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Geological Environment of Some Radioactive Mineral Deposits in South Australia

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The environments of the known uranium occurrences in South Australia are described, and the general relation of uranium mineralisation with sodic granitic rocks is emphasised. All the known large deposits of uranium ore are in rocks of Precambrian age of the Willyama Nucleus, but the time range of uranium mineralisation extends from early Precambrian to Lower Palaeozoic times. The eastern margin of the Adelaide Geosyncline forms a uranium province characterised by the iron-titanium-uranium minerals davidite and thorian brannerite.

Introduction

The vigorous and widespread search for uranium ore deposits initiated in South Australia in 1944, and intensified in later years, has resulted in the discovery of a large number of occurrences of radioactive minerals, some of which have proved to be ore bodies of potential economic value. A natural and important consequence of the search, which engaged the attention of a large proportion of the geological and geophysical staff of the Geological Survey of South Australia during the years 1950 to 1955, was the accumulation of a considerable body of diverse fact and opinion on the nature and environment of radioactive mineralisation in the State. A study of this information soon showed that overseas experience would not be applicable in South Australia, and that in some ways the State forms a rather unique radioactive mineral province.

The geological terranes in which radioactive mineralisation occurs have their counterparts in other parts of the Commonwealth, and it is hoped that the discussion of the South Australian occurrences in this paper may assist in the further search for uranium deposits throughout Australia.

Investigations, commenced in wartime and hampered by extreme secrecy and lack of staff, first concentrated on the known radioactive mineral deposits of Radium Hill and Mt. Painter. With a subsequent, chiefly post-war, expansion of activities, detailed investigation of these and other promising deposits proceeded concurrently with a widespread search of all potentially uraniferous areas of the State. In this search the Department of Mines played a leading part, but much prospecting was done and some important discoveries made by part-time prospectors attracted by the then current "boom" atmosphere and "glamour" of uranium search.

Finds were made in widely separated areas of the State and in rocks ranging in age from early Archaeozoic to presumed Cambro-Ordovician. Most of the finds proved of minor economic interest, and all the larger deposits are confined to crystalline rocks of the basement older than rocks of the Adelaide System. However, the minor deposits are of considerable significance in reaching an understanding of the favourable environment for uranium mineralisation. Of particular interest in this respect were the discoveries of uraninite associated with copper mineralisation in slates belonging to the Adelaide System at Nicol's Nob, in the northern Flinders Range, and of davidite in quartz veins and pegmatites intrusive into meta-sedimentary gneisses of presumed Cambro-Ordovician age some 35 miles east-north-east of Adelaide. These finds more than doubled the area of outcrop of potentially uraniferous rocks.

During the period of most active search large deposits were discovered at Crocker Well, Mt. Victoria and East Painter. In addition to these, small deposits of great interest from the genetic viewpoint were found at Myponga and Houghton in the Adelaide Hills, Nicol's Nob previously mentioned, the Port Lincoln area in the Eyre Peninsula, associated with copper lodes of the Yorke Peninsula, and in outlying parts of the Mt. Painter granitic terrane and surrounding Proterozoic sediments.

It is proposed to describe firstly the regional setting of uranium mineralisation in the State, and secondly to give brief details of the important occurrences considered to be of genetic significance.

Regional geology

As a broad picture of the geology of South Australia is necessary for an intelligent understanding of the geological setting of radioactive mineralisation in the State, an outline is given here.

The western half of the State consists principally of the eastern border of the Western Australian Shield. Although essentially stable since Precambrian times, it has undergone sufficient warping movements to permit the accumulation of thin veneers of Mesozoic and later sediments over a large part of its area. A number of shelf areas have been depressed since Precambrian times, including the Eucla basin, the western part of the Great Australian Artesian Basin (termed the Stuart Stable Shelf by Dickinson and Sprigg, 1953a), and the Maurice Shelf (Dickinson and Sprigg, 1953b) in the uninhabited and mid-western desert area of the State. Thin veneers of sediments of Mesozoic and later age have been deposited in

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these shelf areas. In addition, Quaternary and Recent continental deposits are widespread. The effect of all these is to effectively mask a tremendous area of potentially uraniferous crystalline rocks.

The West Australian Shield is bordered to the south-east by the mobile arc of the Adelaide Geosyncline, containing thousands of feet of sediments in thick sequences. In the Geosyncline the Adelaide System rocks have been subjected intermittently to compressive folding throughout Palaeozoic and later time, but a relatively thin layer of practically horizontal sandstones, deposited in a shelf environment contemporaneously with the geosynclinal sedimentation, extends westwards from the geosyncline along the western shore of Lake Torrens.

Of great importance to the occurrence of uranium are the two Precambrian nuclei east of the Adelaide Geosyncline. One, the Paralana Nucleus at the north-eastern tip of the northerly branch of the geosyncline, is comparatively small. The other is the western half of the Willyama Nucleus, extending eastwards into New South Wales and forming in South Australia the Olary Province of Campana (1957).

The rest of the State is occupied by part of the Great Australian Artesian Basin in the north-east and of the Murray River Artesian Basin in the south-east. Minor areas of crystalline rock outcrop are in the Peake and Denison Ranges, the Houghton and Myponga areas in the Adelaide Hills, and the Palmer area bordering the Adelaide Hills to the east.

Regional environment of uranium mineralisation

Although the large amount of factual information on uranium occurrences in South Australia cannot yet be integrated into a comprehensive theory of genesis and mineralisation control, some important generalisations and interesting speculations can be made.

More than 10 years of exploration has shown that significant uranium mineralisation is confined to rocks of the Archaean, Proterozoic and early Palaeozoic eras. Furthermore, the vast majority of the known deposits are in crystalline basement rocks of pre-Adelaide System age. Uraninite is associated with copper in vein deposits in slates of the Adelaide System at Nicol's Nob, but this is an isolated occurrence and is probably not indicative of widespread uranium mineralisation in the Adelaide System rocks. However, one should be cautious in this assumption, as Adelaide System rocks have not been given the close attention accorded the metamorphosed sediments and migmatites of the crystalline basement.

The apparent absence of sizable deposits and widespread uranium mineralisation in Adelaide System rocks is probably related to the genetic association of uranium minerals with granitic rocks in this State. Early Palaeozoic granitic activity is fairly widespread in the south-east portion of the State but, except for the eastern highlands, the area is masked by younger sediments. Except for two areas, in the vicinity of Palmer and Sanderston, some 35 miles east-north-east of Adelaide, and near Umberatana in the northern Flinders Ranges, this activity appears to have little relation to known uranium deposits. Near Palmer, Cambrian rocks of the Kanmantoo Group forming the eastern border of the Adelaide Hills have been invaded by granites, and uranium minerals have been deposited in quartz veins and pegmatite dykes in the metamorphosed sediments. Near Umberatana, cupolas or bosses of sodic leucogranite intrude calc-silicates and hornfels of Torrensian Series. By inference, the general lack of uranium mineralisation in Adelaide System rocks can then be attributed to the limited intrusion of granite in post-Proterozoic times.

Another observed fact of possible genetic significance is that all the large deposits of uranium so far discovered in South Australia occur in Willyama rocks near the eastern margin of the Adelaide Geosyncline, or in Precambrian terrane east of the geosyncline. Associated with this is the fact that the primary uranium mineralisation at Mt. Victoria, Crocker Well and Radium Hill, and in the Palmer-Sanderston area mentioned above, is davidite and brannerite.

It is suggested here that all the uranium mineralisation in this extensive area is genetically related to granitic activity renewed at widely and irregularly spaced intervals of time extending from the early Precambrian to the early Palaeozoic. The regional geological control is believed to be the fundamental crustal weakness of which the Adelaide Geosyncline is the most spectacular expression. The facts suggest the presence at depth of a uranium-thorium-titanium rich granitic magma or reservoir of granitic fluids which was reactivated at intervals by crustal movement to deposit essentially similar suites of uranium minerals in the crust. The facts also suggest that the centre of most intense activity moved southwards with time, the Palmer and associated granitic intrusions being the final events in a long sequence of discontinuous granitic activity.

A further important generalisation to emerge from the study is the constant association of uranium minerals with sodic granitic rocks thought to have been largely formed by sodic metasomatism of pre-existing sediments. Even the uranium occurrences in the Proterozoic Adelaide System rocks appear to have some connection with soda-rich granitic rocks. Related to this association is the inference that all deposits in which primary uranium minerals have been observed and investigated are of high temperature hydrothermal and epigenetic origin.

Radioactive minerals have been found in small deposits on the Eyre Peninsula in the Precambrian rocks of the Gawler Nucleus and even more sparsely distributed in the crystalline rocks of the Musgrave Nucleus (Dickinson and Sprigg, 1953c). These two Precambrian areas differ from one another and from the nuclei east of the Adelaide Geosyncline. The favourable rocks in the Gawler Nucleus are poorly exposed, and it may be this factor that has prevented the discovery of sizable deposits. How-

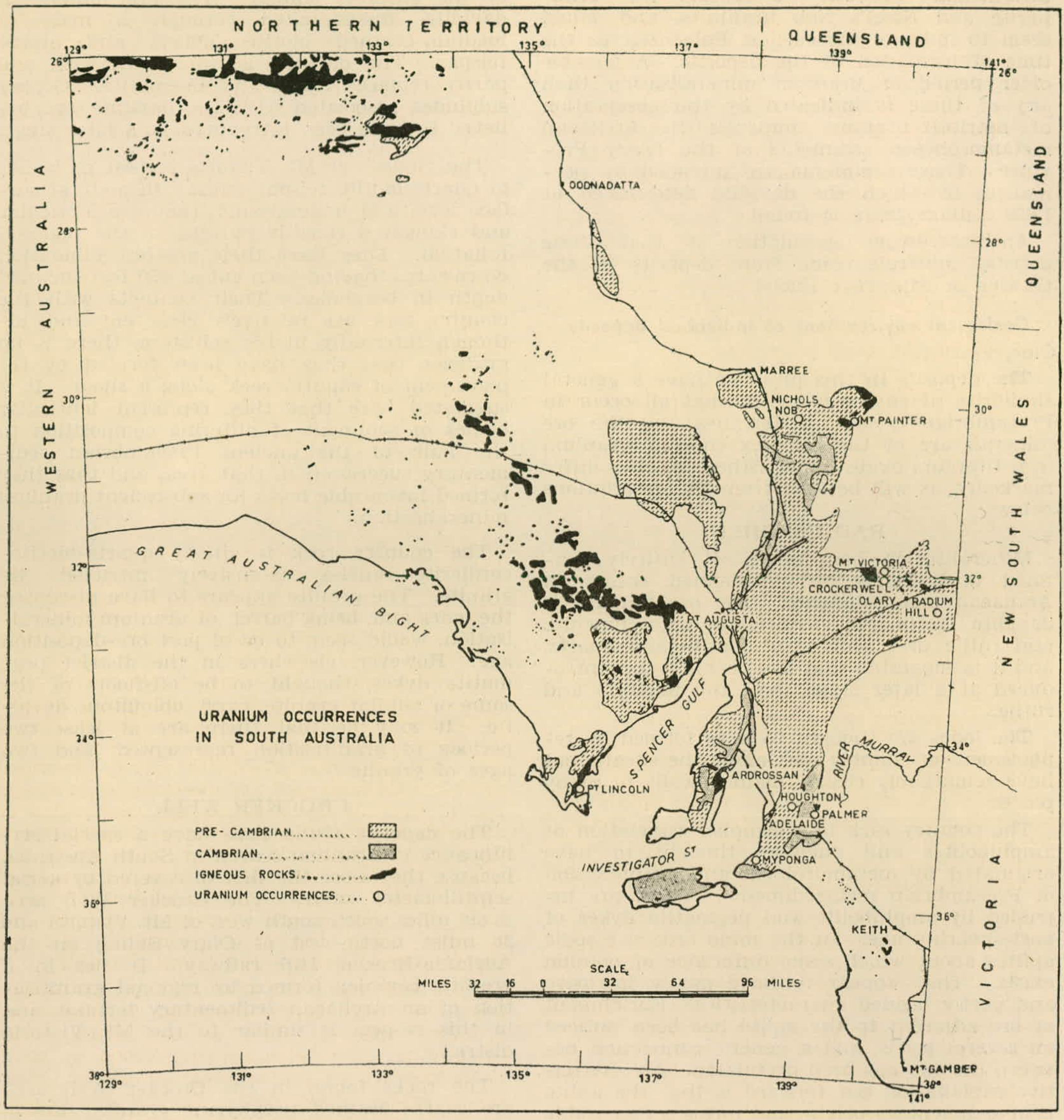

FIGURE 1:—Uranium occurrences in South Australia.

ever, it seems more likely that the environment is generally unfavourable. The age of the gneisses, migmatites, and metamorphosed sediments in which uranium deposits occur on the Eyre Peninsula is not known. It may be that they are of the earliest Precambrian age.

The Musgrave Nucleus is a remote, poorly prospected area of the State, and its relation to the other Precambrian areas of Australia is obscured by superficial deposits. Observed differences are the presence of a complex of ultrabasic rocks, superficially resembling the Sudbury and Bushveld complexes, at the western end, and of masses of charnockitic rocks at the eastern end.

Age determinations, so far confined to minerals from deposits of the Olary Province and the Adelaide Geosyncline, have indicated at least three different epochs of uranium mineralisation. The oldest is that represented by the davidite lodes at Radium Hill and davidite in pegmatites near Crocker Well, estimated to be 1,600-1,700 million years old. There is then a long interval before the mineralisation represented by the brannerite of Crocker Well, determined to have a "best" age estimated at 580 ± 30 million years. The youngest epoch is that of the Nicol's Nob uraninite, 460 ± 10 million years old. The age determined for the pitchblende at Myponga, 510 million years, is intermediate between the Crocker Well brannerite and Nicol's Nob uraninite, and would seem to indicate the earliest Palaeozoic as the time of formation of the deposits. A possible older period of uranium mineralisation than any of these is indicated by the observation of detrital uranium minerals in Archaean metamorphosed sediments of the Olary Province. These sediments are intruded by pegmatites in which the davidite determined at 1,600 million years is found.

An interesting speculation is that these detrital minerals come from deposits in the Gawler or Musgrave nuclei.

Geological environment of individual deposits

Olary Province

The deposits in this province have a general similarity of environment, in that all occur in Precambrian crystalline terranes and the ore minerals are of the complex thorium/uranium iron-titanium oxides type. Otherwise they differ markedly, as will be seen from the descriptions below.

RADIUM HILL

Mineralisation here is almost entirely confined to a series of well-defined shears in Archaean metasediments. The ore mineral is davidite, intimately associated with ilmenite and rutile in a gangue of biotite and quartz, and it is suggested that the uranium was introduced at a later stage than the ilmenite and rutile.

The lodes are thought to have formed by replacement of country rock along the shears, but have remarkably clearly defined walls in many places.

The country rock is a complex association of amphibolites and gneisses, thought to have originated by metamorphism and granitisation of Precambrian metasediments. These are intruded by amphibolite and pegmatite dykes of post-shearing age. In the mine area are sodic aplites about which some difference of opinion exists. They appear to have partly intrusive and partly bedded characteristics. Enrichment of ore adjacent to the aplite has been noticed on several levels, and a genetic connection between the two has been postulated. An alternative explanation put forward is that the aplite is of sedimentary origin, and forms a favourable horizon for ore deposition.

The older amphibolites are thought to be metamorphosed calcareous sediments, and seem particularly favourable host rocks. A thickening in lode and an increase in grade is normal where the lode shears pass through them.

The ore shoots are irregular, and have no well-defined pitch. This is probably due to the fact that the principal ore control is stratigraphic, and mapping has not yet elucidated the detailed fold pattern in the sediments.

MT. VICTORIA

These deposits are analogous to those at Radium Hill, 55 miles to the south-east, in that they occur in a granitised Precambrian terrane, and the mineral composition and paragenesis of the lodes is similar. The ore mineral is davidite, disseminated through a mass of medium-grained biotite, quartz and albitic felspar. The davidite is intergrown with and partly replaces rutile and haematite. Copper sulphides associated with the davidite are believed to have been introduced at a later stage.

The "lodes" at Mt. Victoria consist of bodies of quartz-biotite-felspar schist. In plan, at surface level and underground, they are lenticular and elongated roughly parallel to the regional foliation. They have their greatest dimension downwards, having been cut at 850 feet inclined depth in boreholes. Their contacts with the country rock are relatively clear cut and, although internally highly schistose, there is no evidence that they have been formed by replacement of country rock along a shear. It is suggested here that they represent lenticular bodies of sediment of differing composition to the bulk of the ancient Precambrian sedimentary succession in that area, and that they formed favourable hosts for subsequent uranium mineralisation.

The country rock is chiefly quartz-biotite-cordierite gneiss extensively intruded by granite. The granite appears to have disrupted the lodes and, being barren of uranium mineralisation, would seem to be of post ore-deposition age. However, elsewhere in the district pegmatite dykes, thought to be offshoots of the same or similar granite, carry ubiquitous davidite. It may be that there are at least two periods of granitisation represented, and two ages of granite.

CROCKER WELL

The deposits found here have a special significance in uranium search in South Australia, because they were the first discovered by aerial scintillometer survey. The Crocker Well area is six miles south-south-west of Mt. Victoria and 36 miles north-west of Olary Siding on the Adelaide-Broken Hill railway. It lies in a granitic complex formed by regional granitisation of an Archaean sedimentary terrane, and in this respect is similar to the Mt. Victoria district.

The rocks found in the Crocker Well area are coarse grained porphyritic granite, fine to medium-grained adamellite, alaskite, and aplite in dyke-like and small irregular bodies, granite pegmatites, a granodiorite of sedimentary origin, migmatites, and numerous unassimilated lenses of the highly metamorphosed Archaean sediments.

Although in a general way the porphyritic granite and adamellite form the cores of several migmatitic masses, garlanded by ungranitised sediments, in detail, as might be expected, the relations of the various rock types are far from regular. Thus in the Crocker Well area, which contains the largest adamellite mass, numerous small and large bodies of sediments occur within the borders of the adamellite and conversely, tongues of the adamellite extend into the sediments.

The ore bodies at Crocker Well consist of the complex thorium-uranium-titanium oxide, brannerite*, occurring in a rectangular fracture pattern in the adamellite at Crocker East and in pseudo-breccia at the original prospect. This rock appears to have acted as a favourable host, as brannerite mineralisation is practically confined to it, in spite of a close juxtaposition between the ore bodies in the adamellite and barren metasediments. The ore mineralisation is apparently genetically associated with the intrusion of the aplite and alaskite. These appear as thin veins in the fractures in the adamellite, and partly replacing it adjacent to the fractures. The alaskite, aplite and adamellite are thought to have been produced by sodic metasomatism of pre-existing sediments.,

The age of the brannerite has been determined at 580 ± 30 million years, and as the aplite/alaskite is considered to be a late stage product of the regional granitisation, the age of the latter is regarded as fixed. There are, however, factors, too numerous to be discussed in detail here, which throw some doubt on this conclusion and, as at Mt. Victoria, there may be more than one period of granite formation represented.

Davidite is also present in minor amount in the adamellite, and xenotime and monazite occur in a tabular granodioritic intrusion cutting both the adamellite and alaskite at the original Crocker Well discovery.

Numerous minor deposits occur in the vicinity of Crocker Well, chiefly at the peripheries of granitic masses and frequently associated with pegmatite dykes.

Mount Painter Province

Some of the earliest discoveries of radioactive minerals were made in this province, and when wartime need for uranium became urgent it was the scene of the first intensive investigation of uranium deposits in the State. The principal deposits are those of East Painter, but in recent years a number of minor deposits have been found near the periphery of the Archaean granitic complex in Adelaide System rocks.

EAST PAINTER

The deposits here consist of mineralised masses of a peculiar granitic breccia, similar in some ways to the breccia at the original Crocker Well prospect, except that the rock types present in the breccia are gneiss and schist and a red granite. The country rocks are gneisses and schists of Archaean age, recognisable as metamorphosed sediments. They have been granitised to varying degrees, and two types of massive granite are found. These are a red granite, characterised by a large red felspar and usually stressed and sheared, and a relatively unstressed white granite. The breccia masses are usually associated with the red granite and have features suggesting that some of the breccia fragments have been transported. The breccia is cemented in places by abundant chlorite and haematite.

The most abundant ore mineral is the copper-uranium hydrated phosphate, tobernite. Other secondary minerals recognised are autunite, metatorbernite, uranophane and gummite. The primary source of most of the uranium has not been recognised, the only primary mineral present being fergusonite. Uranium mineralisation is confined to the breccia masses at East Painter, and occurs principally along shears.

MINOR OCCURRENCES IN PROTEROZOIC SEDIMENTS PERIPHERAL TO THE ARCHAEAN COMPLEX

A number of occurrences of primary uranium minerals have been discovered in recent years in the basal beds of the Adelaide system adjacent to the contact with Archaean metasediments in the Mt. Painter Province. The favourable horizon appears to be a series of calc-silicates, representing metamorphosed limestones, near the base of the Torrensian series. These are also the host rocks for most of the copper ore bodies in the district, and there is an apparent genetic association of copper and uranium. Where identified, the primary uranium mineral has proved to be mainly pitchblende, occurring in lodes with a variable gangue, chiefly quartz, in the lodes in the calc-silicate beds.

The source of the uranium and copper has not been definitely determined, but it seems likely that they have their origin in the dying phases of granitic activity associated with the formation of the Adelaide Geosyncline. Granitic masses of boss-like form have been described by Mawson and Dallwitz (1945), intruding calc-silicates of the Torrensian Series close to their contact with the Archaean metasediments at the south-westerly margin of the Mt. Painter massif. In the absence of another source, it is logical to assume that the magma represented by these intrusions gave rise to the copper-uranium ore bodies in the vicinity.

NICOL'S NOB

The chief interest of this deposit is that it was the first discovery of uranium in Adelaide System rocks. Nicol's Nob is 20 miles east of Lyndhurst railway siding on the Pt. Augusta-Alice Springs railway, and some 50 miles west-south-west of Mt. Painter.

Uraninite has been identified from sideritic copper veins occupying narrow fractures in the axial plane of a broad anticlinal fold in Proterozoic slates. The gangue material consists of siderite, calcite and quartz. The age of the uraninite has been determined at 460 ± 10 million years by radio active dating methods. This places it in the early Palaeozoic, and possibly it originated from the same source as the pitchblende in lodes in the Torrensian calc-silicates nearer Mt. Painter.

The Adelaide Hills Province

A number of uranium occurrences, none of which have so far proved of commercial value,

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*When first discovered, the radioactive mineral at Crocker Well was thought to be a new species to which the name "absite" was given. Later study showed it to be a member of the uranium/thorium titanate series, with most of the properties of brannerite. Because of the distinctive appearance of the greenish-brown variety with a vitreous lustre, the name absite is retained for field use.

have been discovered in the Archaean cores of the folded Proterozoic sediments of the Adelaide Geosyncline, and in gneisses and granitic rocks of Cambro-Ordovician age on its eastern margin. Though the host rocks of many of the deposits are Archaean gneissic metasediments, the evidence points to a Palaeozoic age of formation.

HOUGHTON

Fifteen miles north-east from Adelaide, uraniferous minerals occur disseminated in minor fractures in an albite actinolite gneiss thought to have been formed by soda metasomatism of previously metamorphosed sediment. The gneiss is a member of a succession of Archaean rocks, known collectively as the Houghton diorite and comprising a number of varieties or rocks consisting essentially of albitic plagioclase and ferromagnesian minerals.

The ore mineral is so finely divided as to make accurate analysis difficult. It has been tentatively identified as davidite, and is genetically associated with the sodic metasomatism. No obvious structural control, other than a general alignment of radioactivity parallel to the banding of the metasediments, has been observed. The age of the mineralisation is in doubt, because the metasomatism may have taken place at any time between the early Archaeozoic and late Proterozoic, according to the geological evidence.

MYPONGA

Some 40 miles south of Adelaide, pitchblende was discovered in fractures in gneiss forming part of the southernmost Archaean core of the Adelaide Geosyncline.

The country rock consists of highly metamorphosed sediments converted to various types of gneiss, which, in the mineralised area, have been deformed in a simple monoclinal fold. The gneisses have been intruded, and in places altered, by potassic felspar pegmatites.

The ore minerals occur as impregnations of the foliation in a biotite-perthite gneiss at the crest of a minor fold. Rich pockets were found, but the ore did not extend downwards to any great depth. The age of the pitchblende has been estimated by radioactive dating methods at 510 million years.

PALMER-SANDERSTON

The most recent discoveries of radioactive minerals of any significance have been made by a prospector in the gneisses and granite of the Palmer District, 40 miles east-north-east of Adelaide. These rocks are thought to be metamorphosed and granitised sediments of the Kanmantoo Group of Cambro-Ordovician age.

The rather limited amount of detailed investigation to date indicates the most favourable host rock to be diopsidic gneiss and the richest concentrations of davidite to be in thin pegmatite veins transverse to the foliation of the gneiss.

The radioactive minerals are davidite and thorian brannerite, occurring in quartz veins and pegmatite dykes, both parallel and transverse to the regional foliation of the gneiss. The age of the mineralisation, post-Kanmantoo, is fixed on geological grounds, and in view of the fact that davidite younger than the Proterozoic is not known elsewhere in South Australia, should be cautiously regarded until fixed by radioactive dating.

Gawler Nucleus

A considerable number of uranium occurrences have been found on the Eyre Peninsula, and deposits amounting to several thousand tons of low-grade ore have been investigated in the neighbourhood of Port Lincoln.

The deposits near Port Lincoln lie in an Archaean terrane of augen gneiss, granite gneiss, foliated granite and amphibolite formed by granitisation of Precambrian sediments and intruded by sodic pegmatite dykes. The ore mineral consists of minute spheroids of pitchblende surrounded by uranophane-gummite haloes, disseminated interstitially, throughout granulated granitised metasediments. The sediments have been transformed by intense sodic metasomatism, though it is not certain that the uranium was introduced by soda-rich solutions.

The Musgrave Nucleus

This area comprises a large variety of Archaean rock types including adamellites, which elsewhere are favourable hosts for uranium mineralisation. However, it differs from other Archaean provinces in the State in the presence of a suite of ultrabasic rocks and of large masses of charnockitic rocks. Because of its remoteness, prospecting has been sporadic and largely unsystematic, and the noted absence of mineralisation may be due to insufficient search. Uranium minerals present are wholly associated with pegmatites and include allanite, euxenite and cyrtolite.

Yorke Peninsula

Unimportant finds of uranium associated with copper have been made in dumps of old copper mines in the Archaean of the Northern Yorke Peninsula. The association with copper make it likely that this mineralisation is of the post-Proterozoic epoch which introduced copper into Adelaide System rocks.

Future prospects for uranium discovery in South Australia

In this State, as in other parts of Australia, future prospects depend largely on demand, because without it the economic incentive is lacking for recommending expensive prospecting campaigns. However, assuming that increased demand will reactivate uranium search, prospects can be summarised as follows.

The Olary Province offers good prospects. Investigations to date have been intensive only in the Radium Hill and Crocker Well districts. Through considerations of lack of time and personnel, large areas of potentially uraniferous rocks were omitted in the aerial scintillometer survey of the Province; attention being concentrated on the most promising outcrop areas.

Also, apart from the extensive activity in the Radium Hill area, very little systematic ground scintillometer search has been done.

In the Mt. Painter district much of the potentially favourable areas remain virtually unexamined, particularly in view of the relatively recent discovery that basal beds of the Adelaide System are favourable for uranium deposition. The district is considered to have reasonable prospects for the discovery of smaller lode-type ore bodies.

In the Adelaide Hills area the best prospects seem to lie in pegmatitic and vein type ore bodies in the granitic rocks and granitised metasediments of the Palmer area. Prospects for future discovery in the Houghton and Myponga Archaean inliers are poor.

In the remainder of the State prospects are rather enigmatic. In both the Gawler and Musgrave Nuclei large areas of rocks occur of a type which, in the Olary Province, are favourable hosts to uranium mineralisation. It is probable that both regions have been too little explored to dismiss them as unfavourable.

The other areas of potential uranium-bearing rocks are the Yorke Peninsula, offering only very poor prospects, and the extensive outcrops of Upper Proterozoic and Cambrian rocks of the Adelaide Geosyncline and associated shelf sediments.

The upper Proterozoic and Cambrian sediments of the northern Flinders Ranges, the Olary Arc, and the shelf sediments west of Lake Torrens have been practically neglected as far as uranium search is concerned, because the discovery of uranium associated with copper deposits in them coincided with a slackening in demand and a consequent slowing down of prospecting activity. However, the uranium mineralisation associated with the copper so far observed is weak, and prospects for large ore bodies in these rocks cannot be considered good.

The Palmer-Sanderston area has not been sufficiently investigated to give a firm opinion. The davidite and brannerite mineralisation associated with sodic granitic rocks give it obvious similarities with the Olary Province, and it can be regarded as a promising area for future investigations.

It is concluded that on the whole reasonable prospects exist for the discovery of further uranium ore bodies in South Australia.

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(a) p. 429 and Fig. 2.

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Instrumentation Problems in Radioactive Prospecting

By J. Daly and D. F. Urquhart*

The paper gives a brief review of problems in the design of equipment for radioactive measurements required in the search for radioactive minerals. The equipment is used for two main purposes, assaying and prospecting. Equipment required for assaying introduces no special problems. However, the use of radiation measuring equipment in moving vehicles, which is of great value in prospecting, involves fundamental difficulties, apart from any practical problems which may arise. As examples, the design of equipment for bore logging and for airborne surveying is discussed. Bore logging involves serious practical problems, and the response of a bore logger is greatly affected by the time constant of the equipment. In airborne prospecting the practical difficulties are not serious, but the response is influenced by the fact that only radiation from the surface can be detected. A simple method is described for calculating the response of moving detectors.

Introduction

The design of equipment for radioactive measurements is nowadays a fairly well standardised procedure. All that is involved is the selection of a detector capable of giving the information required, and a suitable means of registering this information. The properties of detectors and registering circuits are the subject of a vast literature, in which they are discussed at any desired level of physical and mathematical complexity. A summary at a rather elementary level is given in a publication of the Bureau of Mineral Resources, Geology and Geophysics (Daly, Urquhart and Gibson, 1956); no discussion is given here.

In connection with the search for radioactive minerals, radioactive measuring instruments are required for two purposes, assaying and prospecting.

For the purposes of the present paper the question of instrumentation in connection with assaying may be treated in summary fashion. The measurements required have no unusual features, and the equipment does not differ in design from that used for similar measurements in other fields of nuclear study.

The various methods of assaying may be distinguished as follows:—

(i) Beta or gamma counting, comparing the radiation from the unknown sample with that from a standard. This requires a detector, which may be either a geiger tube or a phosphor and photomultiplier, and counting equipment.

(ii) Simultaneous beta and gamma counting. The equipment is the same as in (i). This method of assaying is fully described by Daly, Urquhart and Gibson (1956).

(iii) Methods involving gamma ray spectroscopy. If a gamma ray spectrum of the radiation from the unknown sample is observed, much more accurate information is obtained on whether the material contains uranium or thorium or both, and its state of radioactive equilibrium. The Bureau has found that there is no advantage in taking a full gamma ray spectrum, but that adequate information is obtained by counting gamma rays of a few selected energies. The standard method now used by the Bureau involves a beta count, taken simultaneously with two gamma counts at selected energies. A scintillation detector must be used for the gamma counts. In addition to the usual counting equipment, pulse amplitude analysers must be provided to select the desired energies. The design of such circuits is standard.

(iv) Delayed Coincidence Methods. These methods have been suggested, but it is not known that they have been used much, presumably because they require a considerable amount of equipment. It is theoretically possible to estimate the amount of any radioactive element which produces a radioactive daughter product of very short half life, by isolating the radiation from each element, and measuring the number of disintegrations of the daughter product which occur within an appropriate time interval after each disintegration of the parent. The equipment required would include gating units, time delays, and coincidence units, in addition to the usual detectors, amplifiers and counting equipment.

(v) Methods involving alpha ray spectroscopy. Such methods are attractive in principle, as both uranium and thorium are alpha emitters. A method of this type has been developed and used in the Bureau, and is described by Howard (1958). The detector is a gridded ionisation chamber, followed by a high gain amplifier and an alpha ray spectrometer. The major drawback to alpha counting methods is the difficulty of preparing the sample in a suitable form.

For prospecting purposes, the information required is a measurement of gamma ray intensity at a chosen point. Consideration of

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* Bureau of Mineral Resources, Melbourne. Manuscript received March 13, 1958.

sensitivity, ease of operation, and maintenance restrict the choice of detectors to Geiger tubes, or scintillation detectors, and of registering circuits to ratemeters. The design and construction of portable Geiger or scintillation ratemeters is standard, and such instruments are now well known. The purpose of the present discussion is to consider the problems involved in the extension of these elementary techniques.

Attempts at the extension of geophysical techniques take the following obvious directions:—

(i) Mounting of equipment in moving vehicles, such as aircraft, thus increasing the speed of coverage.

(ii) Construction of continuously recording equipment, which is a necessary consequence of (i).

With ordinary geophysical techniques, the difficulties involved in such extensions are practical ones only. This does not necessarily imply that the difficulties can be easily overcome. Taking some familiar methods as examples, the airborne magnetometer problem has been solved as regards total force instruments. However, notwithstanding continuous research, airborne magnetometers for the measurement of specific components of the earth's field are still in the early experimental stages. In recent years, airborne electromagnetic equipment has been developed successfully. On the other hand, the difficulties in the construction of an airborne gravity meter have so far proved insuperable, and on present indications will remain so for a long time.

The statement that only practical difficulties are involved implies merely that there is no reason in principle why an instrument cannot be devised which will give fundamentally the same information as the instruments at present used on the ground. With radiometric equipment, this is not the case. Apart from practical difficulties, any extension of radioactive prospecting equipment to moving vehicles involves difficulties in principle, as a result of which the information obtained from the modified method is fundamentally different from that obtained by a static measurement of radioactive intensity. These difficulties are due to the following fundamental causes:—

(i) Unlike gravitational, electrical or magnetic fields, radioactive intensity is not a potential field.

(ii) Any radioactive measurement requires the counting of a random process. This necessarily involves a time constant.

As an example of the application of these principles, instruments for two types of radioactive measurements (for radioactive borelogging and for airborne radiometric prospecting) will be discussed.

Radioactive bore logging

Radiometric logging is used for two main purposes.

(i) Stratigraphic logging, used in oil drilling.

(ii) Logging of holes drilled in the exploration of deposits of radioactive minerals.

The problem in stratigraphic logging is generally to record the position of formations of considerable width, with radioactivity which may be very slight. The main purpose of the work is usually to assist in correlation of geological formations from one drill hole to another. High sensitivity is necessary, but the actual nature of radioactive minerals in a formation, or the precise width of the formation, are not matters of prime interest.

Logging of exploratory holes on the other hand, usually involves relatively strongly radioactive formations which may be quite narrow. High sensitivity is not required, but it is of great importance to obtain an estimate as accurate as possible, of the width and grade of the formations. The discussion here is confined to the second type of logging.

The principles of design of a borelogger will be discussed in some detail, because it may be of interest to observe the non-scientific factors which are often of prime importance. The following practical considerations have considerable influence in the design and construction of a bore logger (in Australia at least):—

(i) Shallow exploratory drill holes are kept to as small a diameter as possible for economic reasons. Practically, this means that the probe of a bore-logger for general use must enter an EX hole, and thus its diameter cannot exceed 1⅜in.

(ii) Drill holes commonly pass through crumbling ground, particularly in the weathered zone. The probe of the logger is liable to stick in such ground. This complicates the mechanical design of the logger considerably.

(iii) There are great advantages in using only components which are readily available. The situation in this regard has improved in recent years, but there is still a wide range of components which can be purchased from stock in U.K. or U.S.A. but which are not available in Australia.

(iv) The designer is practically restricted to the use of a standard type of cable, as cable manufacturers could not consider the construction of a special type of cable for which no large market exists.

There is no problem in the design of the detector or registering circuit. The detector is either a Geiger tube, or a phosphor and photomultiplier. Until recently, photomultiplier tubes of small size were not available, and it was impossible to construct a scintillation probe of diameter less than 2in., so that scintillation loggers could not be used in EX holes. Recently, however, a photomultiplier tube of diameter ¾in. has become available, and scintillation probes can now be made which will enter an EX hole.

The registering circuit is a ratemeter of conventional design. The problem for the designer arises from the fact that detector and registering circuit have to be connected by a cable some hundreds of feet long, which introduces a heavy capacitative load. Theoretically, this can be overcome in several ways, which are all considerably hampered by the practical matters listed above. For example, the probe could include impedance matching elements such as cathode followers or pulse transformers, provided it were big enough. Such a probe, however, would have to contain its own power supplies, otherwise a multiconductor cable of special design would be necessary. Such cables are not readily available.

Also, if more components are built into the probe, it is more expensive, and the chance of loss must be minimised. If batteries are included in the probe, they will have to be replaced periodically. The engineering problem of designing a probe which can be easily dismantled to replace batteries, and which will stand up to the water pressure encountered at depths of some hundreds of feet, is considerably more difficult than would at first appear.

These considerations lead to some form of compromise which usually has obvious drawbacks. Two examples may be of interest. The design of loggers specially constructed for the Bureau of Mineral Resources was based on the following primary considerations:—

(i) The loggers were required for use in EX holes at Rum Jungle. There was reason to expect that the ground would be extremely bad.

(ii) Some of the holes were horizontal holes drilled underground. Push rods were necessary in such holes.

(iii) The only Geiger tubes available were glass tubes about ¾in. diameter. As the probe was restricted to 1⅜in. diameter, there was no room in it for other components.

Based on these requirements, equipment was designed in which the push rods were an integral part of the equipment, to be used in all holes. This has the following advantages:—

(i) The push rods provide all the strength necessary, so that a low cost, low capacity cable of low mechanical strength could be used.

(ii) The cable selected was a Pt 11 M coaxial. Because the push rods could be used as an electrical connection, the equivalent of a three conductor cable of very low capacity was available. This was made the basis of an ingenious device for reducing the effect of the cable capacity still further.

The use of the push rods means that the probe must be raised and lowered by hand. The method of reading is to halt the probe at each reading point for a time sufficient for the ratemeter to reach a steady reading. Distances are measured along the push rods, and no errors due to cable stretch are introduced. The probe is relatively inexpensive. However, continuous recording is not easily practicable.

This equipment has been used for several years, and is still in use. Its only serious drawback is that it is slow and cumbersome.

An example of an alternative design is the "Deedle bug" designed by the U.S. Bureau of Mines. The Geiger tube used on this equipment is metal-walled, and is about ¾in. diameter. It requires no protection, and itself forms the probe. The tube is of the high current type, and measurement involves recording the steady current flowing so that the capacity of the cable does not affect the design.

Because the diameter of the probe is considerably smaller than that of the smallest hole, it is not so liable to stick, and the cable provides strength sufficient for retracting the probe. The probe is driven by a winch, which also controls the paper drive of a pen recorder. Continuous recording is therefore possible.

Provided the hole is reasonably clear so that free motion of the probe is not hindered, this type of logger is convenient in use. Its disadvantage is the effect of the time constant on the response. The response may be calculated by a very simple process.

The action of a logger involves feeding a current proportional to the count rate at any instant into a resistance-capacity circuit, and measuring the voltage developed. If R and C are resistance and capacity, and I(t) the current input, the voltage developed is given by:—

$$\frac{Cdv}{dt} + \frac{v}{R} = I(t)$$

or $$\frac{RCdv}{dt} + v = R.I(t)$$

RC is the time constant of the circuit.

If an analytical expression for I(t) may be written down, the equation can be solved for V immediately. It can also be solved very

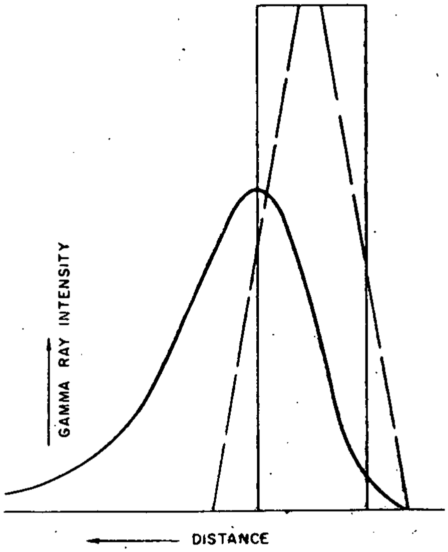

FIGURE 1:—Response of continuously recording borelogging.

conveniently by graphical methods, as discussed, for example, by Bailey and Somerville (1938).

The application of this to the logging problem is shown in Figure 1.

The ideal log which should be obtained when passing a band of uniform radioactivity is shown by the rectangular graph. Due to the finite length of the probe, the actual current input to the ratemeter will have the trapezoidal wave form shown. Graphical integration over the time constant of the equipment gives the response as shown by the curved line, which differs very considerably from the ideal log.

A priori, it would appear doubtful that such a simple theory could provide a realistic basis for the discussion of bore logging problems. However, the theory has been tested by a set of experiments performed by Territory Enterprises Ltd., at Rum Jungle, the results of which were made available to the Bureau of Mineral Resources. The results indicate that the logs calculated by this method fit observations with surprising accuracy, and that this simple treatment can be used with confidence to predict the response of loggers to various conditions.

One significant conclusion from this work is that unless the radioactive formation is considerably wider than the length of the probe, the shape of the log has only an indirect relation to the width and grade of the formation, and it is quite impossible to obtain accurate information on the width and grade of narrow formations from logs of this type. A detailed report on this work will be issued in a Bureau publication.

Airborne radiometric prospecting

An obvious method of increasing speed of coverage in prospecting is to use equipment mounted in a motor vehicle or an aircraft. Continuous recording is necessary. Equipment mounted in a motor vehicle has been used successfully by the Bureau of Mineral Resources, but it has been found that the efficiency of this method is limited by topography to a much greater degree than might be expected. The use of aircraft offers greater possibilities. There is no particular problem in the design of equipment for this purpose, nor is the time constant of the equipment an important factor, to a first approximation. However, other problems arise, which are due in principle to the fact that radiation intensity is not a potential field.

The ideal method of prospecting would fulfil the following two requirements:—

(i) It would register radiation coming from any ore body.

(ii) It would register only radiation coming from ore bodies.

Neither of these conditions can be satisfied by any method, because it is possible to record only radiation arising from surface material. Unless an ore body actually outcrops, it may have little or no expression in surface radioactivity. On the other hand, any prospecting instrument will register all surface radioactivity, from whatever source.

Prospecting with a hand instrument involves testing the level of radioactivity at various points, and tracing the source of any anomalous radioactivity (not usually a very easy matter). An improvement in speed of coverage can be obtained by mounting the detecting equipment in a light aircraft. This is flown over the terrain at the lowest possible altitude. The site of any anomalous radioactivity is marked on air photos and the marked sites are later examined on the ground. In principle, this amounts to performing the operation of ground prospecting at the speed of a light aircraft. For the purposes of a mining company wishing to prospect a limited area, this is a very satisfactory method and has been widely and successfully used.

However, it has difficulties where a full routine coverage is required, particularly where information has to be published. The information must be published in some form of map. An inaccurate map is of no value, and an accurate map cannot be prepared from air photos alone, but requires additional control by other surveying methods. The most economical solution is to fly the area, locating the position of the aircraft, not on air photos, but by means of some other surveying method, such as Shoran. An accurate map is prepared separately by the usual methods, and the survey results combined with it.

This involves carrying the necessary positioning equipment in the aircraft. For this reason, a large aircraft is necessary which must fly at greater heights and speeds than a light one. The disadvantage of the greater height may be minimised by mounting the detector in a "bird," which is trailed below the aircraft in flight. This method has been used successfully by the Bureau, although it introduces difficulties in actual flying which may become serious in areas of rough topography. The extra height and speed mean that the response of the detector averages over a large area, and is affected to some extent by the time constant of the equipment. It is therefore one stage more difficult to relate it directly to conditions on the ground.

A comparison with aeromagnetic methods will help to make the matter clearer. The magnetic field on the ground consists of the effects due to magnetic bodies of geological significance, at various depths, with in many instances a superimposed magnetic "hash" due to detrital material of irregular magnetism, which may obscure the more significant indications completely. In an airborne survey over the same ground the effect of the "hash" disappears, because it is caused by near surface material having random polarity in either sense, and its integrated effect at a distance is zero. The radioactive case is quite different.

Radioactive intensity cannot have a negative value, so that irregularly distributed surface radioactivity will certainly have a positive integrated effect at a distance. Instead of simplify-ing the picture as in the aeromagnetic case, an airborne radiometric survey is likely to confuse it unless the results are examined with special care.

The effect of time constant on the response can be evaluated by the same theory used for bore logging. However, the problem is rather more difficult, because allowance must be made for the absorption of radiation in the air. The equation:—

$$\frac{RCdV}{dt} + V = R. I(t)$$

applies as before, but the expression for I(t) is more complicated.

The effect on the detector of an elementary area of radioactive material at a distance r is proportional to $\frac{e^{-\mu r}}{r^2}$ where μ is the absorption factor for gamma radiation in air. The value of I(t) at any instant is therefore proportional to $\frac{e^{-\mu r}}{r^2}$ dr, where the integral is taken over the total area of radioactive material observed by the detector at the instant in question. This integration may be performed graphically. Work is in progress to test the application of this theory to actual prospecting problems.

Conclusion

The above brief review indicates that the usual principles of the design of radioactive measuring equipment are quite adequate to any of the requirements of radioactive prospecting. There are plenty of difficulties involved in the discovery and exploration of deposits of radioactive minerals, but there is no reason to expect that their solution will be made any easier by improvements in instrumental design.

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Some Oxide Ceramics as Reactor Materials

By K. D. Reeve*

Although the conventional advantages of ceramics lie in their favourable high temperature properties, reactor technology has not yet made use of these properties, mainly because of their uncertain behaviour under irradiation. The fabrication and properties of beryllia, uranium dioxide and thoria, and of mixed oxide systems, are discussed.

Introduction

Ceramics, principally carbides and oxides, are currently being considered for use in nuclear reactors as:

(i) Moderators. Graphite is a ceramic moderator. Beryllia is a better moderator, not sensitive to oxidation at high temperatures, which is an advantage over graphite.

(ii) Fuel-Element Material. The use of ceramic fuels is projected for high temperature systems for which metallic fuels are not available, or for other systems where ceramics are desirable replacements for metals, usually because of chemical inertness and structural stability.

(iii) Future uses may include canning material and control rod material.

Oxides have the advantage over carbides of not being sensitive to oxidation at elevated temperatures; coolant compatability problems are therefore simplified.

The oxides considered here are principally beryllia, uranium dioxide, and thoria. Some discussion is included on wholly ceramic dispersion type fuel element systems.

Beryllia

Introduction

Beryllia is an attractive possibility as a moderator on nuclear grounds, and because of its refractory nature and chemical inertness, has special advantages for high temperature systems. The resistance to attack of dense material by liquid metals at moderate temperatures is also a desirable property for application in liquid metal-cooled or liquid metal-fuelled reactors.

In 1944, a nuclear power reactor containing pebbles of beryllia and uranium dioxide in a pebble-bed cooled by molten bismuth was proposed (Daniels 1957). A modified proposal for a high temperature, helium-cooled, beryllia-moderated power reactor to be built at Oak Ridge was accepted, and a research programme started (Daniels 1957; Howe 1956). The design consisted of beryllia tubes impregnated with uranium dioxide and suspended inside beryllia moderator bricks. The project was later abandoned because of insufficient experience in the fabrication of complicated ceramic bodies, and in the engineering use of ceramics. Present reactor uses are restricted to low temperature applications as reflector (David 1957) in the U.S.A.E.C. bulk shielding facility (swimming pool), the Low Power Water Boiler (LOPO) and the High Power Water Boiler (HYPO) and as moderator (McCreight and Sowman 1956), in the Los Alamos, Homogeneous Aqueous Reactor (HAR) and in the Oak Ridge Aircraft Reactor Experiment (ARE) (Bettis et al. 1957). Although in the latter case, the moderator is cooled by liquid sodium, the sodium is enclosed in stainless steel tubing and does not come into contact with the beryllia. These applications are based on the nuclear properties of beryllia; advantage has not yet been made of its favourable high temperature properties.

The available data on the physical, chemical and mechanical properties, and on the fabrication and uses of beryllia, have been reviewed recently by White (1955).

Physico-chemical properties

Beryllia crystallises with a slightly distorted hexagonal wurtzite structure (Jeffrey et al. 1956). No phase changes have been observed up to the melting point of 2,570°C. The theoretical density is 3,025 gm. cm.$^{-3}$. Thermodynamically, beryllia is a very stable oxide, with a free energy of formation of —140,000 cal. per mole. at 0°C.

Fabrication

The techniques of crucible production by slip-casting and sintering high-fired (1,700°C) beryllia are fairly well established (Norton 1947; Murray et al. 1954a). The latter workers describe the Harwell procedure in which beryllia containing 0.8 to 2 per cent. impurities was sintered at 1,700-1,800°C after slip-casting, resulting in zero porosity, translucent crucibles. During this study it was observed that the purer material sintered less readily. The practice of hot-pressing beryllia and other refractory oxides as developed at Harwell has also been described (Murray et al. 1954a & b; Murray 1954). Beryllia can be hot-pressed in graphite dies at 1,600-1,800°C and 2,000lb./sq. in. to 98 per cent. theoretical density.

The rigid purity requirements for nuclear reactor uses have directed attention to the fabrication of high purity beryllia, in which near theoretical density is much more difficult to attain. The fabrication by cold-pressing and sintering and by hot-pressing of a high purity beryllia has been studied at Harwell. (Livey et al. to be published). Small additions of calcium oxide and magnesium aid densification, but pure beryllia does not sinter to high density, irrespective of surface activity of the original powder.

The status of beryllia production technology

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in the U.S.A., France and the U.S.S.R. was summarised in papers presented to the 1955 Geneva Conference (Kaufman and Kjellgren; Caillat and Pointud; Meyerson respectively, 1956).

In the U.S.A., a pure grade of Brush beryllia is calcined to 1,150° and hot-pressed in graphite dies at 1,600-2,300° at pressures of 200-500lb./sq. in., producing articles of near theoretical density. Alternately, beryllia is fused, crushed, ground and classified, before cold-pressing, slip-casting or extrusion. Densities achieved on sintering this material range from 2.0 to 2.6 gm. cm.$^{-3}$ (67-87 per cent. theoretical).

In France, a grade of oxide containing less than 0.04 per cent. impurities is used for briquette production by hot-pressing. Beryllium hydroxide is precipitated and calcined under very accurately controlled conditions, then hot-pressed in graphite dies at temperatures up to 1,900°C, and pressures of 2,000-2,500lb./sq. in. Briquettes of rectangular shape 50 by 100 by 100 mm. of mean density exceeding 2.90gm. cm.$^{-3}$ have been produced by this method.

In the U.S.S.R., a pure grade (less than 0.03 per cent. impurities) is fabricated by one of three methods. Tamping of a pre-calcined material (1,200-1,800°C) in graphite dies and sintering at 1,800° produces articles of density 2.1-2.2 gm. cm.$^{-3}$. Hot-pressing in graphite dies at 1,800°C and a pressure of 200-300lb./sq. in. produces bodies of density up to 2.75 gm. cm.$^{-3}$. Hot-pressing under vacuum at 1,850-1,900° and 30-300lb./sq. in. produces bodies of density 2.9 gm. cm.$^{-3}$, which are exceptionally pure and strong, although this is obviously an uneconomic production method.

In general, near theoretical density in highly pure beryllia can be produced readily only by hot-pressing, although for certain applications addition of oxides which assist sintering after cold-forming may be allowable. As hot-pressing is not at present an economical process for large scale production, considerable effort is now being directed to methods of attainment of comparable densities by cold-forming and sintering techniques without resort to additives.

Quirk et al. (1957) have studied the effect of calcination temperature of four grades of beryllia on the density attainable on hydrostatically pressing at 100,000lb./sq. in. and sintering in hydrogen at 1,450°. Calcination at 800° resulted in better than 95 per cent. density for beryllia containing 0.15, 0.25 and 1.0 per cent. impurities, although the purest sample (0.03 per cent. impurities) could be sintered only to 75 per cent. density. Higher calcination temperatures, particularly above 1,200°, reduced attainable densities in each case. The basis of this method of achieving high density after cold-pressing and sintering at such low-temperature is in the highly surface-active nature of the low-temperature calcined material. Specific surface areas for 800°C calcined material were 30-80 m^2/g.; for 1,200° calcination, only 2-20 m^2/g. Variations with calcination temperature of the lattice parameter, which passes through a minimum at 1,000°, suggested that strain due to impurities might be important. The reason for the poor sintering behaviour of the purest product is not fully understood. Addition of 1 per cent. magnesia before calcination (to 800°) followed by sintering to 1,450° resulted in better than 90 per cent. theoretical density for all four powders. The high shrinkage on sintering of compacts pressed from very fine surface active powders places a serious size limitation on production of dense bodies by this method.

In the course of a study on the sintering properties of fine magnesia powders, preliminary results on the densification of surface active beryllia powders at temperatures as low as 800°C were obtained (Livey et. al. 1957). Beryllia powder prepared by calcination of beryllium hydroxide at 700° was heated to 800° in a Stellite die under a pressure of 10 tons/sq. in. Although the compact was cracked, the density was 90 per cent. theoretical, and the method is promising.

Thermal properties

Beryllia is outstanding amongst ceramic oxides in its excellent thermal shock resistance; the main contributing factor to this behaviour is the exceptionally high thermal conductivity. Recent determinations of thermal conductivity on slightly porous samples indicate a strong dependence on the method of fabrication, probably due to differences in pore size and orientation. This effect has been pointed out by Murray (1954), while comparing the conductivities of hot-pressed and slip-cast-sintered beryllia of the same porosities. The hot-pressed samples had a higher conductivity in each case. The thermal shock index $\left(\frac{k_c}{E}\right)$ was determined as a function of temperature for hot-pressed beryllia of 2 per cent. porosity and for slip-cast-sintered beryllia of 13 per cent. porosity. The index fell off sharply with temperature up to 1,000°, but was always approximately twice as high for the hot-pressed as for the sintered specimens.

Two recent determinations of thermal conductivity are summarised in Table 1. Those obtained at MIT (Kingery et al. 1954) were on slip cast and sintered beryllia of density 2.7-2.86g. cm.$^{-3}$, and those at NBS (Ditmars and Ginnings, 1957) on similarly prepared material of density of 2.6 g. cm.$^{-3}$. Both sets are uncorrected for porosity.

TABLE 1.—THERMAL CONDUCTIVITY OF BERYLLIA

T (°C)	k (cal. sec.$^{-1}$ °C^{-1} cm.$^{-2}$ cm.) MIT	NBS
100	.50	.38
200	.40	.27
300	.30	.20
400	.21	.16
500	.15	.13
600	.11	.11
700	.08	.097
750	.07	.092

2

The lack of agreement between these results may be due to differences in density, in starting material and method of fabrication, and in view of the marked difference in the rates of change of conductivity with temperature, probably in part due to inherent inaccuracies of one or both methods.

Mechanical and high temperature properties

The bend strength of hot-pressed beryllia at room temperature is usually in the range 20,000 to 30,000lb./sq. in. (White 1955), provided that excessive grain growth has not occurred. Loss of strength due to large grain growth in beryllia and other ceramics is a well-known phenomenon. Bend strengths of up to 40,000lb./sq. in. were noted for the dense beryllia prepared from active powder, for average crystal size in the sintered material of less than 20 microns (Quirk et al. 1957). The strength was observed to fall off sharply with increase in crystal size, being only 500lb./sq. in. at 200μ microns. The importance of avoiding excessive grain growth, which occurs during high temperature sintering, but is not essential to it, is thus established. Sintering temperatures should be kept as low as possible.

The Russian work (Meyerson 1956) refers to exceptionally strong beryllia produced by vacuum hot-pressing, although no figures are quoted. There is little doubt, however, that there is much scope for improvement in the strength of beryllia by controlling the fabrication process.

It is generally agreed that the mechanical properties of beryllia assume lower values with rise in temperature. Figures summarised by White (1955) indicate an appreciable fall in bend strength, compressive strength and elastic modulus from room temperature to 1,000°C, although unpublished Harwell results (Scott) on hot-pressed material indicate no loss in strength at 1,000°C. Baroody et al. (1951) report that plastic flow occurs in beryllia at 1,000°C. Table 2 shows reported creep rates for beryllia of 30-35 per cent. porosity under a stress of 95lb./sq. in. (Sinott 1949).

TABLE 2.—CREEP OF BERYLLIA

Temperature °C	Creep Rate Per cent. hr. x 10^4
1095	3.1
1150	7.0
1205	14.6
1260	100

Brittan and Sibbitt (1947) give some results on the high temperature behaviour of hot-pressed beryllia. At Battelle, a bend strength of 15,000lb./sq.in. was measured at 1,370°C, and no deformation was observed at 1,630° for several hours under a load of 170lb./sq. in. "Norton" hot-pressed beryllia showed only a slight tensile strength decrease up to 1,460°. Slight expansions on reheating were noted. The high temperature behaviour of beryllia is undoubtedly sensitive to method of fabrication and extent and distribution of porosity; dense hot-pressed material probably creeps negligibly below 1,200°C, although more detailed work is needed.

The vapour pressure of beryllia is given as 3.6 x 10^{-4}mm. at 2,000°, and 2.4 x 10^{-3}mm. at 2,140° (Brittan and Sibbitt 1947). A more important limitation on the use of beryllia at elevated temperatures is in the reaction with water vapour, which becomes appreciable above 1,300°. (Livey and Murray 1956). Weight losses of 0.5-0.7 per cent. after 2½ hours at 1,400°C in an atmosphere containing water vapour at 90 mm. mercury pressure were quoted by Brittan and Sibbitt (1947).

Permeability to gases is an impotant property for possible reactor application. Low density products have considerable open porosity, and hence, are quite permeable. Open porosity has been observed to persist to quite low total porosities (Murray et al. 1954). However, it is reported that 96 per cent. density beryllia appears to be "impermeable" in ½in. thicknesses at room temperature, and that Degussa slip-cast beryllia is "gas-tight" at 1,000°C (Brittan and Sibbitt 1947). No quantitative data has been published.

Resistance to liquid metals

Beryllia is reported to be attacked by liquid sodium-potassium at 500°-600°C unless the specimen is very dense and highly "vitrified." (Kelman et al. 1950). Although no reliable figures are available, pure hot-pressed beryllia is probably unattacked by oxygen-free sodium or sodium-potassium at these temperatures. Oxygen in the liquid metal may be a major cause of beryllia attack.

Irradiation behaviour

Present results on the behaviour of BeO under irradiation are incomplete. Irradiation to 10^{19} n.v.t. has been observed to result in serious decreases in compressive strength, a 40 per cent. loss in thermal conductivity, and up to 1 per cent. expansion (Billington 1956). Preliminary British results (Bacon and Wilson 1955) showed that irradiation to 7 x 10^{20} n.v.t. caused lattice expansions of 0.088 per cent. in the c-axis and 0.033 per cent. in the a-axis. This change could be annealed out to the extent of 20 per cent. after eight hours at 500°, and 95 per cent. at 1,100°C. Very much more work is needed, particularly at neutron energies and irradiation temperatures where helium production due to (n, α) and (n, 2n) reactions may be of importance.

Summary

The present situation on the possibilities of the use of beryllia as moderator in high temperature reactor systems can be summarised as follows:—

- (i) Beryllia is an efficient moderator from the reactor physics point of view.
- (ii) It is compatible with coolant gases to high temperatures, provided water vapour is excluded.
- (iii) There are no phase changes up to 2,570°C. There is probably little loss of strength up to 1,000°C or higher.

(iv) In common with most other ceramics, beryllia is brittle, which is an undesirable engineering property.

(v) The thermal shock resistance, although high compared with most other ceramics, may still not be adequate for parts subject to a high heat flux, or to high rates of heating and cooling.

(vi) Reactor design with points (iv) and (v) in mind can minimise these unfavourable properties.

(vii) Fabrication of dense material is at present carried out by hot-pressing, an expensive method for large-scale production. It may be possible in the future to use cold-forming and sintering methods.

(viii) Fabrication of complicated shapes in large sizes is difficult.

(ix) The behaviour under irradiation is not fully established. In particular, the possible serious decreases in thermal conductivity would lower thermal shock resistance.

Uranium dioxide and thoria

Introduction

The three oxides of most interest as fuel or fertile material are uranium dioxide, plutonium dioxide, and thoria. Little unclassified information is available on plutonium dioxide (see Drummond and Welch 1957).

The uses of uranium dioxide as fuel element material may be described as follows:—

(i) Use in high pressure water systems, based on the excellent corrosion resistance of uranium dioxide.

(ii) Use as replacement for uranium metal fuel in gas-cooled reactors, allowing the temperature to be raised above that of the α-β phase change (660°C). Inertness towards possible metallic canning materials compared with uranium is also an advantage here.

(iii) Use in cermet dispersion-type fuel elements (Howe 1956; Weber and Hirsch 1956); the uranium dioxide may be simply a convenient form in which to disperse the uranium (e.g., Ae-UO_2, Geneva reactor) or temperature conditions may be such that only the oxide is compatible with suitable matrix metals.

(iv) Possible use in ceramic dispersion type fuel elements for high temperature reactors.

Uses of thoria may be described as follows:—

(i) Use of thoria in suspension (aqueous, liquid metal), as powder, or in massive form as a convenient and inert form of thorium for breeding by the TH-U233 cycle.

(ii) Use as "inbuilt" breeder in UO_2-ThO_2 fuel elements.

(iii) Possible use in a ceramic dispersion type fuel element, with fissile and fertile material both dispersed in the matrix.

Fabrication

The technology of uranium dioxide and thoria has been reviewed recently (Johnson and Curtis 1956; Murray and Livey 1956). Both are typical ceramics, and can be fabricated by the usual ceramic techniques.

URANIUM DIOXIDE

This oxidises readily on heating in air to give successfully non-stoichiometric oxides $UO_2 + x$ and U_3O_8 (with destructive increase in volume); U_3O_8 decomposes above 1,200°C to UO_2, containing a slight excess of oxygen. U_3O_8 formation must be avoided, and consequently UO_2 must be protected from oxidation during the sintering process and in use, except at temperatures above 1,200-1,300°C.

Stoichiometric UO_2 is difficult to sinter to high density in argon, but non-stoichiometric oxide sinters much more readily. The favourable sintering properties of British non-stoichiometric oxide ($UO_{2{\cdot}13}$) are thought to be due partly to the fine particle size and high specific surface of the material, and partly to the oxygen excess (Williams 1957). Conditions of preparation of UO_2 powder to achieve optimum sintering properties are very critical. Both stoichiometric and non-stoichiometric material can be hot-pressed to high density, but the latter is not suitable for the hot-pressing of large sizes, because reduction to UO_2 occurs in the outer layers adjacent to the graphite die, differential sintering rates are set up, and cracking occurs (Murray and Livey 1956).

Non-stoichiometric oxide is known to be appreciably more volatile at elevated temperatures. Results discussed recently by Williams (1957) indicate that stoichiometric oxide, although more difficult to fabricate, may be preferable for reactor use because volatility and structural changes at high temperatures are minimised.

THORIA

Thoria shows no non-stoichiometry, and is generally more difficult to sinter to high density than non-stoichiometric uranium dioxide. However, starting from very finely ground, pure thoria (99.9 per cent.), zero-porosity translucent crucibles have been prepared at Harwell by slip-casting and sintering at 1,825°C (Murray et al. 1953).

Striking improvements in sintering behaviour have been obtained by addition of ½ w/o CaO before sintering at 1,700°C. The density attainable was improved from 86 per cent. to 96 per cent. of the theoretical by this means (Johnson and Curtis 1954). Effort has recently been directed to obtaining this effect by the use of other additives which on nuclear grounds may be less undesirable. Calcium fluoride, strontium oxide, and vanadium pentoxide have similar, but smaller effects, than calcium oxide. Addition of beryllia is only very slightly beneficial; uranium dioxide retards sintering (Curtis and Johnson 1957; Arenberg et al. 1957).

Some properties of uranium dioxide and thoria are presented in Table 3, with corresponding properties of beryllia included for comparison. Figures quoted on fabricated material are representative ones for material of at least 90 per cent. theoretical density.

TABLE 3.—PROPERTIES OF BeO, UO_2 and ThO_2 (a, b, c).

	BeO	UO_2	ThO_2
Crystal structure	Wurtzite (hexagonal)	Fluorite (cubic)	Fluorite (cubic)
X-ray density (g.cm^{-3})	3.025	10.95	10.15
Melting point (°C)	2570	2800	3200
Colour	White	Brown-red	White
Thermal conductivity (cal.sec^{-1} cm^{-2}cm °C^{-1}			
20°C	0.5-0.6	0.02	0.02
1000°C	0.05	0.008	0.007
Modulus of elasticity (lb./sq. in.) 20°C	50×10^6	25×10^6	21×10^6
Coefficient of thermal expansion (°C^{-1})			
0-1000°C	9.3×10^{-6}	10×10^{-6}	9×10^{-6}
Bend strength 20°C (lb./sq. in.)	20,000	16,000	12,000

(a) White, 1955; (b) Johnson, et al, 1957; (c) Livey, 1957.

Discussion

One of the most serious disadvantages of uranium dioxide as a fissile material, which is necessarily subject to high heat fluxes, is its low thermal conductivity and hence sensitivity to thermally induced stresses. The advantages of dispersion in a high conductivity material (metallic or beryllia), particularly for highly rated fuel elements, are thus apparent. However, massive uranium dioxide as fuel is being intensely investigated in Canada, U.S.A. and the U.K., and it seems likely that the low thermal conductivity will not prove as big a disadvantage as might be supposed.

Thoria, as fertile material, may not be subject to such severe thermal stresses as fissile uranium dioxide but the low thermal conductivity may be a disadvantage here also. Chemical processing of sintered thoria is difficult, and it may be advantageous from this point of view to use cold-consolidated powder, which could be processed more readily. However, heat production during breeding, combined with low thermal conductivity, may result in high centre temperatures and sintering. The advantages of avoiding sintering in thoria for future chemical processing are shown in the time required for nitric/hydrofluoric acid dissolution of thoria calcined to various temperatures (Curtis and Johnson, 1957), given in Table 4.

TABLE 4.—DISSOLUTION OF THORIA.

Calcination Temperature (°C)	Dissolution Time (hours)
700	1½
1,200	2½
1,630	5½

Ceramic dispersions

Introduction

If reactor requirements are such that a ceramic oxide fuel is desirable, there are two major alternatives:—

(i) Use of UO_2 fuel, natural or slightly enriched, with its attendant disadvantage of low thermal conductivity and probable cracking during use or

(ii) use of a dispersion type fuel, in which enriched UO_2 is dispersed in a ceramic matrix.

The matrix should have a higher thermal conductivity than uranium dioxide, should be capable of fabrication to low porosity, should be reasonably stable under neutron irradiation, and should not react with uranium dioxide under fabrication or operating conditions.

Some possible systems will now be considered.

Uranium dioxide-thoria

The fabrication of prototype Borax IV fuel elements from this material has been described previously. (Handwerk and Noland, 1957). This is not a true dispersion type system, because uranium dioxide and thoria form a complete series of solid solutions (Lambertson, et. al. 1953). The equilibrium state of uranium dioxide-thoria bodies is thus a uniform solid solution, which forms readily during fabrication. There is no improvement in thermal conductivity over uranium dioxide; moreover, these mixtures do not sinter as readily as either thoria or non-stoichiometric uranium dioxide.

However, the system has some advantages, viz.:—

(i) UO_2-ThO_2 can be fabricated by the air-sintering of U_3O_8-ThO_2 mixtures; air-sintering is not possible with UO_2.

(ii) The ThO_2 is present as an "in-built" breeding material.

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(iii) UO$_2$-ThO$_2$ bodies are stable in air; excess oxygen can be accommodated interstitially in the solid solution fluorite lattice. For less than 50 w/o UO$_2$ in the solid solution, no additional phases have been observed on oxidation (Anderson, et al. 1954).

Beryllia-thoria

This system has the advantage of a high conductivity matrix material; no solid solutions or compounds are known, and thus a dispersion of uranium dioxide in a beryllia matrix should be stable if liquid phase formation is avoided. A eutectic in this system has been observed at 2,150° (Lang, et al. 1956). A liquid phase will begin to form at this temperature, setting the upper limit for use of beryllia-uranium dioxide bodies at somewhat less than 2,150°.

Extensive work was carried out on beryllia-uranium dioxide dispersions at Battelle, Oak Ridge and Argonne as part of the Daniels Pile Project (Howe, 1956). Brittan and Sibbitt (1947), describe the fabrication of 2 w/o and 10 w/o dispersions of uranium dioxide in beryllia. These were made by hot-pressing a mixture of fine beryllia and U$_3$O$_8$ powders at 1,800°C, and 95 per cent. theoretical density could be achieved this way. The U$_3$O$_8$ breaks down to uranium dioxide which is present in the compact as a fine micron-size dispersion. The thermal conductivity, melting-point and bend-strength were reported to be the same as for hot-pressed beryllia.

These bodies were unstable in air, and crumbled to powder at 700° due to U$_3$O$_8$ formation. Above 1,000°C volatilisation of oxidised specimens occurred. Impregnation of porous beryllia with UO$_3$ and subsequent decomposition is also described.

Preliminary results on the behaviour of beryllia-uranium dioxide bodies under irradiation have been reported (Billington, 1956). After one in 2 x 10^5 uranium atoms had fissioned the thermal conductivity of 2 w/o and 10 w/o uranium dioxide in beryllia had dropped by a factor of six without signs of saturation. The linear dimensions increased by one per cent. and the compressive strength and elastic modulus decreased by 30 per cent. However, severe damage from fission product recoil atoms to the beryllia matrix would be expected in these fine (less than 2 microns) dispersions of UO$_2$.

Recoil atom escape and also the volume per cent. damage to the matrix in such systems generally can be decreased by increasing the particle size of the dispersion to 50-100 microns. (Weber and Hirsch, 1956). Methods of producing uranium dioxide grains of such sizes have been summarised by Johnson, et alia (1957).

Beryllia-uranium dioxide-thoria

The additional advantages of this dispersion are:

(i) thoria is present as an "inbuilt breeder" and

(ii) bodies made from this material are stable to oxidation.

This system has been studied at Harwell (Reeve and Williams, 1958). Because a eutectic has been observed in the beryllia-thoria system at approximately 2,200°C and almost the same beryllia composition as for the beryllia-uranium dioxide eutectic, viz., 60-70 mol.% BeO (Dukelski, 1906), and because uranium dioxide and thoria form a complete series of solid solutions, it was expected that the ternary system would behave as a pseudobinary system. This has been confirmed. The maximum allowable temperature for use of beryllia-uranium dioxide-thoria would be somewhat less than 2,150°, because it has been observed that at 2,150° a liquid phase is beginning to penetrate the beryllia matrix. This is obviously undesirable in a dispersion-type fuel element. Fabrication of such dispersions in beryllia to near theoretical density is easier to achieve than for pure beryllia.

Other oxides as matrix

Other possible matrix oxides are magnesia, alumina, silica, and zirconia. Thermal conductivities are inferior to beryllia; possible advantages are superior resistance of the matrix to neutron irradiation, and superior retention of fission product gases. At present not enough data is available on which to base a choice. Some data on fission product retention in magnesia-uranium dioxide and alumina-silica-uranium dioxide bodies has been discussed by Williams (1957). Dispersions of 100 micron uranium dioxide in magnesia of 75 per cent. theoretical density released only 20 per cent. of the xenon produced at 800°C. However, as magnesia and uranium dioxide have some mutual solid solubility, this is not an ideal dispersion system since solution may occur slowly during high temperature use. Xenon release from a 100 micron uranium oxide dispersion in an impervious alumina body containing some silica bond was negligible at 800°C. However, uranium dioxide was found to be present in solution in the glassy bond; which is not desirable in a dispersion type fuel.

Silica and zirconia both suffer from phase changes, and there is little to recommend them at present, although fused silica is reported to be stable under irradiation (Simon, 1957).

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The "Jigged-Bed" Ion Exchange Pilot Plant for Uranium Extraction

By G. L. Herwig, R. M. Stewart* E. A. Swinton, and D. E. Weiss†

A high efficiency technique for the extraction of uranium without the necessity for countercurrent decantation or filtration is being developed by C.S.I.R.O., in conjunction with the Australian Atomic Energy Commission, Territory Enterprises Pty. Ltd., and the Permutit Co. Ltd., of England. Rio Tinto (Australia) Ltd. has also contributed to the work. After leaching under normal conditions, the leach pulp is split by a small hydrocyclone into a slimes fraction of 20 to 30 per cent. solids content, and an underflow of sands at 60 to 80 per cent. solids. The sands are washed by gravity flow through a cascade of low-head cyclones and discharged with virtually no loss of soluble uranium. The slimes, at a pulp density between 20 and 30 per cent., depending on the characteristics of the ore, are pumped upwards through a column, which contains a floating bed of a special coarse size ion-exchange resin held against a screen across the top of the column by the flow of pulp. Blockage of the bed and the screen are prevented by a slow, gentle pulsation applied to the flow of pulp. To reduce the size of the plant, the operation has been made continuous—eluted resin being fed to the top of the column, and uranium-loaded resin being taken from the base of the bed, washed and eluted in a separate system. Two extreme types of ore have been tested on a five to 10-ton per day scale in a column 1ft. in diameter. On both a free settling and a poor settling ore the efficiency has been found to be such that very substantial savings could be achieved over conventional plants. Prolonged trouble-free operation has been obtained during tests at Rum Jungle into the application of the technique to Dyson's ore.

Introduction

A continuous adsorption process, the "jigged-bed process," was developed in the laboratories of the C.S.I.R.O., and it enabled a fine suspension to be passed through a virtually compact bed of a granular adsorbent, by applying a pulsation to the flow. It was suggested that the technique might be applicable to the extraction of uranium from leach pulps by ion-exchange. It is necessary for conventional ion-exchange (Preuss and Kunin 1955) to prepare a clear solution from the leach pulp either by filtration and washing, or by countercurrent decantation. This step is always expensive, sometimes prohibitively so, and incurs substantial losses of dissolved uranium in the discarded solids. A technique which would enable ion-exchange resins to be efficiently contacted directly with the leach pulps, and thus eliminate filtration, would be very desirable.

The United States Atomic Energy Commission, at Grand Junction, U.S.A., has developed a simple and direct method for treating dilute slimes (5-8 per cent. solids) with a special ion-exchange resin of 10-20 mesh bead size (Hollis and McArthur 1955). It is termed the "basket machine" resin-in-pulp (R.I.P.) process and five commercial plants are operating on this principle in the U.S.A. The process involves multiple stagewise contact of the resin with the pulp, and requires a large plant. It is only justified economically when the ore to be handled is very difficult to filter or settle. A continuous process has also been developed based on the Higgins "jerked-bed" contactor, but this process handles dilute slimes (5-8 per cent. solids), the particle size of which is critical (Arehart *et al.* 1956). A multistage mixer-settler process has also been developed (Infilco-Techmanix), using airlifts to mix the resin and pulp.

If an R.I.P. process could be developed in which the actual ion-exchange process was almost as cheap and efficient as that of the conventional ion-exchange process, the economies to be derived from the elimination of the filtration or decantation step could be realised in full, and R.I.P. could be advantageous even with ores with good filtration or settling characteristics.

After demonstration of the "jigged-bed" principle in the laboratories of C.S.I.R.O. in 1954, a pilot-plant column (1ft. in diameter) was developed, with the assistance, both financial and technical, of the Australian Atomic Energy Commission, Rio Tinto Co. Ltd. (Mary Kathleen Uranium) and Territory Enterprises Pty. Ltd. (Rum Jungle). Pulps investigated have been prepared from two widely different samples of ore possessing bad and excellent filtration characteristics. After initial trials on White's ore in August 1955, work was performed on a sample of free-filtering ore from Mary Kathleen, from September 1955 to April 1956, on both a continuous and batchwise basis. Trials on Dyson's ore, from Rum Jungle, were commenced in March 1957, and after modifications to the system the pilot plant was reassembled on the mill site at Rum Jungle and tested under longer conditions of continuous operation during November and December 1957.

The jigged-bed process has been the subject of a series of patent applications in Australia and abroad. An early batch version of the pro-

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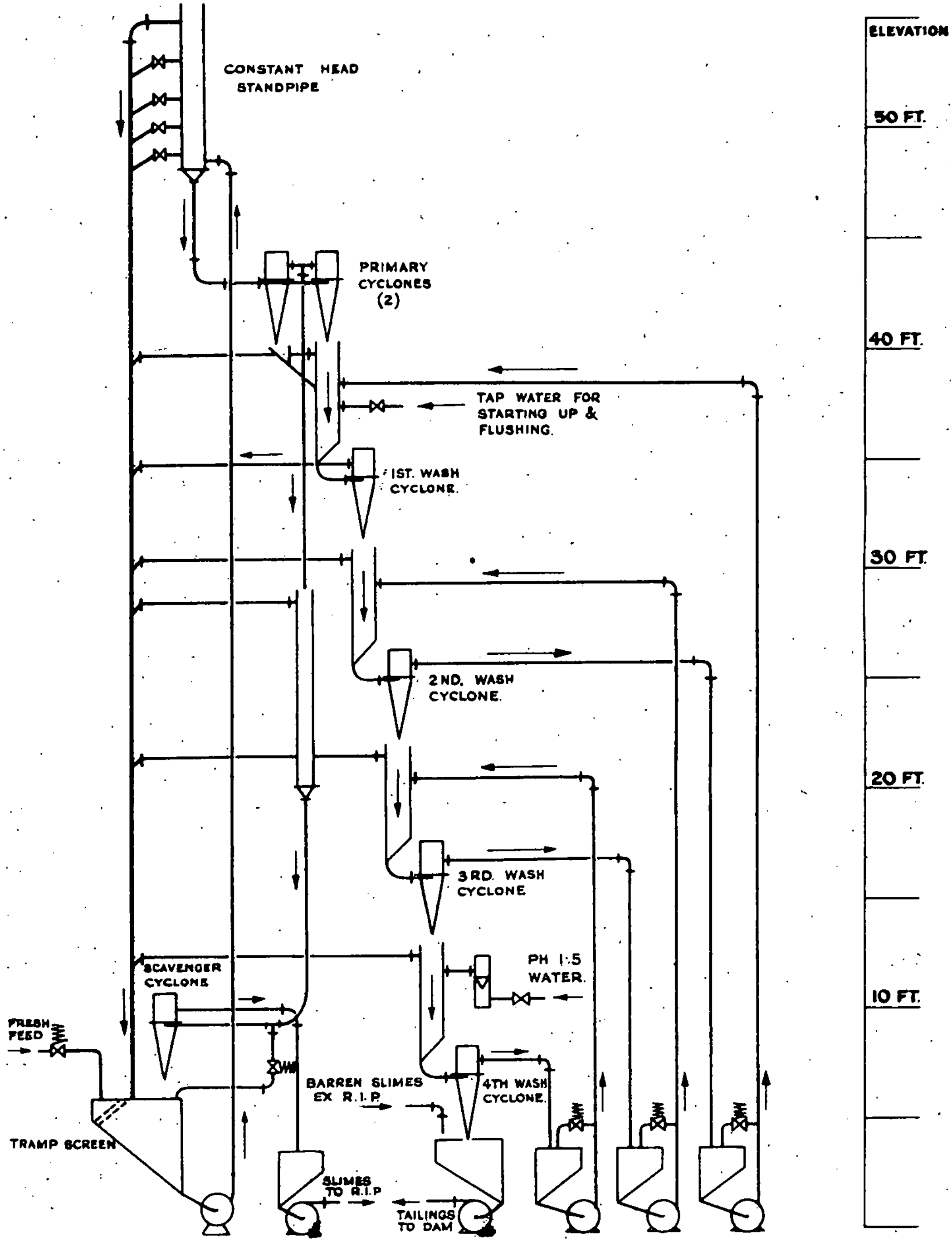

FIGURE 1:—Suggested layout for a 200-tonper day cyclone desanding unit.

cess has been previously described (Swinton and Weiss 1956) and also an earlier continuous ion-exchange process based on the principle of mineral jigging (McNeill, Swinton and Weiss 1955).

Development of the full scale process by the licensees, the Permutit Co. of London, is proceeding.

Description

In the jigged-bed technique, leach pulp from which the sands have been removed is pumped upwards through a column of anion-exchange resin at flow rates comparable with those through conventional ion-exchange columns. A bed of 10-20 mesh anion-exchange resin, floating in the pulp, forms a semi-compact bed against the underside of a 30 mesh screen across the top of the column. Blockage of the bed and the screen is prevented by applying a slow gentle pulsation to the flow of pulp by a pneumatic system. Substantially complete extraction of the uranium is obtained in one pass. The presence of the suspended slimes does not affect ion-exchange with the resin. Eluted resin is fed continuously into the top of the bed, and loaded resin is taken continuously from the base of the bed, washed, and eluted in a separate system to recover the uranium.

The column can deal with slime pulps containing up to 40 per cent. by weight of suspended solids, but sands must first be removed from coarse leach pulps. Passing leach pulps through a hydrocyclone yields an ideal product. It reduces a leach pulp containing 40 to 50 per cent. solids directly to a slime overflow containing 20 to 40 per cent. solids by removing the plus 300 mesh sands as an underflow containing 60 to 80 per cent. solids. The sands are washed very efficiently and economically by gravity flow down a cascade of hydrocyclones operating with a low pressure drop, a small amount of wash water being pumped countercurrently to the flow of sands.

Adsorption column

During the development programme, various systems were tried out, but in this paper only the five-ton per day pilot plant as finally developed and operated at Rum Jungle will be described in the following sections.

Pulp preparation

Grinding and leaching of 60-ton samples of ore was conducted with mill equipment aiming at a nominal 10-mesh grind, which had been proved previously to leach satisfactorily with sulphuric acid at 50 per cent. pulp density. After leaching and adjusting the pH to a value of 1.5, with limestone, the pulp was held at leach consistency in the pachuca, and circulated through a ring main to prevent undue quantities of sands settling into the bottom outlet. Quantities were withdrawn daily for the removal of sands, and were passed through the cyclone unit which delivers slimes to two 1,800-gallon storage pachucas from which the feed to the jigged column was taken.

The cyclone system is shown diagrammatically in Figure 1, which is a line diagram for a design for 200 tons per day throughput. The pilot plant system was identical in form, but used only a single primary, a scavenger, and three washing cyclones. Fresh pulp from the storage pachucas was mixed in a sump at ground level with the wash waters from the sands washing section and diluted from 50 per cent. solids to 30 to 40 per cent. solids. This pulp was pumped by a 2in. rubber-lined centrifugal pump to a constant-head standpipe 40ft. above ground level, the excess overflowing back to the sump. The pulp in the standpipe, under a constant head of 15ft., flowed through a 3in. diameter hydrocyclone made from sheet steel lined with Linatex soft rubber. The inlet was approximately ¾in. in diameter, the vortex finder pipe ½in. in diameter, and the diameter of the outlet at the apex of the cone could be adjusted by a screw action from ⅜in. to $\frac{3}{16}$in. This cyclone was termed the primary cyclone.

The underflow, at 70 to 75 per cent. solids, comprised nearly all of the +300 mesh material, along with a proportion of entrained slimes. It fell directly into the head of a standpipe, 7ft. high, where it mixed with countercurrent wash water and then passed through the first washing cyclone, of 2½in. diameter. From the first washing cyclone, the underflow of sands, containing about 66 per cent. of solids and substantially free from slimes, fell directly into the succeeding standpipe and so on until the sands had been washed almost completely free of uranium by three stages of countercurrent decantation with the ratio of overflow to underflow solutions being between 5 to 1 and 10 to 1. Acidified wash water was metered to the standpipe feeding the last washing cyclone and the overflow from this was pumped by a ¾in. pump to the standpipe feeding the penultimate cyclone, and so on.

The overflow from the primary cyclone at the top of the cascade ran into a standpipe which fed a 3in. diameter "scavenger cyclone" near ground level. The overflow from the scavenger cyclone was pumped away as product. The underflow, normally a 30 per cent. suspension of 200 to 300 mesh sands, fell into the original feed sump for recycling. In the event of sand surges or blockages, causing the overflow from the primary cyclone to contain a coarser product than normal, the scavenger cyclone would discharge these sands back to the system, thus protecting the quality of the product. A control system actuated according to the density of the underflow from the scavenger cyclone, could operate a valve for discharging sands before the scavenger cyclone became overloaded with them, but in pilot plant operation this was done manually.

Only one pump—the feed pump to the primary cyclone—dealt with abrasive pulp. All other pumps elevated overflows from the washing system, which were either clear solutions or dilute suspensions of slimes. Sand slurries were not pumped at all so that blockages which would have arisen from settling were eliminated. The standpipe system in the washing cascade allowed the system to become self-balancing since the height of pulp in each standpipe adjusted itself to the flows required. In the event of blockage of any cyclone, the respective standpipe overflowed through a safety line back to the original feed sump, raised the level therein and stopped the feed of leach pulp by means of a valve controlled by a float.

The complete system, using a primary cyclone having a diameter of 3in., dealt with 10 to 15 tons of ore per 24 hours, and could be controlled to yield any pulp density required between 15 to 35 per cent. solids. The proportion of +300 mesh material in the slimes product (excluding 100 mesh trash) varied according to the pulp density produced, from 1 per cent. of the suspended solids at 20 per cent. pulp density to 5 per cent. of the suspended solids at 35 per cent. pulp density. Even the pilot scale unit required no more than occasional supervision and simple techniques were devised to enable the plant to be stopped and started at short notice without trouble.

Slimes from the cyclones were brought to the required pulp density in two 1,800-gallon storage pachucas, and as required, were pumped to a small feed tank through a vibrating stainless steel screen, which removed wood fibres and other trash not removed by the cyclones. It has been proved that 100-mesh screening is necessary to prevent the ion-exchange bed from blocking, but this has not caused any difficulty since an area of 2sq. ft. of vibrated screen is ample to deal with 200 gallons per hour of desanded pulp.

Adsorption column

The adsorption column, shown diagrammatically in Figure 2, consisted of a 3ft. 6in. length of rubber-lined steel pipe, flanged at both ends, and fitted with sight glasses and branches. The pilot plant column, nominally 1ft. internal diameter, was slightly tapered, from 11½in. at the top to 13in. at the base. It was closed at the top by a 30-mesh screen made from monofilament nylon supported on a performated perspex plate, with an open overflow section above it. The base was closed by a shallow rubber-lined steel section provided with a side inlet of 3in. diameter. A

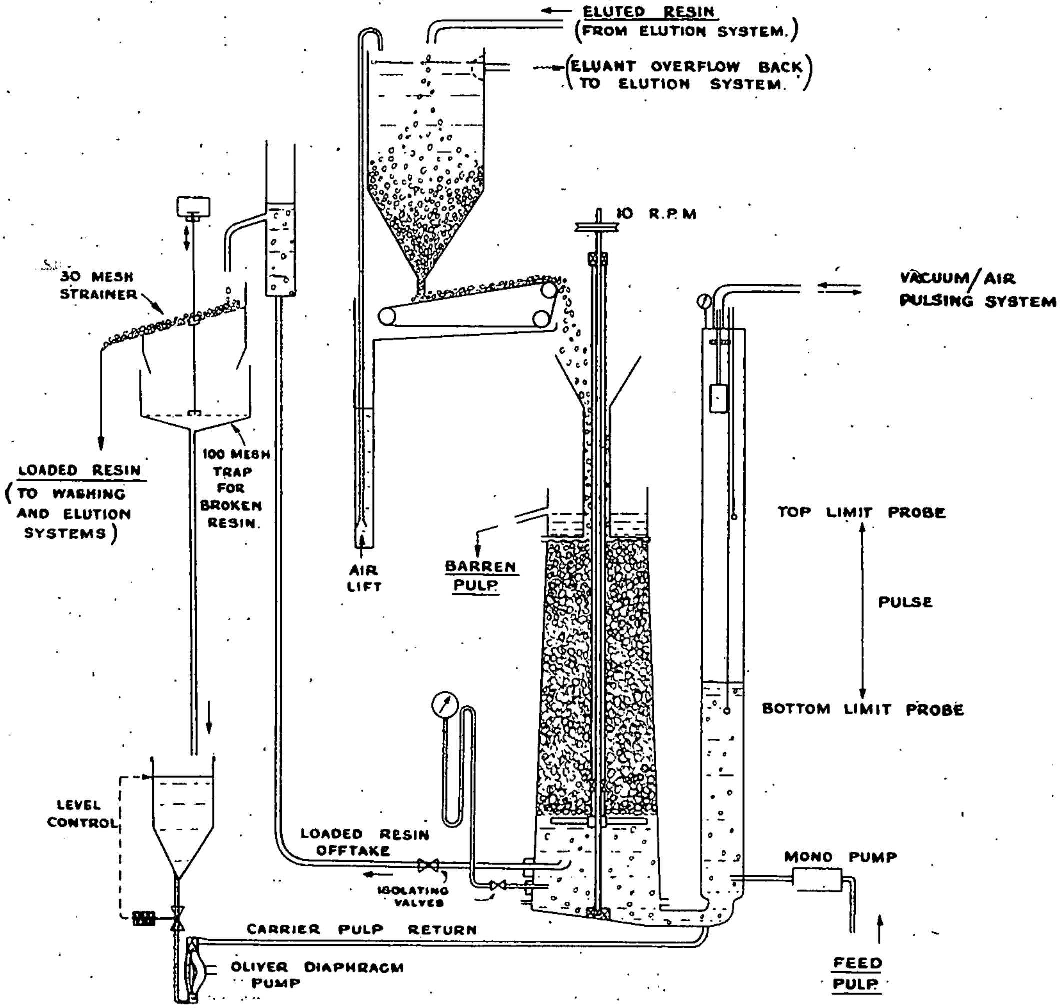

FIGURE 2:—Schematic diagram of the "jigged-bed" adsorption unit.

combination of a sloping base and a baffle plate provided as much distribution of the flow as was necessary.

The 3in. inlet led through a right-angled bend to the pulsator pipe which was 4in. in diameter and 6ft. high. Pulp was pumped into the base of this pipe through a variable speed "Mono" pump, which did not allow pulsations to pass back into the feed pipe and provided positive control of the feed rate. The pulp flowed up through the bed of ion-exchange resin and out over the overflow at the top of the column, at rates between one and three gallons per minute.

A pulse was generated by applying vacuum and compressed air alternately to the top of the pulsator pipe by means of solenoid valves. The pulse was controlled by a timing cam and limit switches. The amplitude could be adjusted to suit the particular pulp viscosity and flowrate by adjusting the pressure of the air and vacuum, and the levels of the limit switches.

A cylindrical hopper of 3in. diameter just protruded through the nylon screen at the top of the adsorption column. Eluted resin was metered into this hopper, and was drawn into the column by the pulsation to form a semi-compact bed of resin extending from the screen downwards to a depth of 2ft. 9in. The pulse generated a movement of one to 2in. in the bed of resin at a frequency of 10 strokes per minute.

Loaded resin offtake system

Resin, containing adsorbed uranium, was withdrawn from the base of the bed by running a flow of about a half a gallon per minute of the feed pulp out through a spigot placed below the bed and through a 10ft. length of ½in. diameter polythene tube into a small surge tank so that the surge caused by the pulsation in the adsorption column was damped. From the surge tank the flow gravitated to a 30-mesh screen, which strained off the loaded resin entrained in this flow. For plant operation, a displacement wash on this screen would be desirable. The pulp passing through the screen was pumped back to the base of the column through a 100-mesh screen to remove fragments of broken resin.

Loaded resin wash system

The resin was next passed over a 30-mesh vibrated nylon screen submerged in water to take out broken resin resulting from attrition occurring in the adsorption column. In plant operation resin fragments must be removed either continuously or intermittently, but preferably the screening should be performed on the eluted resin. The loaded resin was then given a thorough wash to remove adhering pulp, by being fed continuously into a steep-sided cone where it fell in turbulent flow against a rising stream of water which was injected at a point 1ft. above the apex of the cone and overflowed the cone through a guard screen. The resin settled below the point of water injection and was taken off continuously as a drained cake by mounting the outlet at the apex of the cone one to 2in. above a moving belt made from 30-mesh nylon screen running over rollers. Water drained through the resin and through the open mesh of the belt at such a speed that a carpet of resin was laid down of constant cross-section in the same way as a belt feeder operates on a dry granular solid. By sloping the belt gently upwards to a height of about 4in. above the loading point, water held in the interstices of the resin cake drained back by capillary action, yielding an almost dry cake which discharged from the belt as it passed over the upper roller. All of the washings were passed through a 100-mesh screen to recover and measure the amount of broken resin. In normal operation the washings would be used in the cyclone unit for washing sands.

Elution apparatus

The elution equipment to be used on a plant scale was being developed simultaneously by the Permutit Co. of London, and will not be described here. In the 1ft. diameter pilot plant an "elution jig" was used as a matter of convenience because it was developed some time previously and, though clumsy, operated reliably (McNeill, Swinton and Weiss 1955). It was recognized that it was loaded beyond its capacity, and that it was to be superseded, so no further development work was performed. However, some features may be of interest, and a brief description is included.

A rectangular trough was divided into six cells 9in. square, 1ft. deep by transverse partitions. Each cell contained a 30-mesh nylon screen near its base, and a cam-driven diaphragm pulsator below the screen. The diaphragm compartments were separated one from the other, but above the screen the cells intercommunicated through a system of transverse weirs. In each cell a weir retained a bed of resin to a depth of two to 4in. and an inverted weir dipping down into the bed of resin from the top of the trough separated zones of clear liquid 6in. deep above these beds. Each pair of weirs was so arranged that resin flowed from the top of the bed in one cell to the base of the bed in the succeeding cell.

By pulsating the liquid in the cells by the diaphragm as in a mineral jig, the resin bed became semifluidised and flowed from cell to cell by virtue of its own hydrostatic head. A counter-current flow of eluant was obtained by passing the flow upwards through the last resin bed, and by allowing the clear liquor above the bed to overflow a weir. It was then elevated one to 2in. by a rudimentary airlift into a standpipe feeding the next cell, so that it would flow up through the next bed in turn. The overflow weirs in each cell were adjusted to maintain a hydrostatic balance between each cell such that there was negligible flow of liquid through the labyrinth along which the resin flowed. This adjustment was stable.

Resin overflowed from the last cell into a sump from which it was elevated by an airlift into a settling cone which provided a storage of eluted resin submerged in eluant. Liquor from the settling cone overflowed back to the airlift sump.

Eluted resin feed

Resin flowed from the base of the eluted resin settling cone through a spigot of 1in. diameter on to another 30 mesh nylon belt feeder. The speed of this belt was used to control the flowrate of resin throughout the whole system. Eluant draining through the belt was returned to the settling cone by an airlift.

The resin was drained by sloping the belt upwards as previously described and could readily have been rinsed if desired. It was then discharged into the hopper at the top of the adsorption column. Each downward pulse in the adsorption column retracted the resin bed from the upper screen by a distance of one to 2in., leaving a clear space filled with pulp. Fresh resin dropped down the 3in. hopper and spread out over the exposed surface of the bed. On the succeeding upstroke the resin had re-compacted against the upper screen and became non-mobile, so that resin did not return to any great extent up the feed hopper. The type of pulse aimed at was a sharp downstroke, followed by a slow return stroke. Under these conditions, the bed became loose and semi-mobile for approximately two thirds of the time. When the resin became heavily loaded with uranium, its density is such that it falls from the base of the column. It is from this zone that loaded resin is withdrawn. When using pulps of high density or when the resin is relatively lightly loaded, then loaded resin will remain floating in the upflow of pulp. It is then necessary to ensure that the base of the bed is disturbed to release the particles of loaded resin. This can be done by a suitable arrangement of flow patterns in the base of the column, but in the 1ft. column, for reasons of experimental convenience, a slowly rotating rabble arm was installed to ensure a constant depth of bed. At the end of each downstroke, when the bed was in a fully expanded and loose condition, the rabble blades swept the bottom layer of resin into the turbulent zone.

Experimental results

Performance

CYCLONES

Operations of the cyclones has been tried successfully on both Dyson's and White's ores, and full data will be presented elsewhere. A summary of average operating figures for Dyson's pulp is given in Tables I and II. This ore gives an unbalanced grind, containing a large proportion of very fine native slimes, together with sands resulting from coarse grinding of the gangue. The cyclone system separated these most efficiently, yielding a product containing less than two per cent. of plus 300 mesh material.

2.1 gall./min. of leach pulp were processed, yielding 3.3 gall. min. of slimes product at 25 per cent. solids (s.g. of pulp, 1.243; s.g. of contained solution, 1.05). Half the original ore was discharged in the sands fraction, and was washed with 2 gall./min. of acidified wash water.

ION-EXCHANGE

The first trials of the jigged-bed column in its original form were made on pulps prepared from White's ore from Rum Jungle. The column was then improved, and a number of short runs were performed on pulps prepared from ore from the Mary Kathleen Mine, which settled very freely. More extended trials were then made on Dyson's ore from Rum Jungle, which contained a considerable amount of clay.

TABLE 1:—CYCLONE PERFORMANCE OF DYSON'S PULP.

(Thirteen tons of ore per 24 hours)

Pulp Flows

Overflows	Wash Water	Scavenger Slimes	Primary	1st Wash	2nd Wash	3rd Wash
Solids: lb./min	—	10.5	12.46	1.45	0.27	0
Liquids: lb./min	20.9	30.6	35.4	21.95	21.50	21.25
Pulp density		25.5	26	6.2	1.2	0
Underflows	Feed Pulp					Sands
Solids: lb./min	20.5	1.91	11.45	10.3	10.0	10.0
Liquids: lb./min	14.75	4.4	5.15	5.53	5.39	5.04
Pulp density	58	30	69	65	65	66.5
U_3O_8, per cent. of feed in underflows						
Theoretical*	100		16.7	4.35	1.05	0.2
Measured			16.9	6.1	2.6	0.6

U_3O recovery: 99.4 per cent.
Ore split: 51 per cent. to slimes.
Pulp density of slimes: 25 per cent. W/w.
Wash ratio: 4 to 1.

* Derived from mass balance calculations.
Primary and scavenger cyclones: 3 in. diam.
Washing cyclones: 2½ in. diam.

TABLE 2.—SCREEN ANALYSES OF SUSPENDED SOLIDS.

(Per cent. of total dry weight retained on screen named.)

	Feed	Slimes
B.S.S. +100	31.2	0.02
+200	10.6	0.35
+300	6.8	1.42
—300	51.4	98.2

The trials have shown that the following conditions are necessary for satisfactory operation. The resin must consist of substantially 10-20 mesh beads of good mechanical strength. Sands must be removed from the leach pulp, which must be passed through a 100 mesh screen to remove trash to give a pulp containing between 15 to 40 per cent. total solids w/w, dependent on the viscosity, and which must not contain any solids coarser than 100 mesh nor contain more than about 5 per cent. of sands having a particle size exceeding 300 mesh. Flowrates of one to three gall. per sq.ft./min. may be successfully employed. A pulse with a frequency of five to 10 strokes/min. with an amplitude of one to 3in. is satisfactory. The volumetric displacement of the pulp by the pulsation should be approximately five to 10 times the volumetric increment due to the flowrate. A bed of resin from three to 6ft. deep is practicable, and resin may be circulated at rates up to 0.25 gal. w.s.b.*/sq.ft./min. Reliable data are not available for the attrition loss, but on the basis of the figures obtained it is not likely to exceed 100 per cent. per annum of the volume subjected to pulsations. The pressure-drop through the bed is low, as evinced in the following typical examples. For Dyson's ore pulp at a density of 20 per cent. suspended solids, a head of 4ft. in a standpipe produced a flow of 2.3 gall./sq.ft./min. through a bed 3ft. deep, with a pulsation amplitude of 2in. at 10 strokes/min. At the end of each upstroke a momentary peak pressure of 9 lb./sq.in. was registered (equivalent to a head of 16ft. of pulp). For the less viscous Mary Kathleen ore pulp at a density of 20 per cent. solids, a head of 2ft. in a standpipe produced a flow of 1.5 gall./sq.ft./min. through a bed of 3.2ft. deep. For White's pulp, a peak pressure of 4 lb./sq.in. was sufficient to pass 2.3 gall./sq.ft./min. of 33 per cent. pulp min. through a bed 3.0ft. deep.

Prior to the testing of the plant at Rum Jungle, trials had been limited by an inadequate supply of pulp to durations of eight to 15 hours, and no conclusions could be drawn on the long-term practicability of the technique. However, the test run at Rum Jungle on Dyson's ore ran for seven days until it was stopped deliberately. No changes in pressure drop or bed behaviour were observed in that time. The column dealt with pulp densities varying between 1.200 and 1.225, as would have to be expected in mill operation. The column could be shut down for power cuts or maintenance purposes, and restarted within seconds, and gave no trouble. Feed and withdrawal of the resin was simple and reliable. The rate at which loaded resin emerged adjusted itself within 10 to 15 minutes after resetting the rate of resin feed to a new value. The operation was so free of trouble that truly long-term testing has been deferred until a full-scale prototype, having a diameter of 4ft., can be installed.

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*Wet, settled bed of resin.

TABLE 3.—BATCH RUNS ON MARY KATHLEEN PULPS USING AMBERLITE XE123*.

Run No.:	1	2	3	4	5	6	7
U_3O_8 head: gm./litre	4.5	4.4	4.3	2.8	2.0	3.4	3.4
Barrens before breakthrough: gm./litre	.014	.001	.010	.001	.007	.003	.008
Flowrate (gall./sq. ft./min.)	0.95	0.9	1.06	1.23	1.84	1.82	2.05
Bed depth (ft.)	5.0	4.3	4.5	4.5	4.5	4.2	4.5
Average loading at breakthrough: gm. U_3O_8/litre w.s.b.	54	66	43	34	31+	38	37
Solids in pulp, per cent. W/w	25	22	18	20	20	15	15

BREAKTHROUGH CURVE OF RUN 2.

Bed vols. through	U_3O_8 in barrens, gm./litre	Bed vols. through	U_3O_8 in barrens, gm./litre
1	0.001	13.6	0.010
2	0.006	13.9	0.028
5	0.001	14.3	0.090
8	0.001	15.3	0.14
9	0.001	15.7	0.39
11	0.001	16.0	0.43
12	0.001		

* Hohm and Haas Co., Philadelphia, U.S.A.

TABLE 4.—CONTINUOUS RUNS ON MARY KATHLEEN PULPS USING AMBERLITE XE123.

Run No.:		1	2
Duration of run:	hr.	8	8
U_3O_8 in liquor—			
Head:	gm./litre	2.7	3.4
Barrens:	gm./litre	0.020	0.020
Recovery:	per cent.	99.3	99.4
Flowrate of pulp:	gall./sq. ft./min.	1.43	1.23
Solids in pulps:	per cent. W/w	19	18
Volume of resin per sq. ft.:	cu. ft.	3.2	3.0
Depth of compact bed:	ft.	3.2	1.5
U_3O_8 in resin—			
Eluted:	gm./litre w.s.b.	2.8	2.7
Loaded:	gm./litre w.s.b.	44	37
Flowrate of resin:	gall./sq. ft./min.	0.061	0.084
U_3O_8 in eluate:	gm./litre	8.4	12.5
U_3O_8 in barrens in equilibrium	gm./litre	8.4	12.5
with eluted resin:	gm./litre	0.014	0.014
By difference, leakage from column:	gm./litre	0.006	0.006

Efficiency of uranium recovery

CYCLONE-SAND WASHING

Figures for the soluble uranium content of the various underflows are also given in Table I. The measured loss of uranium was two to three times more than the theoretical loss calculated from flow balances. This could be due either to inefficient interstate mixing of the sands with the washings, or to slow diffusion of dissolved uranium from the pores of large particles of grit. Increased residence time and mixing in the interstage standpipes is therefore desirable. Even so, with only three stages of washing, the uranium recovery was 99.4 per cent. A further stage of washing would have increased this to 99.8.

ION-EXCHANGE

Some figures for ion-exchange operation are given in Tables 2, 4 and 5.

From Table 3, it will be seen that in batchwise operation, according to the procedure described elsewhere (Swinton and Weiss 1956), the jigged-bed had an ion-exchange efficiency almost equivalent to that of a conventional column. Thus, the loss of efficiency due to back-mixing caused by the pulsation must have been very small, or perhaps compensated by an increase in efficiency due to reduction in channelling.

The results of two of the short continuous runs on Mary Kathleen pulps are summarised in Table 3. The specific gravity of the pulp was approximately 1.20, and it contained 19 per cent. by weight of suspended solids; flowrates, based on the volume of solution contained in the pulp, were 1.2 to 1.5 gall./sq.ft./min. In steady state, the adsorption column contained about 2cu.ft. of resin, but in the second example, because of slightly lower pulp density and flowrate, only a part of the resin was entrained against the upper screen to form a semi-compact bed and the heavier loaded resin fell into a fluidised zone in the lower part of the column. Provided that a compact bed at least 1.5ft. deep was maintained, there was no difference in the adsorption of uranium. The greater bulk of the uranium was absorbed in the fluidised-zone, and the compact bed stripped out the remainder of the uranium by "chromatographic" action.

Table 5 summarises the ion-exchange results on a longer run on Dyson's pulp, using the final form of the column.

In general, in these and other runs, resins with loadings of 25 to 56 gm. U_3O_8/litre w.s.b. were taken off from the adsorption column, depending on the relative rates of flow of leach liquor and resin. From these resins, eluates containing from four to 14 gm. U_3O_8/litre were produced.

In all runs the elution system was overloaded, and in consequence incompletely eluted resin was recycled back to the adsorption column. A proportion of the loss in the barrens pulp was due to the inefficiency of the elution system rather than to leakage through the adsorption bed, because uranium can only be stripped from the pulp to a limiting concentration which is in equilibrium with the eluted resin fed to the column. The equilibrium concentrations were determined by analysis, and the relevant figures are noted in Tables 4 and 5, where it will be seen that loss of uranium due to the leakage from the adsorption bed was actually very low. The U_3O_8 content of the barren pulp would have been reduced even further by using a bed 5ft. deep, such as was used in the batchwise runs summarised in Table 2. The solution would then have had a residence time in the resin bed of 6 mins. at a feed rate of 2.1 gall./sq.ft./min., or 10min. at 1.2 gall./sq.ft./min.

An understanding of the way in which the efficiency of uranium stripping varies with differing bed depths, pulp flow rates, resin flowrates and elution conditions can be obtained by using standard procedures of chemical engineer-

TABLE 5.—CONTINUOUS TRIAL ON DYSON'S PULP USING PERMUTIT, LONDON, R.I.P. RESIN.

Run No.:		1	2
Time of readings:	hr. from start	23	160-170
U_3O_8 in liquor—			
Head:	gm./litre	1.3	1.04
Barrens:	gm./litre	0.008	0.028
Recovery:	per cent.	99.4	97.8
Flowrate of pulp:	gall./sq. ft./min.	2.20	2.33
Solids in pulps:	per cent. W/w	20	20
Volume of resin per sq. ft.	cu. ft.	2.7	2.7
Depth of compact bed:	ft.	2.7	2.7
U_3O_8 in resin—			
Eluted:	gm./litre w.s.b.	0.7	3.7
Loaded:	gm./litre w.s.b.	43	31
Flowrate of resin:	gall./sq. ft./min.	0.060	0.084
U_3O_8 in eluted:	gm./litre	4.05	3.8
U_3O_8 in barrens in equilibrium with eluted resin:	gm./litre	0.004	0.019
By difference, leakage from column:	gm./litre	0.004	0.009

1. Best recovery during the run.

2. Average of 20 readings.

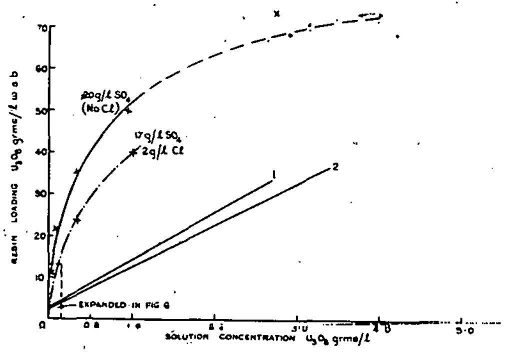

FIGURE 3:—Adsorption equilibrium curves and operating lines for continuous runs on the Mary Kathleen ore using Amberlite XE-123 resin.

ing analagous to those employed for solvent extraction and distillation calculations. An equilibrium curve was plotted for each leach liquor and type of resin used in the column. Small quantities of resin, in the sulphate form, were equilibrated overnight with varying volumes of partly stripped barren liquors, and partly loaded resins were equilibrated with varying volumes of feed solution. The resin and solution phases were analysed separately for their uranium content. The uranium concentration in the various liquors was plotted against the loading of resin in equilibrium with those liquors. Similar curves could be obtained by running a laboratory column on clear solution until breakthrough just occurred, stopping the flow to allow the column to reach equilibrium and then sampling and analysing resin and ambient liquor at various levels. These experimentally derived curves provide an overall measure of the influence of competing ions on the adsorption of uranyl ions by the resin, even though the exact nature of these ions and the mechanism of adsorption is not completely understood.

An example is given in Figure 3 for a leach liquor from Mary Kathleen ore. The empirical curves for other leach liquors are similar in form, but differ by a few per cent. in magnitude depending on the value of the pH and the concentration of competitive anions, particularly chloride and bisulphate ions. It will be seen that resin can be loaded to a maximum of 50 gm. U_3O_8/litre w.s.b., the saturation loading, from a feed solution containing 1.0 gm. U_3O_8/litre. A magnified portion of the equilibrium curve near the origin is given in Figure 4. From this plot it will be seen that resin containing 1 gm. U_3O_8/litre w.s.b. is in equilibrium with a liquor containing 0.0045 gm. U_3O_8/litre, and therefore can only strip a barren liquor to the latter minimum value.

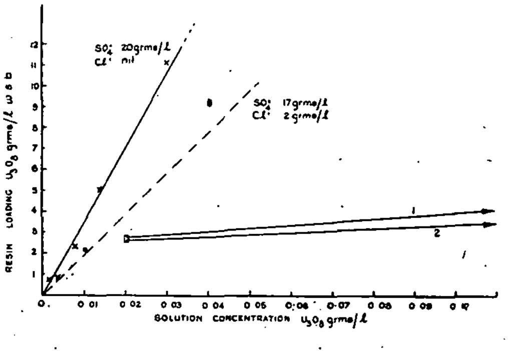

FIGURE 4:—Expanded drawing of the lower portion of Figure 3.

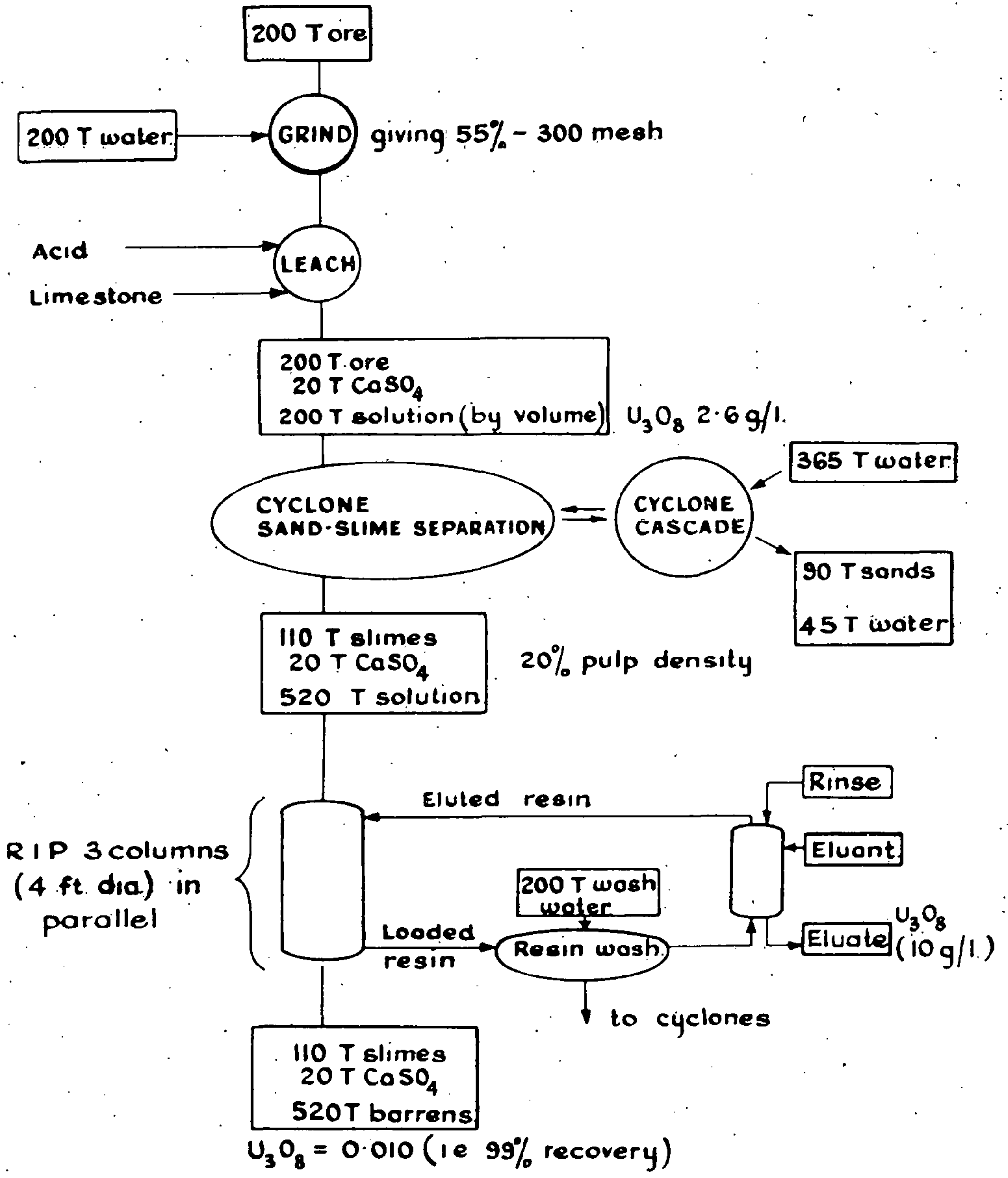

FIGURE 5:—Proposed flowsheet for the treatment of uranium ore A by the "jigged-bed" process.

On these same axes can be plotted the values for the concentrations actually measured in a column in steady state operating conditions, i.e., barrens concentration versus that of the eluted resin, and feed concentration versus that of the loaded resin. The line joining these points is termed the operating line. The concentration differences between the operating line (actual conditions) and the equilibrium curve (the theoretically attainable condition of equilibrium) is a measure of the efficiency of the ion-exchange operation; the more efficient the contacting apparatus, and the faster the rate of reaction of the resin, the closer will operating values approach the limiting equilibrium values.

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The concentration differences can be reduced to a mean figure by using the transfer unit technique involving graphical integration, as in other mass transfer calculations. However, for the data available at this stage, it was sufficiently accurate to approximate the equilibrium curves to straight lines, over the ranges involved, and then to use the logarithmic mean of the end concentration differences, measured along the solution axis.

The mean concentration differences were calculated by this means for the various runs and were inserted, with the other relevant figures, into the usual mass transfer equation

U = K.V. Cm

where U = lb. U_3O_8 extracted per hr.;
V = volume of resin in column (100cu.ft.);
Cm = mean concentration differences (gm. U_3O_8/litre).

K, an empirical factor based on experiment, is then an overall measure of the contacting efficiency of the column. It proved to be fairly constant over the last five runs on the original 1ft. diameter glass column, varying from 2.8 to 3.1lb. U_3O_8/min./100cu.ft. for a mean concentration difference of 1 gm./litre. On the final column, values varied from 4 to 6. By reversing the procedure, this value of the coefficient so obtained can be used to calculate the performance of the column with a fair degree of certainty with differing variables, provided that the flow conditions and bed behaviour are similar to those from which the coefficient is derived. The same type of mathematical analysis can be applied to a continuous elution column. In the case of a stagewise contactor, the McCabe-Thiele stepwise construction can be used.

The loss of uranium in the barren pulp can be calculated for various flowrates of resin, using the experimentally derived factor K, by defining the volume of resin in a column, the flowrate and head concentration of the feed pulp, and the concentration of uranium left in the eluted resin fed to the column. Similarly, the variation of extraction efficiency with pulp flowrate can be calculated. Similar calculations can be made for the performance of the elution equipment. These calculations are valid because they are based on the use of experimentally derived parameters (the equilibrium curve and the value of K) which effectively take into account the many unknown factors concerning the detailed mechanism of the adsorption process.

It is possible to use such calculations to define conditions for operation at a minimum cost, where increased capital outlay on ion-exchange equipment and increased running costs would not bring about economic returns from further recovery of uranium. Such a calculation shows that optimum operation requires a pulp flowrate of 1.5 to 2.5 gall. per sq./ft./min., a resin bed five to 6ft. deep, with the resin flow such that it becomes loaded to about 75 per cent. of saturation (i.e., 35 to 40 gm. U_3O_8/litre w.s.b. from a feed solution containing approximately 1.2 gm. U_3O_8/litre). The elution equipment should elute the resin down to 0.5 gm. U_3O_8/litre w.s.b., and yield a rich eluate containing about 12 gm. U_3O_8/litre. The barren pulp resulting from these conditions would contain less than 0.010 gm. U_3O_8/litre. All these conditions have been encountered in the experimental program.

Discussion

It will not be possible to assess the economics of the process accurately until a full scale jigged-bed unit of 4ft. diameter has been operated for an extended period. However, reliable estimates can be made of the savings to be expected from the replacement of thickeners by cyclones, and a comparison can be made of the relative size of the jigged-bed and conventional ion-exchange units.

Tables 6 and 7 summarise these figures, and for the purpose of this paper the estimates are presented for the hypothetical case of a new installation to treat 200 tons per day of an ore A, which forms a viscous pulp that settles and filters badly. Approximate estimates relating to a normal ore (B) have also been made, which illustrate that the easier an ore is to deal with by conventional methods, the easier it can be handled by cyclones and the jigged-bed process, but the margin for possible savings is less.

A projected flow sheet for ore A is given in Figure 5. Ore is ground to a nominal 10 mesh grind, which releases about 50 per cent. by weight of native slimes. It is assumed that leaching takes place at 50 per cent. pulp density, and that after desanding, 55 per cent. of the ore, with 20 tons of calcium sulphate resulting from pH adjustment, will report to the slimes fraction. The jigged-bed ion-exchange column is assumed to handle slimes at 20 per cent. solids at 2.33 gall./sq.ft./min., as in the pilot plant.

It is calculated that three columns of 4ft. diameter, with resin washing and elution equipment, would be required to handle the flow. For standby duty, a fourth column would have to be purchased.

The alternative conventional process utilises counter-current decantation involving four or five 75ft. diameter thickeners delivering underflows of 40 per cent. pulp density, with four conventional ion-exchange columns of 7ft. diameter.

For a normal ore, such as ore B, it has been shown that because of the much lower viscosity the jigged-bed columns can handle pulps of up to 33 per cent. pulp density more easily than they handle the 20 per cent. pulps of ore A. The volume of pulp to be treated is therefore much less and only two 4ft. jigged-bed columns would be required, though again a standby unit would have to be installed. In the corresponding conventional flowsheet, thickeners of 50ft. diameter would be required, and the washing efficiency would be higher because the under flows would be at 50 per cent. pulp density.

TABLE 6.—ESTIMATED COMPARATIVE COSTS FOR TREATING 200 TONS PER DAY OF AN ORE WHICH SETTLES POORLY, ORE A.

Pulp preparation section	Conventional C.C.D.	Jigged-bed
Capital Costs.		
Thickener units : 4 x 75ft.	£274,000	—
Cyclone units : 7 x 10in.	—	£15,000
Operating costs per annum.		
Labour and maintenance :	£15,600	£11,700
Power :	5,400	4,500
Acidified wash water :	16,900	9,300
Flocculant :	7,800	—
Soluble loss of U_3O_8	63,500*	3,300†
TOTAL	£109,200	£28,800
Relative size of ion-exchange section △		
Volume of solution to be treated (tons/day)	550	520
Pulp density of feed (per cent. solids W/w)	0	20
Number of columns	4 x 7ft.	4 x 4ft. []
Volume of resin (cu. ft.)	800	520

* Based on loss of 3.8 per cent. U_3O_8.

† Based on loss of 0.2 per cent. U_3O_8.

△ Cost data is not yet available for this section of the plant, so a comparison is made of the relative size of the units.

[] One of the four columns is used for standby only, so attrition is calculated on ¾ x 520 = 390 cu. ft. resin.

TABLE 7.—ESTIMATED COMPARATIVE COSTS FOR TREATING 200 TONS PER DAY OF AN ORE WITH AVERAGE SETTLING CHARACTERISTICS, ORE B.

	Conventional C.C.D.	Jigged-bed
Pulp preparation section		
Capital Costs.		
Thickener units : 4 x 50ft.	£200,000	—
Cyclone units : 7 x 10in.	—	£15,000
Operating costs per annum.		
Labour and maintenance :	£14,000	£11,700
Power :	5,400	4,500
Acidified wash water :	14,000	2,800
Flocculant :	6,000	—
Soluble loss of U_3O_8 :	27,000*	2,700†
TOTAL	£66,400	£21,700
Relative size of ion-exchange section		
Volume of solution to be treated (tons/day)	550	280
Pulp density of feed (per cent. solids W/w)	0	33
Number of columns	4 x 7ft.	3 x 4ft. △
Volume of resin (cu. ft.)	800	390

* Based on loss of 1.6 per cent. U_3O_8, assuming same grade as for Table 6.

† Based on loss of 0.16 per cent. U_3O_8.

△ One of the three columns is used for standby only, so attrition is calculated on 2/3 x 390 = 260 cu. ft. resin.

The costs are compared in Tables 6 and 7, where it will be seen that the main saving lies in the installation of cyclones instead of thickeners. It is also estimated that cyclones would cost slightly less to operate than thickeners because of their extremely small size, and the fact that only one pump in the circuit is required to handle abrasive pulp. However, the big savings in running costs arise from the ease with which the sands faction can be washed free of uranium. Less wash water is used, and no flocculant is required. Loss of soluble uranium, which can be economically reduced only to one to four per cent. in countercurrent decantation plants, can be reduced to a negligible level by the cyclone system, firstly because of the much smaller volume of solution entrained in the high density underflow, and secondly because as many as seven cyclone decantations can readily be afforded.

The accurate costing of the ion-exchange step must be deferred until a prototype full-scale column has been tested over a long period, but a number of points of comparison can be made.

In the jigged-bed unit, resin is moved continuously out of the system as soon as it is loaded, and replaced by eluted resin so that there is no hold-up of idle resin as in a conventional fixed bed plant. The units, including the eludition units, are therefore smaller than conventional columns, and this will offer some compensation for the extra cost of the pulse and resin transfer system. Preliminary estimates indicate that for a slimy ore the extra cost will only be a fraction of the capital savings obtained in the pulp preparation stage. With a normal ore, the capital cost will be comparable with that of a conventional set of columns, because a smaller number of jigged-bed columns is required.

Power and maintenance costs will certainly be higher than for conventional ion-exchange, but not excessively so. Attrition of resin has been assumed, on the basis of pilot plant measurements, to be 100 per cent. per annum, based on the volume of resin undergoing pulsation, but because of the continuous movement of resin this volume is much less than the volume of resin in a conventional set. The overall cost of resin replacement is significant, but not all-important; on the basis of estimates for ore A a loss of the order of 600 per cent. per annum would have to be experienced to cancel the savings elsewhere, which are equivalent to the recovery of a further five per cent. of the uranium.

The extraction efficiency of the jigged-bed when combined with an efficient elution system can be assumed on the basis of pilot plant figures to be at least equal to the efficiency of conventional ion-exchange, but it is possible that the usual aspects of continuous countercurrent operation will enable savings to be made both in uranium recovery and eluant consumption. With a normal ore, the lower volume of solution to be treated means that a higher concentration of uranium is fed to the unit, and economies can be expected on this ground also.

In conclusion, it should be remarked that the cyclone and jigged-bed combination has shown great flexibility, and can readily be adapted to changes in ore character. This is one of the most attractive features of the process.

Acknowledgements

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The Wetting of Solid Metals by Liquid Metals

By D. O. Jordan and J. E. Lane*

The experimental results available indicate that the spreading of liquid metals on solid metal surfaces is aided by the formation of either intermetallic compounds, or solid solutions. It is, however, not a necessary condition, because liquid sodium is reported to be able to spread over both solid tantalum and niobium, without the formation of either an intermetallic compound or a solid solution. When intermetallic compounds or solid solutions are formed and spreading has occurred, it is possible that the spreading coefficient would be negative if no changes had occurred at the solid-liquid interface. In such cases, the validity of the use of spreading coefficients, and contact angles, to describe the conditions at the interface is doubtful. Surface-active agents are substances which alter conditions at phase interfaces. It has been shown that oxides and sulphides considerably reduce the spreading of liquid metals on solid metals. Fluxes, and many other materials, aid this spreading. These substances therefore can be classified as surface-active agents.

Introduction

The wetting of solid metals by liquid metals has been used as a technical process in soldering and brazing for many centuries. In recent years, interest in this and associated phenomena has increased considerably because of the use of liquid metals as coolants in nuclear reactors and to the possible use of liquid metal slurries in which the fissile material constitutes the solid phase (Taylor, 1955; Frost, 1956a, 1956b, 1957).

The authors are engaged in the study of the wetting of solid metals, uranium in particular, by liquid sodium. This review is intended to discuss the wetting of solid metals by liquid metals in general, but a greater emphasis will be placed on wetting by liquid sodium.

Definition of wetting

In technical processes, wetting has been defined (Moilliet and Collie, 1951) as any process in which one fluid is displaced completely, or in part, from the surface of a solid or liquid by another fluid phase. This definition is adequate for such processes as the wetting of textiles, flotation of minerals, and the manufacture of pigmented paints. However, liquid and solid metal surfaces in air are almost invariably coated with a film of oxide which will considerably modify the surface properties. If the solid metal is completely freed of the oxide film and degassed under vacuum, the wetting process cannot be described in terms of the above definition because there has not been a simultaneous displacement of one fluid by another. This difficulty may be overcome by defining wetting as any process during which a solid and a liquid come into contact and form a solid-liquid interface.

In spite of the criticism which has been made of the use of contact angles as a measure of the degree of wetting of solids by liquids (Osterhoff and Bartell, 1930; Adam, 1941), this method has been frequently used in the experimental measurement of the degree of wetting. A great deal of the confusion that has arisen in the literature concerning the use of contact angles may be overcome by proper definition. In this review, a contact angle of zero will indicate complete wetting, a finite contact angle will indicate partial wetting, and a contact angle of 180° will indicate that no wetting has occurred.

The surface free energy of liquid and solid metals

The surface free energy of liquid metals is high compared with those of organic liquids and water. There are very few metals with surface free energies less than 200 ergs/sq. cm., whereas a surface free energy of 50 ergs/sq. cm. is high for an organic liquid.

The theoretical approach to the calculation of surface free energies of metals has followed the relationships based on the free electron theory of metals (using the simple Sommerfeld theory) or relationships connecting the surface free energy with other properties of the metal. The latter relationships have been discussed by Hildebrand and Scott (1950) and by Bondi (1953). Notwithstanding the criticism of the free electron theory by Skapsi (1948), Taylor (1954) has calculated the surface free energy of liquid sodium using the free electron model relationships, but has obtained poor agreement with the experimental values.

The surface free energies of a number of liquid metals have been determined experimentally, and a large proportion of these results have been tabulated by Lyon (1952).

The surface free energies of a few solid metals have been measured in recent years. They have been obtained by either of two methods: firstly, the method using the dihedral angle theory proposed by Smith (1948) and secondly, a method utilizing the viscous creep in thin wires at temperatures just below the melting point of the metal. The results of some of these measurements are given in Table 1.

Several methods of calculating the surface free energy of solid metals have been developed, and these have been summarized by Taylor (1955). Unfortunately, these calculations have not been thoroughly tested owing to the lack of experimental data.

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TABLE 1.—THE MEASURED SURFACE FREE ENERGIES OF SOME SOLID METALS

Metal	Temperature range °C	Surface free energy ergs/sq.cm.	Reference
Au	700-800	1200	Tamman and Boehme, 1932.
Au	1017-1042	1400 ± 65	Buttner, Udin and Wulff, 1951.
Ag	875-932	1140 ± 90	Funk, Udin and Wulff, 1951.
Cu	1200-1300	1450	Udin, Shaler and Wulff, 1949.
Cu	800-900	1800	Bailey and Watkins, 1950.
Sn	215	685	Greenhill and McDonald, 1953.

The wetting of solid metals by liquid metals

A liquid metal will spread over a solid if the following inequality holds:

$$\gamma_{SV} > \gamma_{SL} + \gamma_{LV} \qquad (1)$$

where γ_{SV} is the surface free energy at the solid-vapour interface, γ_{SL}, that at the solid-liquid interface, and γ_{L}, that at the liquid-vapour interface. This inequality is often written in the form

$$S = \gamma_{SV} - \gamma_{SL} - \gamma_{LV} \qquad (2)$$

where S is the spreading coefficient, and spreading will only occur if S has a positive value.

Frost (1957) reports that equation (1) is satisfied in the systems solid tantalum or niobium and liquid sodium. The tantalum and niobium both have extremely high surface free energies (approximately 2000 ergs/sq. cm.), and the liquid sodium has a surface free energy of approximately 200 ergs/sq. cm. The interfacial free energy at the solid-liquid interface could thus be as high as 1500 ergs/sq. cm., and the inequality, (1), would still be satisfied.

An apparent deviation from the above relationship occurs with the system solid copper-liquid lead. The surface free energy of solid copper is, in all probability, much greater than 1,000 ergs/sq. cm., because the value for liquid copper exceeds 1,000. The surface free energy of liquid lead is approximately 450-500 ergs/sq. cm. These figures would suggest that a contact angle of zero for liquid lead in contact with solid copper would be expected, but this is not observed (Bailey and Watkins, 1950). Bailey and Watkins have measured the solid surface free energies of this system, together with the solid-liquid interfacial free energy using the method of dihedral angles (Smith, 1948). They obtain the following average values for the surface and interfacial free energies: clean copper (solid), 1,800 ergs/sq.cm.; between copper (solid) and lead (liquid), 340 ergs/sq.cm.; copper (solid) with absorbed lead, 780 ergs/sq.cm. If, therefore, clean copper were brought into contact with liquid lead, spreading should theoretically occur because the initial value of the spreading coefficient would be positive. However, such conditions are not experimentally realisable because lead vapour is rapidly adsorbed onto the copper surface. The considerable reduction in the surface free energy of the solid copper surface thereby produced is sufficient to produce a negative spreading coefficient.

In most systems in which the liquid metal has been observed to spread over a solid metal, i.e., forming zero contact angle, the above relationship does not operate because the two metals are capable of forming either intermetallic compounds or solid solutions (Daniels 1935; Tamman and Ruhenbeck 1935; Daniels and Macnaughton 1937; Bailey and Watkins 1951). Three different types of behaviour are described by Bailey and Watkins (1951), when a solid metal plate is immersed in a liquid metal, withdrawn and the excess liquid drained off by tilting the plate through a small angle. These are:

(a) The liquid metal forms a uniform coating on the solid metal,

(b) The liquid metal film collapses on draining, the liquid forming drops which adhere to the surface of the solid.

(c) No liquid adheres to the solid metal surface either as a film or as drops. These groups were classified by Bailey and Watkins as (a) wetting, (b) de-wetting, and (c) no-wetting.

In the investigation of 38 pairs of pure metals chosen from the following: Solid metal, iron, nickel, copper, gold, silver; liquid metal, silver, antimony, tellurium, zinc, lead, cadmium, bismuth, aluminium, tin. They found that a uniform film was formed with the 25 pairs that could form intermetallic compounds under the experimental conditions, and with the seven pairs that were capable of forming solid solutions. Of the remaining pairs which formed neither intermetallic compounds nor solid solutions, three formed collapsed films and three showed no adhesion at all. In a more rigorous test on selected pairs of metals, the metal plates were dipped vertically into the liquid metal and withdrawn vertically, so that the draining angle was 90°. The pairs which could form solid solutions had more stable coatings than those which formed intermetallic compounds.

In the more recent and more quantitative studies of Addison, Kerridge and Lewis (1954), the contact angles between zinc, copper, and molybdenum and liquid sodium have been measured using the Wilhemy plate technique. With this method it is possible to measure the contact angle if the surface tension of the liquid is known. With zinc plates, and assuming a contact angle of zero, the value of the surface tension of liquid sodium was found to be 195 dynes/cm. This value is in good agreement with the value of 191 dynes/cm. obtained by the maximum bubble pressure method (Taylor 1954) and that of 202 dynes/cm. obtained by the drop weight method (Addison et al. 1955). With copper and molybdenum plates, a comparable value of the surface tension of liquid sodium could only be obtained if it were assumed that the contact angle was 180°, i.e., that conditions of non-wetting prevailed. These data support the view that the formation of intermetallic compounds or solid solutions at the interface produce wetting, since zinc is known to form at least one intermetallic compound with sodium, whereas copper and molybdenum apparently do not.

Although the formation of intermetallic compounds and solid solutions frequently produces initial spreading of the liquid metal over the solid, should the intermetallic compound or solid solution be a solid at the temperature of the experiment, and if its solid surface free energy be less than that of the parent solid, the value of the final spreading coefficient may become negative. The liquid film will then coalesce into droplets and de-wetting occurs (Milner 1957).

The effect of added substances on surface free energy and wetting

The addition of oxides, sulphides, other metals and fluxes to the solid metal-liquid metal systems alters the conditions at the interface and these substances can therefore be correctly classed as surface-active agents.

Oxides

The surface free energy of most metal oxides is lower than that of the corresponding metal (Bondi, 1953). Metals which are contaminated with an oxide surface will therefore not be wetted as easily as metals with clean surfaces. Addison et al. (1956) found that zinc plates, with a heavy coating of zinc oxide, had initial contact processes, and are of such varied composition that different modes of action must be operative. Dean (1924) has suggested that chloride fluxes owe their efficiency to the evolution of HCl, which removes the oxide film from the solid surface. If iron is galvanized with zinc, a zinc-ammonium chloride flux may be used. It has been suggested (Daniels, 1932) that the oxygen atoms on the iron surface are replaced by chloride atoms. When the fluxed surface is dipped into the molten zinc, the chlorine is removed as zinc chloride, and the metallic zinc then wets the steel surface. In these suggested mechanisms of fluxing, the definition of wetting as replacement of one fluid by another is valid. Coffman and Parr (1927) have shown that dry HCl gas will reduce the surface tension of solder. Tasman and Arntz (1930) showed that the spreading of mercury on silver, gold, copper and zinc was quickened if the spreading was carried out under dilute sulphuric or hydrochloric acid.

More recently it has been suggested that an electromechanical effect aids the action of fluxes (Latin, 1938; Bailey and Watkins, 1951). It is suggested that if a zinc-ammonium chloride flux is used with solder on copper sheet, the tin will be taken into solution in the flux, and electrolytically deposited on to the copper sheet, thus aiding the wetting of the copper sheet by the solder. The open circuit voltages of this type of cell, using different metal chloride fluxes, have been measured by Bailey and Watkins (1951).

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Minerals in New South Wales for Atomic Energy

By E. O. Rayner*

The geological nature, extent and future prospects are outlined for those minerals occurring in New South Wales which may have application in atomic energy developments. Uranium and thorium-bearing deposits as possible sources of fuel materials are discussed, and reference also made to availability of minerals of zirconium, beryllium and other metals which may be used as sheathing, moderator, heat transfer or constructional materials for nuclear reactors.

Uranium mineralisation is divided into groups and the preferred mineral, rock and structural associations are indicated. The uranium deposits have not been proved of immediate economic importance, and ore reserves would not total more than several hundred tons, but further testing is warranted.

Zircon and monazite, as sources of zirconium and thorium respectively, occur in significant amount in heavy-mineral beach sand deposits of the N.S.W. coast and adjoining Southern Queensland coast. Proved and probable reserves total 3,000,000 tons of zircon and 34,000 tons of monazite; of each total approximately one-third is in N.S.W. In addition, large possible reserves may exist in areas as yet incompletely tested. The nature and origin of these deposits is reviewed.

Beryl, source mineral for beryllium, is present in pegmatites of the Broken Hill district and in lode formations of New England. Small tonnages of beryl will continue to be won as a by-product in felspar mining at Broken Hill. Short notes are added on other minerals which might become important in nuclear energy.

Introduction

The growth of atomic energy projects has created new uses for a number of metals and minerals, and has imposed upon States and nations the need to search for and take stock of their resources of these substances.

This paper seeks to outline the nature, extent and geological setting of all those minerals in New South Wales which have, or may acquire, an application in atomic energy developments.

Uranium

Uranium-bearing rocks in New South Wales may be described as occurring in three groups which are geologically and geographically distinct:

(i) Precambrian—Broken Hill district (Far Western N.S.W.).

(ii) Lower to Middle Palaeozoic (epi-Ordovician to late Lower Carboniferous)—Central and Southern N.S.W. (e.g., Blackfellows Dam, Carcoar and Whipstick).

(iii) Upper Palaeozoic (Permian)—New England district (North-eastern N.S.W.).

In the Broken Hill Precambrian region the uranium-bearing minerals are found chiefly in rocks of the Willyama Complex (Archaean), the age of mineralisation being tentatively regarded as late-middle Precambrian (Houghtonian). Primary davidite occurs in the Thackaringa area in pegmatites, aplites, and microgranites, and as hydrothermal, high temperature deposits in shear zones (Rayner, 1955, 1956). The davidite (iron-uranium titanate with rare earths) is in metamict form in association with rutile, hematite, ilmenite, magnetite and biotite. The rock and mineral assemblages closely resemble those of the davidite-producing field of Radium Hill in South Australia, though there the host structures of the fissure deposits are more regular and persistent. Selected davidite from Thackaringa contains approximately 4 per cent. U_3O_8, but small bunches of ore taken in bulk and consisting of the intergrowth referred to above, assay 0.25 per cent. U_3O_8 and less. On present information the occurrences are small and scattered, but the Thackaringa area might prove capable of producing several hundred tons of low-grade davidite ore by selective small-scale mining and the raw product or a concentrate may be amenable for treatment at Radium Hill.

Other occurrences in the Broken Hill field, chiefly of secondary uranium minerals, are in shear zones (e.g., Eldoo Creek) or associated with copper (e.g., Copper Blow) and silver-lead lodes (e.g., Great Western and Hen-and-Chickens).

The Broken Hill Precambrian field lies between and may be correlated with those of South Australia and Cloncurry-Mount Isa (Queensland). These regions form a common metallogenetic province in which uranium is associated with such elements as titanium, iron, thorium, rare earths, lithium, beryllium and fluorine. Economic metallic associates include copper, cobalt and silver-lead-zinc. The province may also extend to the Northern Territory.

The Broken Hill district is part of an arcuate belt, through east-central Australia, which contains the most significant uranium deposits of the continent. This belt—which for descriptive purposes might be termed the Australian Uranic Arc — includes the uranium-bearing Precambrian terrains of South Australia (e.g., Radium Hill, Crocker Well and Mount Painter); New South Wales (Broken Hill); Queensland (e.g., Mary Kathleen); and the Northern Ter-

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* Geological Survey of New South Wales. Manuscript received March 13, 1958.

ritory (e.g., Rum Jungle and South Alligator River). Broadly, this belt lies near the eastern rim of the ancient Precambrian shields of western and central-northern Australia, and corresponds with the western fringes or hinge lines of the main shelf zone through east-central Australia.

The major uranium deposits (and some other large base-metal lodes) occur not within the stable cratonic zones, but in the relatively unstable, more mobile, belts which lie off the shield or in intercratonic geosynclines. These favourable zones, which have suffered a considerable degree of folding and faulting, also feature abnormally high thermal gradients, as at Broken Hill and Mount Isa, and may well represent belts of relative crustal thinness. Within the large stable cratonic zones which form a large proportion of the western part of Australia there is less indication, and perhaps less likelihood, of large-scale uranium mineralisation.

The uranium-bearing deposits of the second and third groups referred to earlier—namely Palaeozoic, in a range from epi-Ordovician to late Permian—are associated with several stages in the growth of the Tasman Geosyncline. This complex mobile belt of deposition has undergone a number of periods of orogenic movement, igneous intrusion and ore deposition.

At Blackfellows Dam, near Nymagee in central N.S.W., uranium minerals are present in a small fissure lode within the Erimeran granite. The secondary uranium minerals, meta-torbernite, meta-autunite, and curite, are associated with copper, lead and zinc minerals. Assays from a short length of this secondary zone range from 0.18 to 1.87 per cent. U_3O_8 across widths of up to 30 inches. In addition, nodules of pyrolusite in a clay matrix on the footwall have been found to have an absorbed outer layer of very highly radioactive radium and daughter products. As yet there has been no testing of the primary zone, but the nature of the occurrence does not preclude the possibility of some pitchblende or uraninite in depth.

At Carcoar, 130 miles west of Sydney, primary and secondary uranium minerals occur with cobalt, molybdenum, and copper deposits. The uranium-bearing lodes are on the flank of a body of hornblende diorite, and are localised in shears within intruded slates and andesites and at the contact between these rocks and the diorite. The diorite is in turn intruded by a granite batholith which crops out half a mile from the mineral deposits. Small uraninite crystals are disseminated in cobaltite and in molybdenite. Radiometric grid surveys reveal that the radioactivity is mainly limited to the old cobalt workings, except for several adjacent anomalies where subsequent testing has uncovered secondary uranium minerals. The known cobalt leases have been worked out, and there are virtually no proven uranium reserves. Recent geophysical surveys by the Bureau of Mineral Resources have revealed some strong linear anomalies which are worthy of testing.

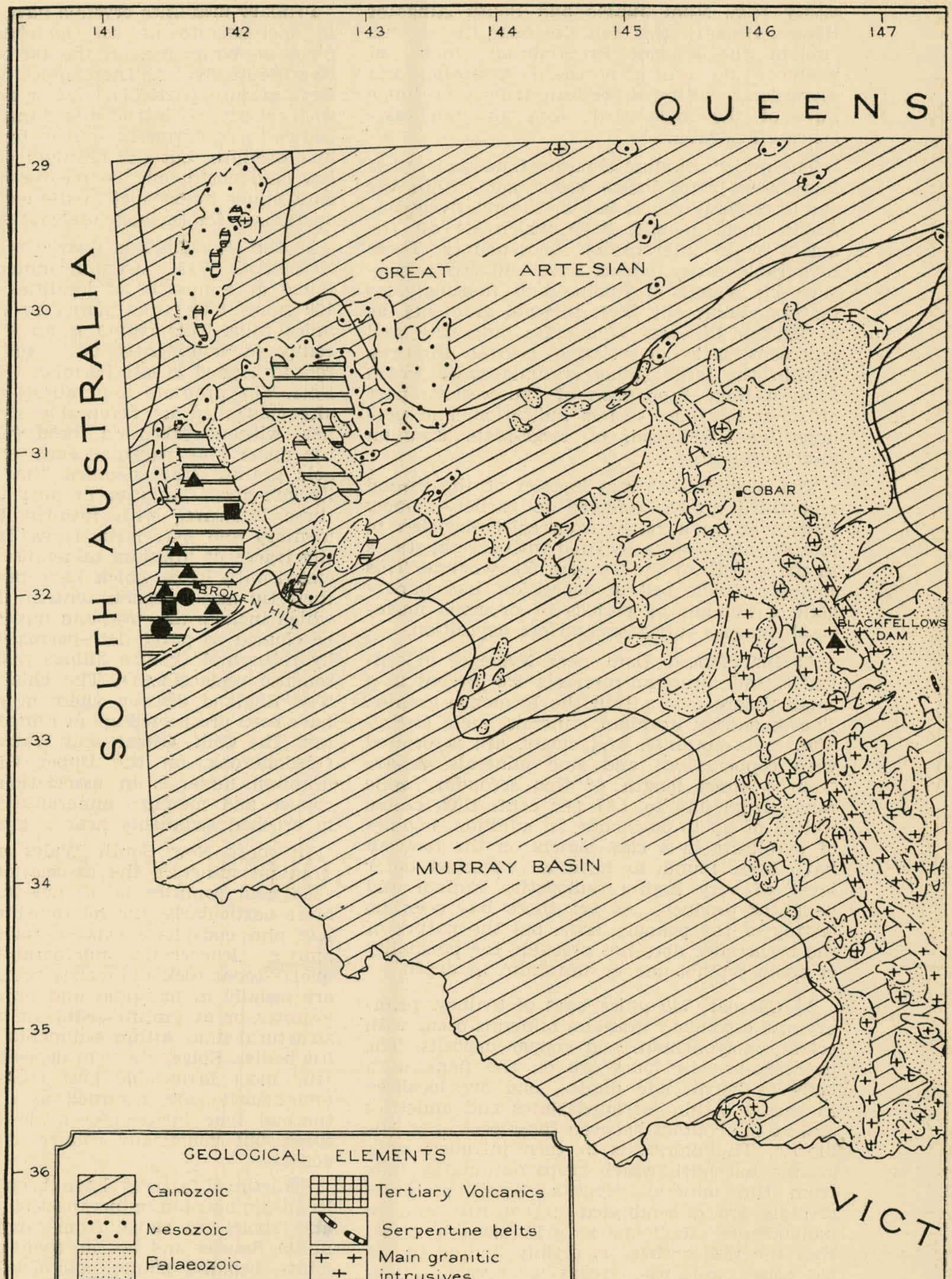
Primary uraninite crystals have been detected in molybdenite of the molybdenum-bismuth pipes at Whipstick, on the far south coast of New South Wales. These pipes are within the Bega granite, particularly at or near its contact with overlying intruded sediments. The orebodies were formerly mined for molybdenum and bismuth, but the uranium content is very low, and could only be recovered economically as a minor by-product in the event of renewed operations for molybdenite and bismuth.

In the New England district of north-eastern New South Wales, uranium minerals have been found at a number of localities within a belt 150 miles long from north to south and 100 miles wide. This region is an elevated orogen composed dominantly of a suite of granitic rocks intruded in late Permian (Hunter-Bowen) time. The uranium mineralisation, though locally controlled by favourable structural traps, has, without exception, been introduced into the upper and marginal zones of the youngest acid granite (the so-called "tin" granite) and its late-phase variants, or near the contact of these intrusives with invaded Permian sedimentary and pyroclastic rocks. In many cases, the traces of uranium mineralisation are associated with lodes which have been worked for their tin and tungsten content. The ore-bodies which include this uranium mineralisation may be classed as partly late-pegmatitic and partly hydrothermal (fissure fillings with some metasomatic replacement). The chief areas of the New England district where uranium minerals have been prospected are at Emmaville, Torrington, The Gulf, Gilgai, and Watson's Creek. At Gordonbrook, on the Upper Clarence River, uranium minerals in association with cobalt, copper and mercury mineralisation are found in crushed sediments near a granite contact.

In most New South Wales occurrences of uranium minerals the association of uranium with acid granites is a noteworthy feature. More particularly, the relationship is with such late and end-phase expressions as pegmatites, aplites, leucocratic micrograniites, greisens, quartz-topaz rock and quartz veins. The deposits are usually in marginal and upper portions of granites or at granite-sediment contacts, or in structural lines within sediments close to granitic bodies. Epigenetic vein deposits in sediments (the most favourable host rocks being slates and schists) are regarded as a later, hydrothermal type introduced—at lowered temperatures, but genetically connected with granitic activity.

Structural features have been important in localising uranium mineralisation. These favourable structures range from large shear zones, faults, fissures and breccia zones down to minor joints, bedding planes, microfractures and interstitial openings.

The chief metallic associates of the uranium mineralisation are copper, cobalt and molybdenum, and to a lesser extent bismuth, tungsten, tin, and silver-lead-zinc. There is no notable association of uranium with the numerous gold, gold-antimony and major copper-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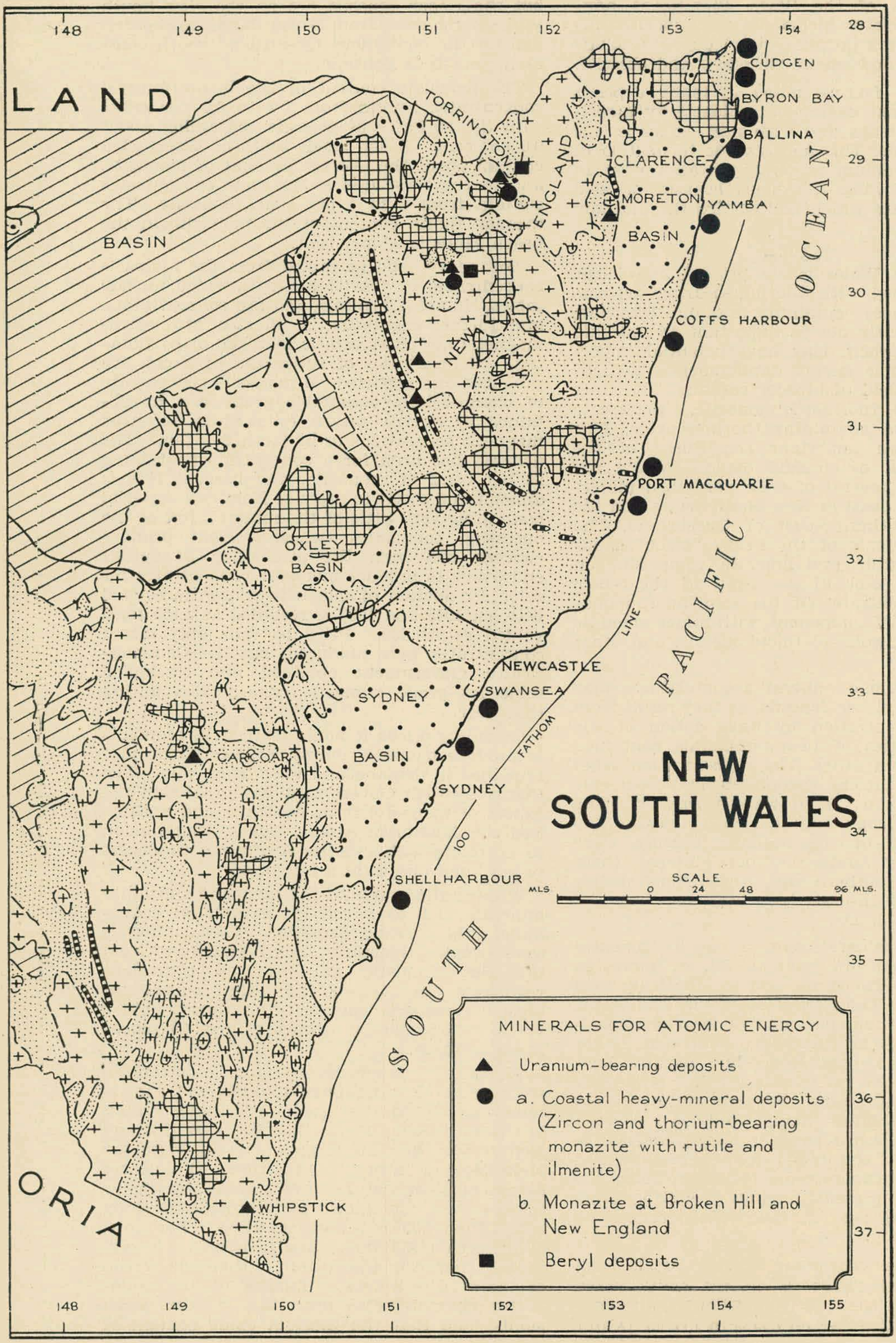
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PORT MACQUARIE
OXLEY
BASIN
PACIFIC
LINE
FATHOM
100
NEWCASTLE
SWANSEA
SYDNEY
BASIN
SYDNEY
CARCOAR
SHELLHARBOUR
SOUTH
NEW SOUTH WALES
SCALE
MLS 0 24 48 96 MLS.
ORIA
WHIPSTICK
MINERALS FOR ATOMIC ENERGY
▲ Uranium-bearing deposits
● a. Coastal heavy-mineral deposits (Zircon and thorium-bearing monazite with rutile and ilmenite)
b. Monazite at Broken Hill and New England
■ Beryl deposits
148
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gold deposits of the State. Significant non-metallic associates include biotite, fluorite (including the dark purple variety) quartz (including the blue and smoky varieties) and chlorite.

A common feature noted in many deposits is an abnormal degree of radioactivity associated with gossans and with iron oxides in the secondary zone. This appears due to fixation of secondary uranium minerals by absorption in goethite, following the neutralisation of acid sulphate waters which had carried the uranium in solution.

Thorium

New South Wales is, on present knowledge, better endowed with thorium-bearing deposits than with those containing uranium. Thorium, though currently not as important as uranium as a fuel element, may have potential importance in atomic energy developments, particularly in the field of breeder reactors. The chief ore mineral of thorium is monazite, a phosphate of rare earths containing thorium oxide. This mineral is an important constituent of the heavy-mineral beach-sand deposits which are exploited for several of their contained minerals along the coast of New South Wales and the adjoining southern coast of Queensland. The main constituents of the heavy "black" sands are zircon, rutile, and ilmenite, comprising on the average some 97 per cent. of the whole natural concentrate. Of the remainder, monazite is the chief component, with minor amounts of cassiterite, gold, platinoid metals, and other minerals.

These economic mineral accumulations may be classed as placer deposits, as they result from natural concentration by wave action of the small quantities of heavy minerals contained in ordinary sea sands. The concentration takes place chiefly during cyclonic storms, when gale force winds cause high wave turbulence. The deposits are commonly enriched towards the northern end of the beaches, because wave action during storms is more intense there than at the southern end, where some shelter from the south-easterly winds is given by southern headlands.

The heavy-mineral seams range in thickness from less than an inch to 6ft. or more, in width from 5-100 yards, and in length up to a mile or more. The richest accumulations so far exploited are on present-day beaches which have been cut into and reconcentrated. Other deposits, some very large but usually low-grade, are found on former beaches landwards of the present strand, beneath dunes and in emerged heathlands. Some windblown concentrations have been formed on later transgressive dunes. Some of the inland deposits are in raised beaches and terraces some 10-15 feet above the level of the present beaches, resulting from coastal emergence in late Pleistocene and Recent times.

The deposits occur along the New South Wales coast from Shellharbour on the south coast, northwards to the Queensland border, and continue on the Queensland coast to Fraser Island, but the richer deposits are on the New South Wales north coast from Byron Bay to the border, and on the contiguous Queensland south coast as far north as Southport.

The monazite content of the heavy mineral concentrates ranges from 0.2 to 4.0 per cent., with the majority of deposits containing about 0.6 per cent. Beach sands south of Woolgoolga contain about the average amount of monazite, and in the Newcastle sector some concentrates contain from 2 to 4 per cent. monazite. The average thoria content of the monazite in the beach sands is 6.6 per cent.

The heavy sand as mined from the seams is concentrated by gravity methods, which include tables, Humphrey spirals, and fan concentrators, and further separation is effected by electromagnetic and electrostatic methods or by flotation. Concentrates of rutile and of zircon are produced and exported. A monazite concentrate of commercially acceptable grade can be obtained by gravity means followed by repeated magnetic separations or by froth flotation. The demand has been for a concentrate containing 95 per cent. of monazite, and because this is difficult and costly to produce from raw material containing very little of the mineral, few of the operating companies (whose current primary concern has been the production of rutile) endeavour to produce monazite concentrates for sale. Production of monazite concentrates sold between 1948 and 1956 is recorded as 2,105 tons, valued at £81,639. Prior to 1948, only mixed concentrates were produced, but the Commonwealth Government obtained and stockpiled some low-grade monazite concentrates during that year.

On ruling prices, it would scarcely be economic to produce monazite unless rutile or zircon also remained in demand and were being won. The Commonwealth Government has prohibited the export of monazite from Australia and authorised the Australian Atomic Energy Commission to negotiate with existing Australian producers to buy their output at a fair price.

What is the source of the monazite and other minerals such as zircon in the detrital heavy-mineral beach-sand deposits? These minerals were formerly thought to have been derived from the New England granites and equivalent intrusives in Queensland, the subsequent erosion-transport-deposition cycles being chiefly via the Mesozoic sandstones of the Clarence-Moreton basin.

The sedimentary rocks of this basin do contain these minerals, and have undoubtedly contributed largely to the beach sand concentrates, but there is some doubt that the New England granite was the main original source. First, the New England batholith is restricted in exposure to a length of 250 miles, whereas the heavy minerals are now known to form deposits along more than 700 miles of coastline. Secondly, Whitworth (1956) has stated that rutile is rare in the New England granite; that the zircon of the granite differs in fluorescent and radioactive characteristics from that of the beach sands; and that the thorium oxide content of monazites from New England ranges from 0.1 to 1 per cent., but the beach monazites commonly contain 6 to 7 per cent. thorium oxide. Whitworth has advanced the view that the rutile, zircon and monazite have come originally from the Precambrian crystalline gneisses and schists, and from some of the older granites, by way of successive sedimentary depositions and erosions through subsequent geological ages. A large proportion of the detrital heavy-mineral derivatives from the hinterland formations, on reaching the sea, became concentrated in beach deposits, the particular richness of accumulations on north coast beaches being due to a combination of favourable geological, physiographic and climatic factors.

In addition to the coastal placers, monazite is found in New South Wales in a number of rock formations, and in the Broken Hill and New England fields it occurs in lode formations and in alluvial accumulations along watercourses. In the Precambrian Barrier Ranges around Broken Hill, the monazite is in biotite-quartz gneisses and pegmatites. For example, west of Sentinel Peak, 25 miles south-west of Broken Hill, monazite occurrences are sufficiently concentrated in zones of a biotite-quartz gneiss to be outlined by radioactive anomalies. The total content of thorium is not high, but a monazite concentrate can be readily obtained (6oz. of heavy concentrate panned from 6lb. of rock consisted mainly of monazite and assayed 1.8 per cent. thorium oxide). Monazite is also disseminated in pegmatitic environments at Egebek, and between Broken Hill and Thackaringa.

In the New England district, monazite is an accessory in quartz-wolfram veins and greisens connected with late-Permian granite, and is a common detrital mineral in stream beds. In the Torrington area it is found in lodes at Cow Flat, Dingo Creek, Blatherarm and The Gulf, but the thorium content is usually less than 1 per cent. In alluvial and eluvial tin-mining operations monazite containing approximately 1 per cent. thorium oxide is found in many of the cassiterite concentrates, and constitutes an impurity which has to be removed prior to sale.

Zirconium

Sheathing metals for atomic fuels in reactors require properties of strength, inertness to the fuel material and the coolant, generally a low neutron-absorption factor, and resistance to heat and corrosion (Myers, 1954). Such metals as zirconium, beryllium and niobium have these characteristics. For very high temperature reactors, the oxides and carbides of zirconium and beryllium may have application. Zirconium, apart from its potential in atomic energy projects, has important uses in the ceramic, refractory, electrical, and steel industries.

New South Wales has considerable reserves of zircon, the principal source mineral of zirconium, in the heavy mineral beach-sand deposits which have been described above. The east coast of Australia (New South Wales and Queensland) has been the world's chief supplier of this mineral. Zircon ($ZrO_2.SiO_2$—containing 67.2 per cent. ZrO_2) is the most abundant mineral of the natural concentrate, comprising usually 20 to 50 per cent. of the total heavy minerals. In the case of some material near Clarence River, it is up to 80 per cent. of the total. In the early exploitation of the beach deposits from 1933 onwards, zircon was the chief mineral sought from the sands, but in recent years, with the growth of the titanium metal industry, the chief constitutent exported has been rutile.

The recorded New South Wales production of zircon from 1934 to 1956 exceeds 200,000 tons. In 1956, the last year for which complete figures are available, production was 50,660 tons, valued at £406,313. Not all the zircon won has been exported, and some reserves of zircon concentrates are stockpiled by the producers. Some of these stocks are rough concentrates which will require re-cleaning to upgrade them for eventual sale.

Zirconium usually has associated with it a small percentage of hafnium, and for atomic energy purposes it is essential that the hafnium (which has a high neutron-absorption factor) be removed from the zirconium.

Reference to the nature of the heavy-mineral sand deposits which contain the zircon, and to the ultimate source of the zircon, has been made above in connection with the thorium-bearing component, monazite. Zircon is mainly contributed to the beach sands from the coastal sedimentary rocks, especially sandstones, of the Clarence-Moreton and Sydney basins. In the former basin the zircon is in Triassic and Jurassic strata; in the latter it is distributed through Permian and Triassic sequences. It may be noted that the heavy minerals which are found in the Permian coal measures could not have been derived from the late-Permian New England granites.

Further inland, zircon is a common accessory mineral in most granites; is abundant in creek sands of New England; is present in small percentages in Jurassic and Cretaceous beds of the Great Artesian Basin; is relatively abundant in Devonian quartzites such as those of the Cobar district; and is very common accessory in the Precambrian rocks of the Broken Hill field.

Beryllium

Beryllium metal has an application in reactor developments, both as a sheathing material and as a moderator for slowing down neutrons. Beryllium has other important uses, particularly in special alloys, and is also reported to have been under test for possible use as a rocket fuel.

The chief source mineral of beryllium is beryl, a silicate of beryllium and aluminium. Beryl in commercial quantities is commonly restricted to coarse pegmatites. Distribution within the pegmatites is usually so erratic or sparse that prices which have ruled for beryl ore would rarely repay cost of mining for beryl alone, and the mineral has mostly been won as a by-product in open-cut mining for felspar and mica. Recent advances in flotation technique to recover beryl may result in the exploitation of lower-grade deposits.

Beryl in commercial quantity has been won from the Broken Hill field, chiefly as a hand-picked by-product in the production of potash felspar. In the period 1944-1956, a total of 82 tons of beryl, valued at £7,440, was produced from this district.

In the Broken Hill field the chief centre of production has been at Egebek, 23 miles south-west of Broken Hill. The beryl occurs in pegmatites at the Triple Chance, Lady Beryl, and adjacent felspar quarries. The pegmatites of economic interest are of coarse texture and composed of felspar (microcline and orthoclase, with perthitic intergrowth with albite), quartz, muscovite and beryl, with rarer tourmaline, monazite, rutile, apatite and tentalite (Rayner and Hall, 1949). The host rocks to these coarse transgressive pegmatites are micasillimanite-garnet gneisses, mica schists, aplites, fine-grained concordant pegmatites and amphibolites of the Willyama Complex (Achaean). The Willyama suite is folded dominantly on north-south axes, but the mineralised pegmatites are dyke-like and irregular masses which, together with several dolerite dykes, are set in transverse tensional zones of weakness. These loci are associated with late cross-folding and shearing movements in east-west directions across the earlier main folding.

Beryl in the Egebek area is in well-defined hexagonal crystals which range from less than an inch in length up to individual crystals which are among the largest in the world. One crystal in the Lady Beryl deposit was over 9ft. in length and nearly 3ft. across. Beryl is also present in several pegmatites close to the city of Broken Hill, and a single crystal 6ft. in length was won from one of these localities.

At Euriowie, 50 miles north of Broken Hill, the rarer caesium-bearing variety of beryl known as vorobyevite occurs in pegmatites within Willyama rocks of the Euriowie inlier. These pegmatites also contain cassiterite and the lithium-bearing mineral amblygonite, with quartz and minor amounts of pinite, muscovite, felspar, augellite, apatite, and paragonite. The chief occurrence of the vorobyevite is in the Trident pegmatite. The mineral is white and massive, resembling the caesium beryl from Wodgina (W.A.) known as rosterite; it lacks the defined hexagonal form of the beryl crystals from such localities as Egebek and Broken Hill, and is difficult to pick clean from the associated quartz and amblygonite. Thus, though selected vorobyevite assays 11.5 per cent. BeO (with 1.3 ± 0.3 per cent. caesium oxide), a small amount of ore shipped from the deposit has fallen below the acceptable BeO content of 10 per cent. It is probable that insufficient beryl exists to warrant extraction at present prices, other than in conjunction with tin or amblygonite mining (Hall and Lloyd, 1953).

It has been stated by Mawby (ibid) that at Euriowie the original quartz-orthoclase pegmatites suffered greisenisation and then alkali-rich injection, resulting in polyphase pegmatites characterised by the alteration of felspar to cleavelandite and the formation of white beryl, amblygonite and associated rare minerals.

Although the Broken Hill Precambrian field is the only one in New South Wales which has produced a commercial amount of beryl, small quantities of that mineral have been recorded at a number of localities in the New England region but none of these occurrences can be regarded at present as an economic source of beryllium. In the past, several localities have produced a little beryl of gemstone quality (emerald and aquamarine).

The New England beryl occurs chiefly as an accessory mineral in tin and tungsten-bearing lodes which are within late-Permian granite or near granite-sediment contacts. The general environment is that already referred to in describing uranium mineralisation in New England. Additionally, some beryl is found in alluvial tin deposits in parts of this district. The chief centres of beryl occurrence are Emmaville, Torrington, The Gulf, Elsmore and Tingha. At Blatherarm, three miles north of Torrington, beryl crystals and bromellite occur in pegmatitic and greisenous lode material with wolframite, biotite, monazite and secondary uranium minerals. Probably several tons of beryllium ore could be won from this deposit, but the texture is too fine-grained to allow hand-picking.

Other materials

Niobium has been suggested as a good sheathing material, particularly for fast reactors. It has a high melting point and, combined with good high temperature characteristics, it has reasonable nuclear and fabricating properties. Main sources of niobium are the minerals tantalite and columbite, and other minerals of the isomorphous series of which they are members. Reserves of these minerals in New South Wales may be regarded as negligible, the known occurrences being sparse distributions in pegmatites of the Broken Hill field. Tantalite has been reported from Egebek and Euriowie; tantalite and columbite from three miles north-west of Broken Hill; and microlite (tantalate and columbate of calcium) from Euriowie and Thompson's Siding.

Among heat transfer (coolant) materials which may be suitable for reactors are the molten metals sodium, bismuth, lead and lead-bismuth eutectic. Source salts for the preparation of metallic sodium, particularly sodium chloride, are abundantly available in Australia, but New South Wales is the least developed of all the mainland States in the production of salt from solar evaporating pans and dry salt lakes.

Bismuth minerals are present at a number of New South Wales localities, and have been mined mainly for medicinal use. Known reserves are mostly exhausted, the mines are dormant, and Australian supply is inadequate to meet all domestic demands. The bismuth mineralisation is commonly associated with molybdenite in pipe deposits in granite and at granite-sediment contacts. Lodes of this type occur at Whipstick on the south Coast, which was the chief producer, and in the New England district at Kingsgate, Deepwater and Torrington. In New England, bismuth has also been produced as a by-product in tin and tungsten mining. At Duckmaloi in the central highlands, pyrometasomatic bismuth lodes are developed at a granite limestone contact.

The known New South Wales production to date of bismuth concentrates (including bismuth-wolfram concentrates) amounts to 925 tons, valued at £268,980. Reserves are difficult to forecast because of the irregular and pipe-like nature of the lodes.

Bismuth minerals are present in subordinate quantities in the large tonnages of copper and gold ores of the Cobar field in central New South Wales. For example, cyanide residues from the New Occidental gold mine are reported to contain 0.12 per cent. bismuth, but experiments to recover a saleable product were unsuccessful. However, if large-scale mining is resumed at Cobar the problem of bismuth recovery as a by-product is likely to receive attention.

With reference to lead as a possible molten metal heat transfer material, Australian supplies are very considerable, and in New South Wales the chief source of production of lead is the great Broken Hill line of lode.

Graphite is used as a moderator in atomic reactors. Natural graphite is present in New South Wales at Undercliffe Mountain, Winterbourne Station and South Komungla, and production (chiefly from the first-mentioned locality) has totalled 2,687 tons. However, because the manufactured or artificial graphite is usually used in atomic reactors, details of the natural occurrences will not be elaborated.

Reserves and future prospects

The uranium deposits of New South Wales have not been proved of immediate economic significance, and proved or probable reserves of ore containing more than 0.1 per cent. U_3O_8 would not exceed several hundred tons. However, should the ratio of demand to supply increase substantially—as may happen within a decade with the growth of atomic power plants, or in national emergency — several of the known occurrences may become the focal point of renewed search. In the meantime, such promising fields as the Precambrian terrain of the Broken Hill district have not yet been exhaustively prospected.

Reserves of thorium contained in the monazite of heavy-mineral beach-sand deposits along the coast of New South Wales are considerable. Webb (1957), substantially in agreement with figures quoted by Gardner (1955), quotes the monazite reserves in beach and dune deposits of the New South Wales and Queensland coasts as 16,000 tons in high-grade deposits (+10 per cent. of heavy minerals) with 18,000 tons in lower grade deposits (—10 per cent. heavy minerals), the total quantity of thorium available, therefore, being about 2,000 tons. About 11,000 tons of these combined monazite reserves are in the New South Wales section. In addition, possible reserves of monazite may be substantial in incompletely tested "back" areas, including low-grade (less than 4 per cent. heavy mineral) localities. Small tonnages may also be recoverable from the Broken Hill and New England districts.

Zircon, as a source of zirconium, is abundantly present in the same monazite-bearing heavy mineral deposits of the coastal belt with large amounts of the titanium minerals rutile and ilmenite. On the New South Wales coast about 1,000,000 tons of zircon have been proved, and twice this amount is known in the contiguous southern Queensland coast, including large low-grade reserves in dune deposits on Stradbroke Island. To the New South Wales figure may be added large possible reserves in low-grade areas landwards of the beaches and not yet systematically tested. There are a number of companies engaged in beach and dune mining in New South Wales and Queensland, forming an important and well-established industry.

A small tonnage of beryl, the chief ore mineral of beryllium, continues to be won from the Broken Hill field. The beryl is mainly recovered as a by-product in the quarrying of felspar and reserves are difficult to estimate. It is likely that this field can supply only small tonnages of beryl, and the New England district may also contribute to a minor extent.

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Features of the Development of Chemical Processing for Mary Kathleen Ores

By R. A. Couche and F. R. Hartley*

The development of a suitable process for the recovery of uranium from Mary Kathleen ores is reviewed. Leaching under constant pH conditions was established as the most satisfactory procedure because acid consumption was reduced to a minimum and more selective leaching of the uranium was achieved. Accurate control of acidity was essential. Satisfactory leaching efficiencies were obtained by leaching at a constant pH of 1.7 for eight hours at 35°-40°C. Several other features of the plant are discussed, including the C.C.D. system for solid-liquid separation, and the anion exchange equipment for recovery of the uranium from the process liquors.

Introduction

During 1954 a large uranium bearing ore body, now known as the Mary Kathleen mine, was discovered in the Mount Isa-Cloncurry region of north-west Queensland.

The ore contains three major components—allanite, almandite garnet, and apatite. Subordinate components comprise uraninite, sulphides, diopside, hornblende, scapolite, felspar, calcite and stillwellite.

Exploitation depended upon the development of a suitable process for the economic recovery of uranium. The Research and Development Branch of the South Australian Department of Mines was commissioned in 1955 by the controlling company, the Rio Tinto Mining Company of Australia Pty. Ltd., for this investigation and subsequent development of the process.

Many samples were used during development studies. No sample was similar to the grade of ore expected from mining operations, but the general mineral characteristics were representative of the mine. Details have been reported by the Department of Mines (1956-1958).

Certain features of the development, some unique, are described in this paper.

Milling

The uranium-bearing minerals, principally uraninite, are finely divided and frequently encased in amorphous silica or apatite haloes or shells. When combined with the general characteristics of a hard, high density ore, poor leaching of coarse particles was expected. This was confirmed experimentally, and typical results are shown in Table 1.

To obtain consistent leaching with high recovery, it was necessary to grind the ore to approximately 50 per cent. minus 200 mesh with a minimum of plus 65 mesh. The close sizing thus specified proved too selective for a normal grinding circuit, which produced approximately 20 per cent. plus 65 mesh for a 50 per cent. minus 200 mesh grind. It was necessary to accept finer grinding of approximately 65-70 per cent. minus 200 mesh in order to reduce the plus 65 mesh fraction to less than 2-3 per cent.

TABLE 1.—LEACHING EFFICIENCY FOR VARIOUS PARTICLE SIZES.

Leaching time (hr.)		3	5	7	10
Particle size (Tyler mesh)		Leaching efficiency (per cent.)			
	+ 48	61	71	82	87
— 48	+ 65	77	83	87	91
— 65	+100	75	83	90	95
—100	+150	72	89	93	94
—150	+200	82	93	94	97
—200		85	94	96	97

Finer grinding proved an advantage, because high density ore produced a pulp or slurry of very low apparent viscosity and at 60 per cent. solids the pulp settled rapidly. When the pulp contained only 50 per cent. minus 200 mesh material, it was difficult to maintain the solids in uniform suspension. The hazard of pipeline blockage when pumping the pulp also was reduced considerably by the finer grind.

The grinding circuit for Mary Kathleen will comprise a rod mill in open circuit, with the discharge feeding two ball mills operating in closed circuit with hydraulic cyclone classification. This circuit is expected to give a degree of selectivity better than a normal grinding circuit.

Leaching

Initial experiments

Because of the nature of the ore components, a considerable acid consumption, with high phosphate concentration, was expected in the liquor if acid leaching was used. Consequently, a sample was leached using alkaline conditions. Leaching efficiencies of only 70-75 per cent. were obtained with a mixed sodium carbonate and bicarbonate solution at 70°C.

Acid leaching under fixed acid addition was investigated using two techniques.

(i) "Pug" leaching.—The ore was mixed to a thick even mass with concentrated sul-

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phuric acid and left without agitation for a period. The "pug" was then diluted with water to produce a leach liquor and residue.

(ii) Leaching at various pulp densities.—The pulp was maintained in suspension by mechanical agitation.

Both methods produced high leaching efficiencies, but acid consumption was considerable and the leach liquors were unsuitable for further processing because they contained large quantities of rare earths, phosphates and silica, which precipitated on dilution.

The possibility of leaching under constant pH conditions as a method of reducing acid consumption and the attack of undesired constituents was suggested by Hayton (1956). With this technique acid was added continuously to the pulp to maintain a desired pH value during leaching. Some typical results of early studies are shown in Table 2.

TABLE 2.—LEACHING UNDER CONSTANT pH CONDITIONS.

Sample A:

pH	Acid consumption. (lb. H_2SO_4/ton ore)	Leaching efficiency after 10 hours (%)
1.2	185	95
1.5	125	95
1.7	100	94
2.0	75	90
2.5	50	70

Under these conditions, the concentration of contaminants which affected subsequent processing was reduced to a suitable level. Leaching under constant pH conditions was established as a suitable process, and development proceeded with two objectives, viz., the determination of the optimum process conditions and the specification of suitable process equipment.

The determination of optimum process conditions

The major variants which were studied during development were leaching temperature and leaching time, and their effects on acid consumption and liquor characteristics. The optimum operating pH for these investigations was fixed at 1.7. More recently, leaching selectivity at various pH levels has been studied, and some variation recommended.

THE EFFECT OF TEMPERATURE

Leaching at a temperature of 35°C was shown to be as effective as higher temperatures with regard to leaching efficiency, but less acid was consumed and a liquor with better characteristics was produced. Leaching efficiencies and acid consumption for leaches at different temperatures are shown in Table 3, and the composition of the resultant liquors is shown in Table 4.

Other work at higher leaching temperatures indicated greater acid consumption and larger quantities of contaminants in the leach solution than those shown in Tables 3 and 4.

TABLE 3.—LEACHING EFFICIENCY AND ACID CONSUMPTION AT DIFFERENT TEMPERATURES.

Sample B:

Leaching temperature °C	35	40	45
Leaching time (hr.)	Leaching efficiency (%)		
1	75.0	87.3	87.6
2	89.6	93.6	92.1
4	93.3	95.0	95.3
6	95.3	96.4	95.3
8	96.4	96.7	95.8
16	97.2	97.2	95.8
24	97.2	97.2	97.2
Acid consumption (lb. H_2SO_4/ton ore)	109	113	121

Temperatures as high as 47°C were obtained during continuous leaching on a pilot plant scale, although the acid was diluted to approximately 65 per cent. sulphuric acid and cooled before being added to the pulp. On the assumption that 98 per cent. sulphuric acid was to be used in large scale plant operation, considerably higher temperatures could be expected. Calculations were made allowing for heat of dilution, heat derived from agitation, heat of reaction, and heat losses from the reaction vessels, and it was determined that temperatures of approximately 60-65°C could be obtained in the process at Mary Kathleen unless special measures were adopted to limit the heat input.

Dilution and cooling of the acid prior to addition to the leach was estimated to reduce the operating temperature to 37-40°C. This technique has been adopted, and is a unique feature of the plant at Mary Kathleen It is anticipated that it will reduce sulphuric acid consumption by approximately 20-25 lb. per ton of ore, as well as producing liquors suitable for further processing. It is considered to be one of the features essential for the successful treatment of Mary Kathleen ores.

THE EFFECT OF LEACHING TIME

Batch leaching for 24 hours indicated that economic recovery of uranium could be obtained with eight hours' leaching. A longer leaching time increased acid consumption and liquor contaminants, without any economic increase in uranium recovery.

A statistical analysis of continuous flow pro-

TABLE 4.—COMPOSITION OF LEACH LIQUORS AT DIFFERENT TEMPERATURES.

Sample B:

Leaching temperature °C	Leaching time (hr.)	Liquor composition			
		U_3O_8 (gm./litre)	P_2O_5 (gm./litre)	SiO_2 (gm./litre)	Rare earths (gm./litre)
35	8 16 24	2.52 2.70 2.71	0.17 0.14 0.12	4.61 6.32 6.88	4.87 7.17 7.43
40	8 16 24	2.63 2.68 2.80	0.20 0.12 0.05	5.67 7.02 7.60	6.18 8.00 8.67
45	8 16 24	2.75 2.74 2.64	0.11 0.08 0.04	5.07 6.83 7.12	6.50 8.84 9.15

cesses indicates that a percentage of the feed passes through the system in very much less than the nominal or average retention time, and likewise a percentage remains in the circuit for longer than the nominal time. A longer leaching time was used with continuous leaching to approximate the overall efficiency of eight hours' batch leaching, but similar efficiencies were obtained in eight hours of continuous leaching, which was then adopted as standard. Under these conditions the portion of ore leached longer than the nominal time was considered to be of small consequence, and its effect on the whole was neglected.

During continuous pilot plant operations, when leaching one sample of ore (Sample C) it became necessary to reduce the throughput, and the nominal leaching time thus was increased to 18.75 hours. After approximately two days, the leached pulps became impossible to settle effectively, the acid consumption was high, and considerably larger quantities of contaminants were in solution. The extended leaching time of that portion retained for longer than nominal time was considered to be responsible. Reduction to eight hours' nominal retention and two stages of leaching improved the leaching in all respects. The contrast between short and long retention times is shown in Table 5.

The reduced leaching efficiency was expected, due to the short circuiting of ore through a two-stage leaching circuit.

Further data on the effect of extended leaching time were obtained by batch leaching a sample of the same ore under the same conditions for 84 hours. After 36 hours settling data were unobtainable, and after 48 hours samples were impossible to filter.

Leaching data are shown in Table 6.

TABLE 6.—ACID CONSUMPTION AND LIQUOR COMPOSITION.

Sample C:

Leaching time (hr.)	Acid consumption (lb. H_2SO_4/ton ore)	Liquor composition			
		U_3O_8 (gm./litre)	P_2O_5 (gm./litre)	SiO_2 (gm./litre)	Rare earths (gm./litre)
8	113	7.82	0.56	9.06	24.7
12	125	—	—	8.8	—
16	137	7.93	0.50	11.6	29.7
24	158	7.87	0.60	13.6	34.9
36	131	8.4	0.79	15.9	40.0
48	196	8.6	0.62	14.7	43.0
60	201	—	—	—	—
72	206	—	—	—	—
84	212	—	—	—	—

TABLE 5—CONTINUOUS LEACHING FOR 8 AND 18.75 HOURS.

Sample C:

Leaching Efficiency, Average Acid Consumption, and Leach Liquor Analyses.

Retention time (hr.)	Leaching efficiency (per cent.)	Acid consumption (lb. H_2SO_4/ton ore)	Liquor Analyses		
			U_3O_8 (gm./litre)	P_2O_5 (gm./litre)	SiO_2 (gm./litre)
18.75	97.0	149	7.72	1.03	10.2
8.0	94.5	86	7.75	0.25	7.1

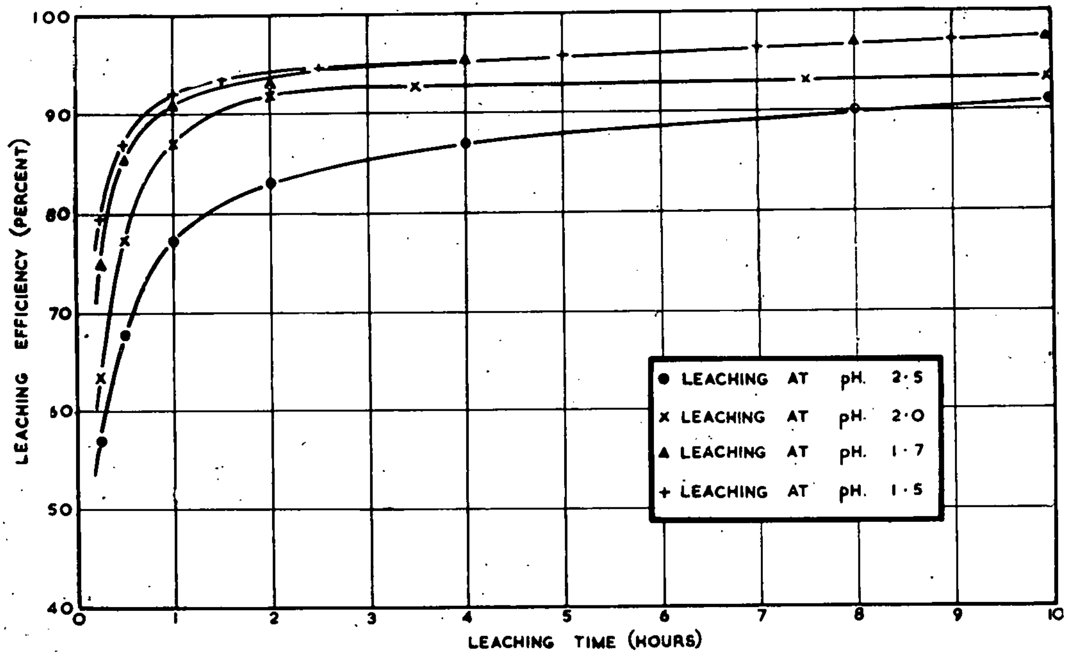

FIGURE 1: Leaching efficiency plotted against time. Sample D.

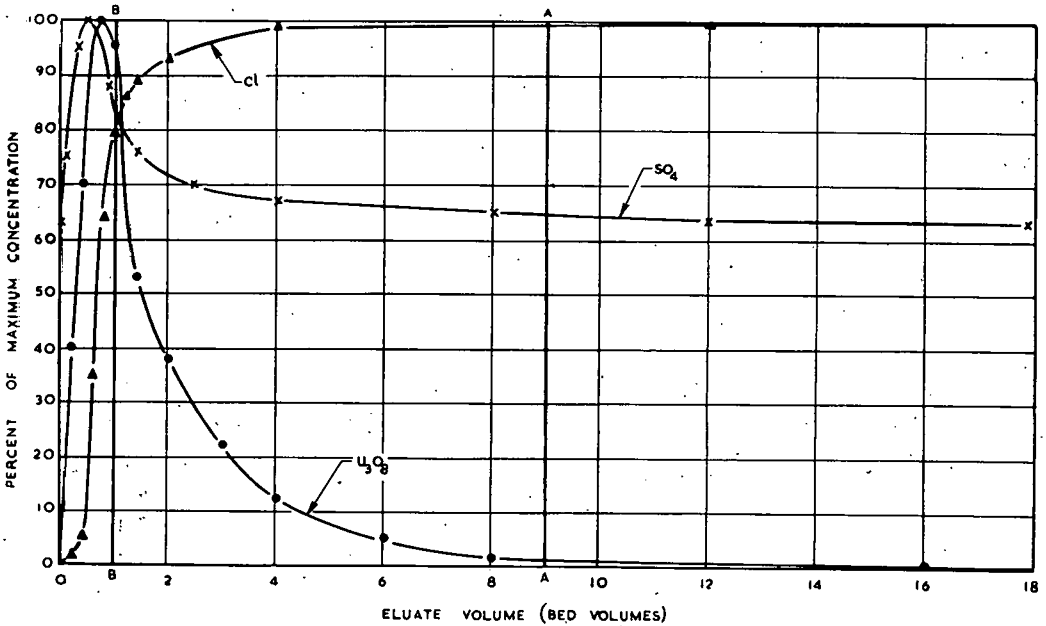

FIGURE 2: Leaching efficiency plotted against acid consumption. Sample D.

TABLE 7.—BATCH LEACHING UNDER VARIOUS pH CONDITIONS. LIQUOR COMPOSITION

Sample D:

pH	Leaching time (hr.)	Liquor Composition		
		U_3O_8 (gm./litre)	SiO_2 (gm./litre)	P_2O_5 (gm./litre)
2.5	0.25	2.41	1.45	< 0.05
	0.5	3.40	1.63	"
	1.0	3.87	1.98	"
	2.0	4.02	2.16	"
	4.0	4.25	2.70	"
	8.0	4.32	3.05	"
	12	4.34	3.48	
2.0	0.25	3.33	2.78	< 0.05
	0.5	4.06	3.52	0.06
	1.0	4.53	—	0.10
	2.0	4.70	4.50	0.12
	4.0	4.73	4.95	0.15
	8.0	4.58	5.69	0.15
	12	4.47	6.83	0.20
1.7	0.25	3.91	2.64	0.13
	0.5	4.41	3.85	0.18
	1.0	4.61	4.65	0.23
	2.0	4.63	6.12	0.25
	4.0	4.65	7.60	0.29
	8.0	4.49	8.82	0.31
	12	4.40	10.20	0.36
1.5	0.25	4.04	3.10	0.22
	0.5	4.36	4.35	0.24
	1.0	4.55	5.40	0.26
	2.0	4.56	6.75	0.34
	4.0	4.46	9.00	0.40
	8.0	4.22	11.4	0.50
	12	3.97	13.5	0.58

LEACHING SELECTIVITY

Recently, Couche and Hartley (1958) have shown that considerable selectivity is obtained by use of higher pH conditions. The ultimate economic leaching efficiency may be obtained within an eight-hour leaching time by use of different pH conditions for different stages of leaching. The advantages are lower acid consumption and improved liquor characteristics.

Figure 1 shows the leaching efficiency plotted against time for ore Sample D, when leached under various pH conditions.

Figure 2 shows the relation between these leach efficiencies and acid consumption.

Table 7 records liquor composition, which also indicates the degree of selectivity obtained.

The key to successful leaching of Mary Kathleen ores has been shown to be an accurate control of acidity. The lower the acid consumption consistent with a high leaching efficiency, the better are the characteristics of the liquor produced. Any factor which increases acid consumption, such as a higher acidity level, longer leaching time, or higher temperature, is most undesirable.

The use of pH as a method of control has been shown to be sound, but strict and accurate pH measurement and control circuits are essential items for process control. Such use of pH for the accurate control of acid addition, and thereby leaching, is a unique feature for the treatment of Mary Kathleen ore, and contrasts with plants using fixed acid additions, or acid addition broadly controlled by pH as a means of reducing excess consumption, rather than a means of producing satisfactory leaching characteristics.

The serious effects of low pH have been shown already. In addition, due to a low free acid content in the pulp during leaching, any restriction of acid flow causes a rise in pH to a level which is ineffective for leaching. In the initial stage of leaching, this pH rise is extremely rapid. After leaching at pH 1.7 for eight hours and washing the solids in a C.C.D. system with sulphuric acid of pH 2.0, the pH of the wash solution rose to 3.5 in four hours. These facts emphasise the degree of pH control required.

THE ROLE OF AGITATION

In order to ensure satisfactory pH control, adequate and efficient mixing was essential. Poor agitation results in localised low pH values close to the points of acid addition, and possibly local high pH values in portions of the tank remote from the pH electrode system. This essential requirement for efficient agitation is also a feature unique in treatment plant design, and contrasts with other plants where agitation merely has to maintain the solids in suspension.

Various pilot scale operations indicated that turbine impellers gave more consistent results than other types of impellers. Table 8 shows typical results.

TABLE 8.—THE EFFECT OF VARIOUS AGITATOR TYPES

Sample E:

Agitator type	Tank number	Leaching efficiency (%)	Acid consumption (lb. H_2SO_4/ton ore)
Turbine	1	92.3	86
	2	95.3	90
	3	96.1	106
	4	97.1	114
Marine Propeller	1	74.4	85
	2	av. 92.8	97
	3		110
	4	94.2	120
Propeller with blades of aerofoil cross-section	1	66.7	70
	2	74.9	78
	3	82.2	92
	4	88.7	94

The specification of suitable process equipment

While continuous leaching involves some excess leaching of a percentage of the ore, other factors such as the treatment of large tonnages and the need for constant acid addition, make this form of leaching desirable.

Pachucas were considered for leaching, as theoretically they were the most economic. They were eliminated because of the close pH control required and the low rate of turnover in a pachuca (approximately 0.25 hours), as well as the possibility of the sand fraction settling out. Leaching tanks fitted with mechanical agitators were selected as essential.

Two banks each of four leaching vessels were recommended. The two banks were chosen in order to reduce the length of overhung shaft required for agitation. The four stages of leaching were chosen to reduce the percentage of "short-circuiting" of incompletely leached material. Turbine type agitators were recommended and are being installed.

Solid-liquid separation

This section deals with both the pregnant liquor recovery, and the clarification of that liquor.

Pregnant liquor recovery

The choice for the major solid-liquid separation was either filtration or continuous counter-current decantation (termed C.C.D.). In the former case lower volumes of higher grade liquor are produced, and this is a factor usually given first preference. Provided sufficient water is available, the adoption of a C.C.D. system, with its superior washing efficiency, is a matter of economics. The higher capital cost and the disadvantage of treating a larger volume of pregnant liquor have to be compared with the revenue received from the added recovery of uranium. Recent practice has favoured C.C.D.

FILTRATION

Filtration data was collected both from leaf tests and pilot plant rotary drum filtration. The pulp was conditioned with flocculent but, although high initial filtration rates were obtained, the cloth blinded rapidly and the rate dropped sharply. Washing efficiency was very poor, and high losses of uranium were recorded. Poor washing is a characteristic of ores leached in acid circuits, and all treatment plants using this method of solid-liquid separation have accepted large operational losses.

Overall loss indicated from the experimental studies was approximately 2-3 per cent. for two stage filtration, and 0.3-0.5 per cent. for three stages. Typical data are shown in Table 9.

TABLE 9.—FILTRATION DATA.

Test	Cake moisture (per cent)	Filtration rate lb./(sq. ft.) (hr.)	Washing efficiency (per cent.)
Leaf (average of three tests)	16.5	155	50
Rotary Drum (new cloth)	17.0	210	30
Rotary Drum (used cloth)	16.1	120	17
Rotary Drum (repulped solids)	16.0	Serious blinding, rate not determined	—

It was estimated that at least six large acid resistant filters would be required for the treatment plant, with their ancillary equipment. The estimated operating charges were high because of the high power consumption of vacuum pumps and the short life of filter cloths. In addition, an added loss of uranium would have to be accepted.

C.C.D. SYSTEM

In contrast to filtration, a C.C.D. system is very efficient in recovery of valuable materials in solution, and the five-stage system under consideration theoretically recovers 99.86 per cent. of the quantity present in the leach liquor. Analyses of washed residues after leaching and unwashed residues after C.C.D. were identical. For practical purposes recovery is complete. The only objections to a C.C.D. system were the capital cost of a five-stage system and the handling of increased volumes of liquor. However, these were more than offset by the higher operating costs and the larger losses incurred by filtration.

THE EFFECT OF FLOCCULENT

Without the use of recently developed synthetic flocculating reagents, the handling of the leached pulps by any means would have been extremely difficult and costly. Both filtration and thickening gave poor performance when unflocculated pulps were used. Figure 3 shows settling rate curves plotted from results of tests which used nil and various quantities of flocculent. These curves show the marked effect of the reagent.

The C.C.D system was based upon studies of pulps flocculated with a polyelectrolyte reagent, and the equipment was designed for use with such pulps. This fact makes the design unique. In designing the thickeners to use such reagents, it was necessary to assume that flocculation in large-scale operations would be as effective as on the experimental scale. During pilot plant operation two factors were observed which affected flocculation, viz., the condition previously referred to as "overleaching," and the method of addition of the flocculent.

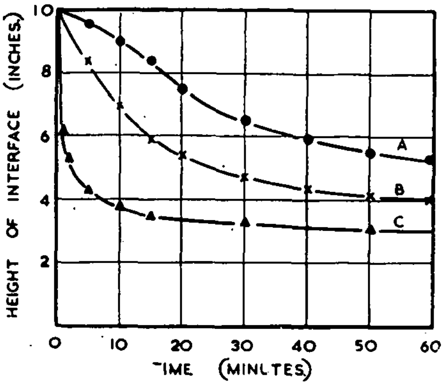

FIGURE 3: Settling curves for Mary Kathleen pulp after leaching for eight hours.
A. Nil flocculent.
B. 0.05lb. flocculent per ton of ore.
C. 0.15lb. flocculent per ton of ore.

Some effects of "overleaching" were noted earlier, but the most serious problem associated with this condition, was the failure of the reagent to flocculate the solids effectively. The increased content of silica, phosphate and rare earths in the leach liquors has been shown in Tables 5 and 6. It is thought that the large quantities of colloidal matter present in the leach pulp adsorbed the flocculent without showing any appreciable flocculation. The only corrective measure was a variation of the leaching conditions, as has been indicated earlier.

Figure 4 shows settling rate curves plotted from tests using pulps leached for various times, and shows the deterioration of settling with excess leaching.

The technique adopted for additions of the flocculent must ensure adequate, but gentle, mixing with the pulp. Pilot plant operations showed this technique may well be critical. Not enough mixing caused poorly flocculated pulp, and if mixing was violent or too long, degradation of the flocs resulted in poor settling.

Large-scale pilot studies showed consistent results. Therefore, in design, it was assumed that at least similar results would be obtained in plant practice. This assumption permitted the acceptance of thickener area and depth determinations based on experimental data. Because of the high density of the Mary Kathleen ore, the pulp settles rapidly to terminal density, and the retention time in any thickener is very short. This factor controls the depth of the compression zone, and in this instance it is found that compression is almost eliminated. Consequently, the thickener depth specified was less than normally required.

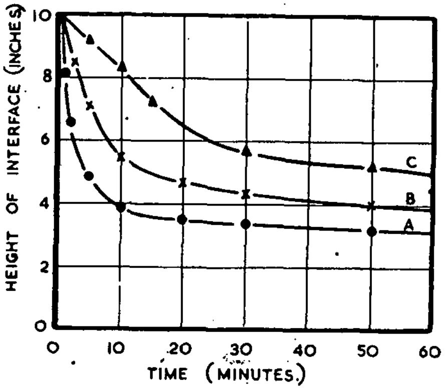

FIGURE 4: Settling curves for Mary Kathleen pulp using 0.15lb. flocculent per ton of ore.
A. 16 hours' leaching.
B. 24 hours' leaching.
C. 36 hours' leaching.

This variation caused some mechanical problems, because the design was not standard, and some modifications had to be made to adjust the thickener mechanisms to meet the specification.

Clarification

The small quantity of slime, approximately 100 parts per million, which is contained in the pregnant liquor from the first stage of the C.C.D. system, has to be removed prior to subsequent treatment by ion exchange. There are several acceptable methods available, and these were considered.

SAND CLARIFICATION

A simple and positive method is sand filtration, but it has limitations. Adequate backwashing and thorough removal of the slime from the sand bed is essential for satisfactory operation. It was found that some method for breaking the slime layer which forms on top of the sand bed was necessary, otherwise large masses of slime settled through the fluidized sand bed when backwashing, and accumulated on the gravel bed. Other disadvantages include the disposal of backwash liquor, and the possibility of handling pregnant liquor containing a higher concentration of solids. In the latter case the backwash cycle would be more frequent, and disposal of this liquor could become an embarrassment.

FILTRATION

Systems include pressure and vacuum leaf types, and rotary drum precoat filtration. Pressure leaf types, similar to the Hercules filter, may be made automatic in operation, but vacuum leaf or "Moore" filters are manually cleaned. A low-grade filter aid is used with these types of filters as a protection for the filter cloth. Disadvantages are the large number of leaves required to obtain the flow rate, the consequent maintenance in renewing filter bags, and intermittent operation. The "Moore" type is widely used in overseas practice.

The ultimate in both theory and chemical engineering practice is continuous precoat filtration. This type provides flexible operation. High rates of filtration may be maintained, regardless of the slime content of the liquor. Pilot plant studies of this indicated satisfactory operation, but there was a tendency toward high consumption of filter aid. Experience on a large plant also indicates that filter aid usage is high, although some overseas operators have reported exceptionally low consumption.

Precoat filtration was selected, and Eimco high submergence units are being installed.

Ion exchange

LIQUOR CHARACTERISTICS

Process considerations

The Mary Kathleen leaching and C.C.D. circuits produce a liquor of a composition well suited to ion exchange extraction of uranium. Table 10 shows some typical analyses of liquors produced on a pilot plant from samples of ore taken at various parts of the mine.

The pH of the liquor from the leaching circuit is 1.7 to 1.9—a range considered nearly optimum for the ion exchange treatment of liquors of low ferric iron concentration. No neutralisation stage is therefore required subsequent to the leaching. The plant, in fact, is designed so that the C.C.D. wash liquor can be acidified to prevent hydrolysis in the process stream.

The variations in the sulphate concentrations shown by the above figures are characteristic of the pH-controlled leaching process. The addition of a fixed quantity of acid, which is the more conventional leaching process, produces a liquor of constant sulphate composition. In this plant, however, the acid consumption will vary with the ore character, and this in turn will be reflected by a variation in the sulphate content of the pregnant liquor. However, this range of concentration is again suited to the ion exchange process.

ADSORPTION

Liquors containing as little as 7.5 grams of sulphate per litre have been tested. There was a tendency to produce "royal" barrens when tests were carried out using only a single column, but no trouble was experienced in the adsorption cycle using multiple columns. The resin in the leading column was substantially converted to the sulphate form before the concentration of the uranium rose to the feed value.

In initial experiments 2in. diameter ion exchange columns were used. Saturation loadings were shown to be high for a process liquor, and loading rates were rapid. On an experimental plant using Deacidite FF resin with a capacity of 1,280 meq. per litre, a bed depth of 3ft. and a flow rate of 2 gallons/sq. ft./min., and a pregnant liquor of composition quoted under No. 2 in Table 10, the following results were obtained:

TABLE 10.—PREGNANT LIQUOR ANALYSES.

Sample No.	Constituent				
	U_3O_8 (gm./litre)	SO_4 (gm./litre)	Fe (gm./litre)	SiO_2 (gm./litre)	P_2O_5 (gm./litre)
1.	0.80	12.5	1.0	2.2	0.16
2.	0.72	19.8	1.4	3.2	0.10
3.	1.0	7.9		1.4	0.04

Saturation loading: 75 grams U_3O_8 per litre of wet settled resin.

Operational loading: 63-67 grams U_3O_8 per litre of wet settled resin, with a recovery of 99.7 per cent. of the uranium.

The breakthrough, taken as 2 per cent. of the pregnant liquor concentration, was sharp, and loading curves were well defined.

This was the picture from the experimental pilot operation, and by and large, is predicted to represent the performance expected from a large plant. The resin to be used in the large plant has a somewhat higher capacity of 1,520 meq. per litre, so that if the loading rate per unit weight is substantially the same, operating loadings of 70 to 75 pounds of U_3O_8 per 100 gallons of wet settled resin can be expected.

SILICA POISONING

Silica poisoning of anion exchange resins used for uranium extraction is experienced in several plants in South Africa from pregnant liquors containing 0.6 to 1.5 grams of SiO_2 per litre. The silica content of the liquor expected to be treated at Mary Kathleen is consistently higher by comparison, and this is expected to lead to silica poisoning of moderate to high severity.

Using a pregnant liquor containing 2.2 grams of SiO_2 per litre, the amount of silica on the resin was found to be 3.0 per cent. after 13 cycles, and 4.3 per cent. after 28 cycles.

The soluble or ionic silica in the above liquor was approximately a third of the total silica, the remainder being termed colloidal.

The poisoning was shown to be related to the age of the liquor, because the ratio of the soluble to the colloidal silica changed during standing. The ionic or soluble silica apparently polymerised and changed to colloidal form, the particle size of which was too large to enter the structure of the resin. It is probable that ionic silica is adsorbed by the resin and later changed to colloidal form which then becomes trapped within the resin. However, there is a definite indication that some silica is also deposited on the outside of the beads.

During an extended pilot operation, severe pressure drops occurred across the resin bed. The pregnant liquor in this case contained approximately 2 grams of total silica per litre, and the columns were operated at 3.6 gallons/sq. ft./min. An increase in back pressure was noticed after a few cycles, and after only 14 cycles this became severe enough to stop the plant. The material which caused the blockage contained 27 per cent. SiO_2, and the resin contained 10.4 per cent. SiO_2. Although silica contributed to the trouble, it is clear that other factors may have contributed to the severity of the blockage.

Although the increase in pressure drop across the resin bed seriously affected the pumping rate, in pilot plant work there was no apparent decrease in the saturation loading capacity of the resin with increasing silica content. The rate of loading may have been affected, but this was not certain.

ELUTION

The operating cost of the ion exchange process mainly depends upon the price of the reagents used for eluent make-up and neutralization.

The eluent to be used at Mary Kathleen is a solution 1.0 molar in sodium chloride and 0.05 molar in sulphuric acid.

The following are some of the elution procedures which were either considered or examined for the Mary Kathleen plant:

Nitrate Elution.—Two stages of elution and two of precipitation are used in South African plants. The whole of the nitrate required for eluent make-up is supplied by nitric acid. The liquor concentration is so adjusted that the eluent contains just sufficient acid to ensure that during the subsequent lime neutralization, sulphate is precipitated as gypsum equivalent to the sulphate eluted from the resin.

There is no doubt that this procedure is the most elegant, in that one regenerant only is required and that a liquid is used for eluent make-up. Sulphate is controlled to a minimum so that elution efficiency is high, and there is no need to wash the product for removal of chloride. Unfortunately, the quite exorbitant price of nitric acid in this country, and the long haulage required for the acid to Mary Kathleen prohibits the adoption of this circuit.

Sulphuric Acid elution.—The use of sulphuric acid for elution was never seriously considered. There is no recycle value in the eluate after precipitation, and the combined cost for the required amounts of acid and neutralizing base is prohibitive. Some consideration was given to a circuit involving sulphuric acid elution followed by solvent extraction. In this case the sulphuric acid can be recycled to the ion exchange plant after its contained uranium has been extracted. The extra cost of this procedure was not justified, however, as there was no incentive to produce a product of higher purity than could be produced directly from ion exchange.

Sodium Chloride—Sulphuric acid elution.—Several variations of the circuit were examined using an eluent of 1 M sodium chloride and 0.05 M sulphuric acid as follows:

(a) The sulphate concentration in the eluent was controlled at 20 to 25 gm/litre.

(b) The sulphate concentration was allowed to increase to 80 gm/litre.

(c) Sodium chloride elution was considered with two stage elution, incorporating the separate neutralization treatment of a high sulphate fraction of the eluate and a chloride recovery stage in the elution cycle.

(d) Consideration was given to a three-stage elution procedure to produce a high grade, high sulphate fraction which could be rejected after precipitation. Both the other eluate fractions were recycled.

In considering which of the above circuits should be used, the decision depends on the local price structure of the various reagents.

A high percentage of recycle of the barren eluate leads to a good recovery of the chloride and savings in salt consumption. For equivalent elution, however, a greater volume is required than when using the alternative procedure where an appreciable volume of the barren eluate is rejected in order to keep the sulphate concentration below 25 grams per litre. Because the acid concentration is the same in both cases, it is apparent that in the first case appreciably more acid is required in the make-up and more lime in the subsequent neutralization. Consequently, the choice of circuit depends on the comparative costs of salt and sulphuric acid. At Port Pirie, where the price of salt is approximately £5 per ton and sulphuric acid approximately three times that amount, there is a greater incentive to save acid.

At Mary Kathleen, salt is approximately £35 per ton, and any single ton of acid being produced on site is worth little more than that at Port Pirie. In this case, therefore, the incentive is in favour of saving salt.

It has been demonstrated that the greatest economy can be achieved for both salt and acid by introducing a second precipitation circuit. Figure 5 (Presgrave, 1958) illustrates a typical set of concentration curves for the uranium, sulphate, and chloride during the elution stage. In the more conventional procedure, the eluate would be divided into two parts as shown by the line AA. The first nine bed volumes would pass to the precipitation circuit, whilst the second nine bed volumes would be recycled to the first part of the next elution. The first half is treated with lime, usually in the form of calcium carbonate, to neutralise sulphuric acid. After removing the gypsum precipitate, the

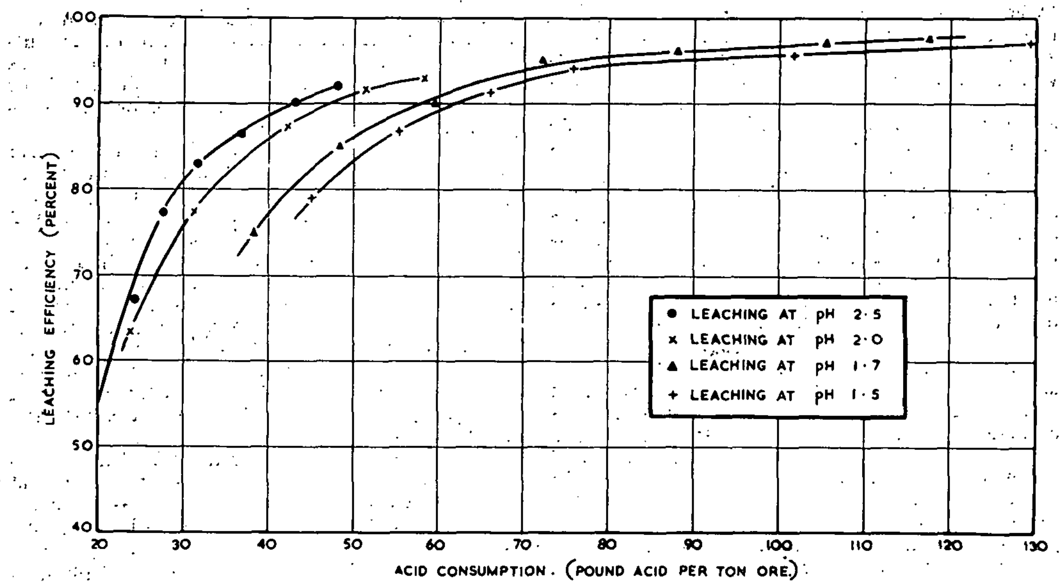

FIGURE 5: Elution curves for uranium, sulphate, and chloride. Resin used, Deacidite FF, capacity 3.4 meg./gm. Eluent used, NaCl, 1.0 M; H_2SO_4, 0.05 m; SO_4, 82 gm./litre. Eluent flow-rate, 0.15 gallons/sq. ft./min. Maximum concentrations, U_3O_8, 32.0 gm./litre; SO_4, 130 gm./litre; Cl, 39 gm./litre.

filtrate at pH 3.5 is treated with magnesia to raise the pH to 6.5 or 7.0 in order to precipitate uranium. The maximum amount of sulphate which can be removed by the calcium carbonate treatment stage is equivalent only to the sulphuric acid originally added to adjust the solution to 0.05 molar in acid strength. None of the sulphate eluted from the resin can be removed in that stage. Consequently, a certain fraction of the barren eluate must be bled from the circuit to control the sulphate concentration in the recycled solution. Because the chloride concentration of the barren eluate is between 25 and 30 grams per litre, this bleed represents an appreciable loss of chloride.

In the modified process to be used at Mary Kathleen, the eluate is divided at a point shown approximately by line BB. As can be seen, this relatively small fraction contains most of the sulphate stripped from the resin, but its chloride concentration is considerably lower than the bulk of the eluate. This fraction is then precipitated separately, and the filtrate is rejected so that sulphate control is effected with a minimum loss of chloride.

At the end of elution, the resin is left substantially in the chloride form. This chloride can be removed by elution with sulphuric acid. Because sulphuric acid must be added to the eluent at the rate of approximately 5lb. per 100 gallons, further economies can be achieved by first using this acid in a final stage of elution to recover an appreciable fraction of the chloride from the resin. The concentration of the acid used for this stage is determined by the final liquor balance. Thus the first half of the elution shown in Figure 5 is nine bed volumes (AA). One bed volume (BB) is rejected in the high sulphate circuit. If we assume that one-third of a bed volume is subsequently gained from the washing and brine make-up stages, the required amount of acid can then be added in two-thirds of a bed volume to readjust the recycled eluate to its original volume. If we assume one bed volume is 300 cubic feet. then the acid required to prepare nine bed volumes of eluate at the rate of 5lb. of acid per 100 gallons is 843lb. of a bed volume is 1,205 gallons. Therefore, the concentration of the acid which can be used in the final elution stage is theoretically 70lb. per 100 gallons, or 1.4N. In practice, any chloride removed by the acid from the resin will be replaced with bisulphate ion. Consequently, the acid must be adjusted to allow for this loss of hydrogen ion, and the concentration will therefore be greater than 1.5N. This concentration of acid is a fairly efficient eluent for the chloride, and, provided that time is available in the elution cycle, approximately 70 per cent. of the chloride can be recovered. The acid lost in this process by exchange with chloride must be taken into account in assessing the economics. Again, this acid could be recovered by washing the resin with water, but the number of stages in the operation tends to make the progress rather unwieldy.

In practice, the economics of any elution circuit are quite sensitive to the overall water balance. The above circuit to be used at Mary Kathleen is particularly sensitive, and it is doubtful whether the savings which can be shown on paper will be achieved in practice.

It is apparent that the total volume increase in the circuit must be kept to quite a low figure. As the product is produced to a specification which calls for efficient washing, such a balance is not easy to achieve without discarding some wash solution of relatively high chloride content. This stage, of course, would counteract the purpose of the whole operation. It is possible to broaden the elution curve by increasing the speed of elution and allowing increase in eluent volume with consequent increase in acid and lime requirement and decrease in uranium concentration. The most efficient operating circuit can only be determined from plant practice.

Features of the plant

Basically, a conventional ion exchange plant is being installed, with associated pumps, tank farm, and precipitation sections. The ion exchange plant itself is being supplied by the Permutit Company of England, and consists of three units each with four columns 8ft. in diameter. Each column contains 300 cu. ft. of resin. The plant is fully automatic, and incorporates the latest improvements in design.

The liquor distributor in the column is designed to minimise backmixing of eluent in the water dome. Such backmixing can result in a considerable loss of eluting solution and cause "royal" barrens or barrens with high uranium concentration. At one time it was proposed to incorporate a drain down cycle in order to simulate air dome operation and recover the solution in the top of the columns before backwashing. The use of the improved distributor was considered a sufficient safeguard.

Full flow, variable area, float type flowmeters are being used in place of differential pressure types for flow indication and control, with the pregnant liquor flowmeter also incorporating an integrator. Integration of the eluate flow at this stage is not highly accurate, and is best incorporated in the circuit after the eluate storage tank.

Open weir boxes are being installed on the outlets of the main process liquor lines from each set of columns as an operating safeguard to facilitate the correction of irregular flow and short-circuiting due to leaking valves. Liquors will flow by gravity from this point to their respective storage tanks.

The controls incorporate an unusually large number of stages in the operating cycle, 11 in all being available, including the standby position. This number was originally proposed in order to incorporate the drain down stages and stages for washing the resin with regenerant solution. When these stages were eliminated, the extra cycles available enabled the modified elution procedure to be incorporated as outlined above. The equipment is designed to cope with the possibility of a high head; due to silica poisoning, developing during adsorption.

The regeneration plant

With the likely occurrence of serious silica poisoning or fouling. a process was developed by the South Australian Department of Mines for regenerating the resin (Lawrie and Presgrave, 1956). Experimental work demonstrated that, when the resin was washed with 0.05 molar ammonium fluoride in 0.5 molar sulphuric acid solution, the silica could be effectively removed. Such a wash could be carried out on a batch principle, or incorporated as a stage in each elution cycle. In the latter case, accumulation of silica on the resin was completely inhibited. Detailed costing subsequently showed that, despite the operating advantages of such a process, it was several pence per pound more expensive than the conventional method of using sodium hydroxide.

Because this latter process involves a change from an acid to an alkaline medium, it is not convenient to incorporate it as part of the cycle. Overseas experience has proved that effective regeneration with sodium hydroxide can only be assured by removing the resin from the process column to a separate, specially designed regeneration plant. When "poisoned," the resin will be transferred by hydraulic displacement to a special processing vessel and treated with 5 per cent. sodium hydroxide solution. After washing and reacidification, the resin will be transferred to a small storage or transfer vessel, and again hydraulically pumped back to the ion exchange plant.

Precipitation and drying

The plant being installed for the subsequent product precipitation stages of the eluate is more or less conventional equipment. Continuous processing has been used in this case for both the high sulphate eluate fraction and for the gypsum and product precipitation from the main eluate fraction.

The product from the precipitation of the high sulphate eluate will be pumped to the product filter, and the overflow from this stage pumped to waste. All waste barren eluate will be pumped through a supercentrifuge to remove small amounts of suspended product which overflow the thickeners. Rotary vacuum filters are being installed for filtering and washing the product.

A through circulation continuous belt dryer is to be used as the final stage of the product treatment. One feature of this dryer is the extruder for feeding the wet product. This consists of a pair of rolls which act as a gear pump and extrude the product through nozzles.

Acknowledgements

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The Location, Mining and Concentration of Uranium Ores at Radium Hill, South Australia

By T. A. Rodgers*

Uranium ore of davidite type is being mined from narrow hypothermal fissure lodes at Radium Hill, South Australia. Modern methods are used in the exploration, development, extraction and concentration of the ore. Techniques and processes, which include scintillation surveys, open overhand stoping, dense medium separation, and flotation, are described.

Geology

Regional geology

The uranium ore deposits at Radium Hill occur in a variety of extensively folded and faulted ancient gneisses and schistose products of metamorphism. This rock environment is believed to be of lower Pre-Cambrian age. Occurrences of uranium bearing minerals of the davidite type, which seem to be associated with hydrothermal conditions of deposition, follow well-defined but irregular fault planes. The ore genesis pattern presents evidence of the existence of high pressures and temperatures during deposition.

Local geology

Although the thickness of the lodes has, in a few instances, been found to exceed 20ft., the mean mining width is approximately 48in. The shears in which mineable ore is found commonly have a moderately steep attitude (50° to 60°).

The uraniferous mineral forms part of a complex association of intergrowths and exsolutions of rutile, davidite, ilmenite, haematite, and other iron-titanium rich minerals. (Figure 2.) Accompanying gangue minerals are frequently black and bronze biotite and quartz. The composition of the pseudo-mineral davidite which yields the uranium has tentatively been described as $(FE, Ln, U)\ (Ti, Fe, V, Cr)_3\ (O, OH)_7$. Recent work suggests that this does not fully express the chemical nature of the substance.

Carnotite, a hydrous potassium uranium vanadate, occurs as a secondary and minor source of uranium. It is confined mostly to the upper ore horizons in the mine.

Exploration

Historical

The discovery of davidite at Radium Hill was made in 1906, by Mr. A. J. Smith, who mistook the black shoaded mineral for tin ore. The presence of yellow carnotite on weathered samples sent to Adelaide for analysis led to the identification of a uranium-radium mineral. Between 1908 and 1931, several attempts were made to produce radium in commercial quantities, but none of these ventures was a financial success. In 1944, at the request of the British Government, a geological investigation of the area was made, but the interest at that time was in deposits which could be considered as possible immediate producers of significant quantities of uranium, rather than in deposits which required deep underground development and presented unsolved metallurgical and chemical problems.

The Mount Painter field was considered to be of more immediate interest, and it was not until 1947 that systematic drilling and other exploratory work was commenced at Radium Hill to assess the value of the uranium bearing ore bodies. In 1952, a contract was arranged with the Combined Development Agency for the sale of uranium oxide produced from Radium Hill ore. The development of ore concentration and chemical treatment processes was undertaken at the same time as the large scale preparation of the ore bodies for mining. Permanent water, power, and railway services, as well as a town, were constructed.

FIGURE 1:—Radium Hill ore x 200. Legend: white, haematite; light grey, ilmenite; dark grey, davidite; black, biotite.

Radiation surveys

After a regional airborne scintillation survey in July, 1951, more localised geophysical examinations were made from low-flying aircraft. Areas of interest disclosed in this manner were subsequently examined by close surface traversing with manual scintillation counters of the LaRoe type FV-5 and the

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* Radium Hill Project. Manuscript received March 17, 1958.

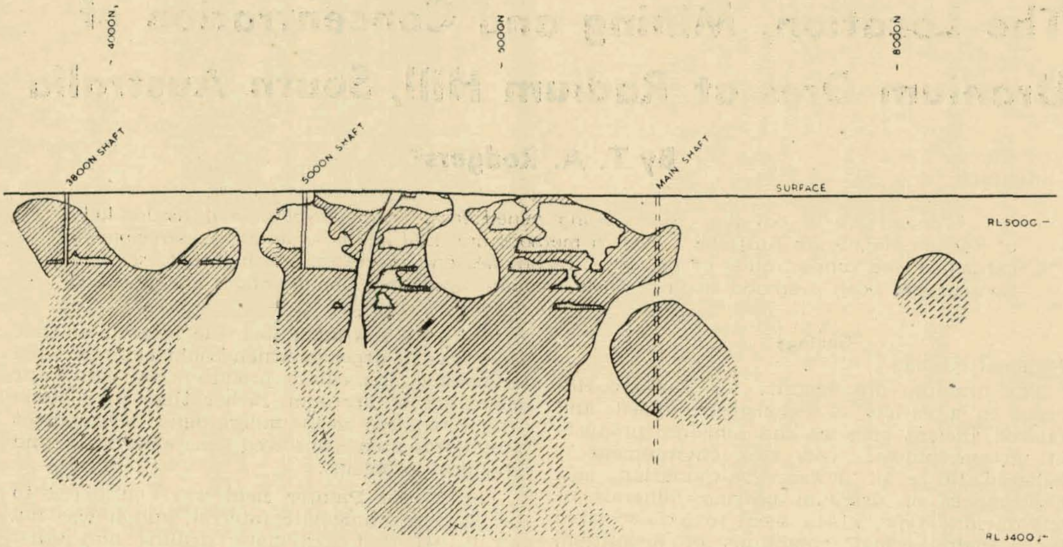

FIGURE 2:—Longitudinal sectional diagram of Geiger lode. Indicated and inferred ore is shaded and main areas of stoping are outlined.

Halross type 939. Broad areas were also examined in similar detail after their selection on purely geological grounds.

The black mineral complex which is sought for its association with uranium, outcrops in part of the area adjacent to the mine. Comprehensive examinations of the locality by close manual scintillation traversing is less effective than might at first sight be assumed because:

(i) where primary ore structures have been exposed to the atmosphere for long periods there appears to be a migration of radioactive minerals leaving a leached outcrop which effectively shields the gamma radiation arising from the ore below, and

(ii) shallow beds of travertine and river alluvium cover about half of the area of greatest potential interest. Where this cover is barren and exceeds two feet in thickness lode radiation is masked.

Fortunately, there is a tendency for sufficient residual radioactive salts to be precipitated from migratory solutions or for sufficient unleached fragments of ore to be retained in the overlying soil, to create anomalies in the isorad pattern. A masked lode at co-ordinate position 4600N 5000E was detected in these circumstances where no visible rock outcrop whatsoever appeared at the surface. It promises to yield many tens of thousands of tons of mineable ore.

Use of other geophysical aids

Ground magnetometer surveys, in the form of reconnaissance traverses and detailed local gridding, have been completed over much of the exposed line of lode. The gneissic country rock is rich in magnetite, and basic igneous intrusives are scattered over the field; faults and the more acid dykes provide planes of discontinuity in the natural magnetic pattern. The magnetic surveys gave considerable support to the hypothesis that the uraniferous lodes favoured the areas of great structural complexity and abnormal igneous activity. Apart from providing this rather general information, they were unsuccessful in small scale ore finding.

Preliminary field work in the use of electromagnetic methods has been undertaken without yet producing conclusive results.

Surface costeaning and diamond drilling

Outcrops are exposed for geological examination by removal of soil and weathered rock by bulldozing and costeaning.

Diamond drilling has proved to be very valuable in determining the lateral and downward extensions of ore in the known uranium lodes and in broad exploratory work. Secondary diamond drilling is carried out from sites chosen in the underground workings. Information disclosed by diamond drilling gives a broad picture of the tonnage of ore available and its uranium content, and provides advance information for the planning of underground shaft sinking, driving, and crosscutting. (Figure 3.)

Mining

Underground development

Entry to the mine workings is through a vertical shaft having a nominal cross section 16ft. by 12 ft.—Figure 4. Its depth at present is 1,370ft. A carefully planned 24-hour cycle of drilling, charging and blasting in benches is observed.

Waste rock is hoisted in two stages to the surface. Except where softer rock is exposed in the walls of the excavation, close timbering or concreting is unnecessary. A steel framework to provide support for the shaft vehicles and services is fixed at intervals in the shaft by heavy steel supports extending into the adjoining rock.

Close tolerances are observed in shaft alignment and from the surface to a depth of 1,200ft. the deviation from the vertical is not more than 0.5in.

At intervals of 100 or 150ft. in the vertical shaft, stations lead through main crosscuts to the uranium ore bodies. Driving within the several lodes is at a size determined by the working clearances necessary for underground excavators of the Eimco 12B type, and by the space requirement for subsequent preparation of ore for stoping. Sub-vertical connections between levels are made by winzing and rising at minimum cross section.

These openings expose the lodes for geological examination and sampling, and provide access for subsequent extraction of ore.

Stope preparation and stoping

The lode attitudes, the ore-country rock boundaries, and the adjoining country rock have characteristics which usually permit overhand open stoping to be adopted. Only rarely does stoping width exceed 48in. Minor tonnages are produced from shrinkage stopes, and from inclined cut and fill activities.

Because of the erratic distribution of the uranium minerals, constant geological examination and close sampling are essential if dilution of valuable material with waste rock, and outright loss of ore, are to be avoided.

Rockdrills used are mounted on pneumatically controlled extensible legs.

Close supervision of underground work is expensive and ineffective. A high-geared system of incentives is applied to both developmental and productive activities. In a heading of a given size, a rate of payment per unit of advance is fixed by negotiation between trained representatives of management and members of small contracting parties. To avoid deliberate over breaking during stoping operations, the scales of payment contain factors related to the completed widths of stopes.

Haulage and hoisting

Broken ore is passed down through the mine in a system of sub-vertical openings of small cross-section. These ore passes lead to loading stations adjacent to the main shaft. A measured quantity is prepared for loading during the passage of a skip from the discharge point at the surface to the underground loading point. Loading is almost instantaneous. Mechanical devices actuated by the arrivals and departures of skips prevent unscheduled discharge of ore into the shaft. Two 3ft. 6in. by 3ft. 6in. by 5ft. 6in. conventional Kimberley-type skips are used. A steel headframe provides support for head sheaves and run-of-mine product segregation facilities.

Two double-drum single reduction 280 h.p. winders control movements of the cage and counterweight, and of the two skips in balance. Maximum rope speed is 1,000ft./minute. Inbuilt safety devices prevent the winder driver from exceeding pre-determined speeds of acceleration, normal travel, and deceleration. Upper limits for winding in each compartment are supplied to a Pickering control.

Drainage and ventilation

Dust produced during rock drilling, blasting, and movement of broken ore, is controlled by providing sprays of water at the point of origin, or in the case of fine airborne particles by brisk ventilation. Apart from the toxicity of

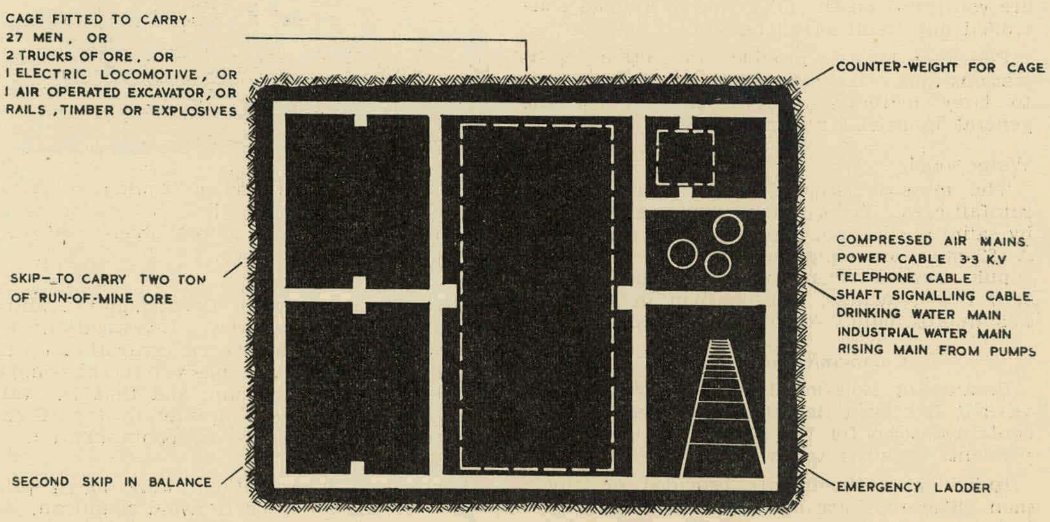

FIGURE 3:—Diagrammatic plan of main shaft.

the uranium component of the dust and its radioactivity, drilling and blasting operations increase the radon content of the mine air. Adequate ventilation is provided to ensure that the radon tolerance of 1 x 10^{-10} curies per litre is not exceeded.

Some re-cycling of mine water is practised, but the build-up in radon content limits the use of this economy.

Pumping stations to deal with the small quantities of highly saline water percolating through the rocks and the surplus water from mining operations are provided at Nos. 3, 6, and 8 levels.

Fresh air is drawn into the mine workings by an exhaust fan placed 1,300ft. south of the main haulage shaft. It is powered by an 80 h.p. motor and air flow is regulated by a variable speed fluid drive. Rated capacity of the installation is 100,000 cubic feet of air per minute—W.G. 3in. Secondary ventilation from the main airways is provided by small axial flow fans.

Filling

In cases where mining of two or more subparallel lodes is carried out contemporaneously, the intervening mass of rock may develop a minor degree of instability. Precautions are taken against the development of major instability by providing residual pillars of low grade or barren material, and by occasional filling. It is anticipated that extensive filling will ultimately be required. A sandy mill waste product is being examined for possible use as a material for hydraulic refilling of depleted stopes.

Industrial services

Power supply

A 130,000V transmission line brings power 130 miles to the field from Morgan. Two main transformers — 132,000/11,000V — 2,000KVA are protected on the 132kV side by interrupters which open circuit under fault conditions and are controlled on the 11kV side by remote controlled oil circuit breakers.

Power at 11kV is provided to sectional substations and is transformed to 3.3kV for supply to large motors — 415V is adopted for the general industrial requirement.

Water supply

The mine is situated in an extremely low rainfall area. Local water supplies are affected by salinity, siltation, and periods of exhaustion. A 52-mile long pipeline provides a reliable supply from a reservoir at Umberumberka in New South Wales. Consumption in 1957/1958 was approximately 30,000,000 gallons.

Community arrangements

Because of isolation from metropolitan services it has been necessary to provide a self-contained town for the 900 employees and dependents required to live on the field.

Individual hutments are provided for single men. These are arranged about a central mess building and a recreation building. One hundred and sixty-five conventional houses are provided for married employees.

The town has been planned so that houses and single men's quarters are spaced about a central sports and shopping area. Amenities include a hospital conducted by the Australian Inland Mission, a post office, two schools, sporting clubs, sewerage, regular train and air services, and some bitumen roads. Regeneration of the meagre natural flora is being encouraged and several thousands of new trees have been planted.

Mineral dressing

Introduction

The development of a large-scale method of concentrating the previously little known mineral, davidite, required the full resources of the South Australian Department of Mines' Metallurgical Laboratories. Investigation suggested that the selective concentration of the davidite portion of the black mineral complex was economically unsound, and that the variable and minute scale of intergrowths (Figure 2) would lead to serious contamination of products.

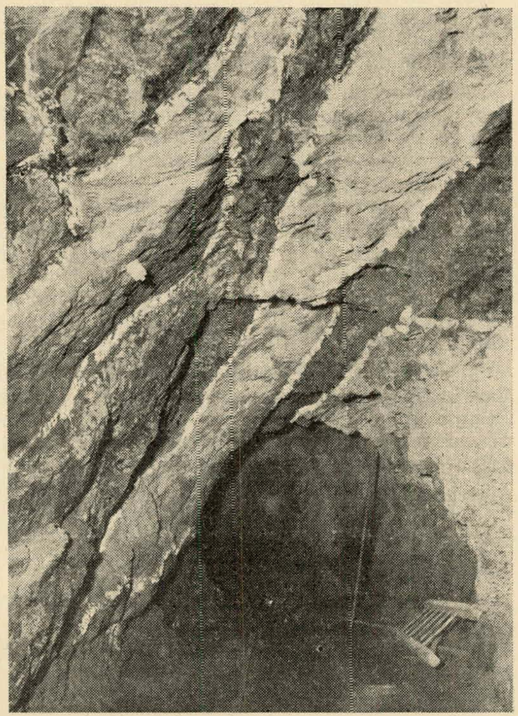

FIGURE 4:—Drive in uranium bearing ore. White chalk has been usd to outline the boundaries between the darker ore and the lighter-toned waste rock.

Subsequent investigations were based principally on the intention to avoid significant loss of uranium by collecting the davidite associates,

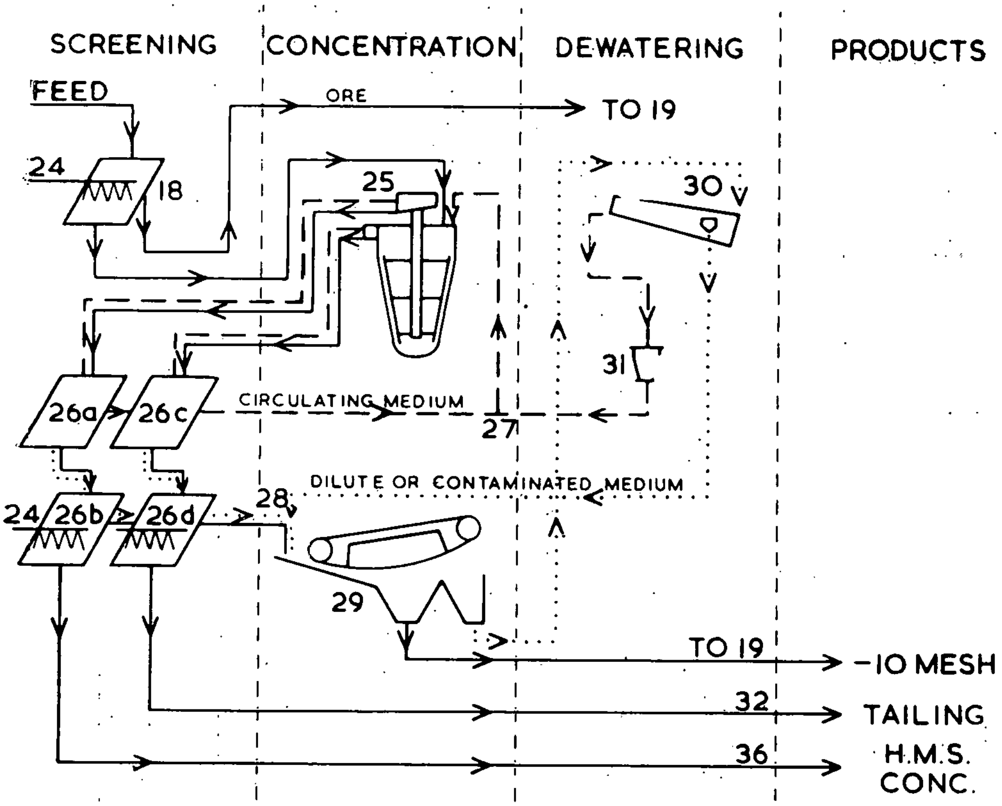

FIGURE 5:—Flow sheet of dense-medium separation process. See key for details.

rutile, ilmenite, and haematite, as well as the davidite itself.

Pilot plant investigations

Magnetic concentration, dense medium separation, and flotation were selected as being worthy of pilot plant scale examination.

The relative efficiencies determined in the course of pilot plant investigations during 1952 are summarised as follows:—

(i) Operation of pilot magnetic plant at Radium Hill.

Feed grade	4.08 lbs. U_3O_8 per ton
Concentrate grade	16.1 lbs. U_3O_8 per ton
Recovery	60 per cent.

(ii) Operation of dense medium separation plant at Radium Hill.

Feed grade	2.04 lbs. U_3O_8 per ton
Concentrate grade	7.53 lbs. U_3O_8 per ton
Recovery	87 per cent.

(iii) Operation of flotation section using dense medium separation product.

Feed grade	7.2 lbs. U_3O_8 per ton
Grade concentrate	18.3 lbs. U_3O_8 per ton
Recovery	85 per cent.

Developmental feed material was used, and the grades of ore referred to give no indication of the grades of ore and concentrate currently being achieved in full-scale production.

Magnetic concentration, because of its lower efficiency, was eliminated, and a large scale concentrator with dense medium separation followed by flotation of fine raw feed and dense medium concentrates was built. The operation of this plant is now described.

Summary of concentration processes

Raw ore is crushed to one-inch size in preparation for initial gravity separation. Following washing and screening, plus ten mesh material is delivered to a dense medium separation unit. The heavy concentrate is reduced to a size suitable for flotation, and the less dense tailings are not subjected to further treatment.

The minus ten mesh material is prepared for and joins the flotation circuit at a point appropriate to its uranium content.

Concentrates from the flotation process are filtered and despatched by rail to Port Pirie.

Crushing station

Coarse ore from the main-shaft vehicles is delivered at a rate which requires crushing to be carried out on two shifts per day, five days a week. Reduction is carried out in two stages by jaw and cone-type crushers in open circuit, with scalping of fines ahead of each stage. The section is designed for operation by one man.

All machines are stopped and started from a control desk and follow an interlocking pattern to ensure starting and stopping in correct sequence.

The crushing station product is of nominal one-inch size. Details of crushing and screening arrangements are set out in Figure 7.

Dense medium separation

A concrete bin having a capacity of 1,400 tons provides storage for crushed ore. Ore is fed from the bin through five discharge openings by remote controlled vibratory feeders to a 14-in. wide belt conveyor.

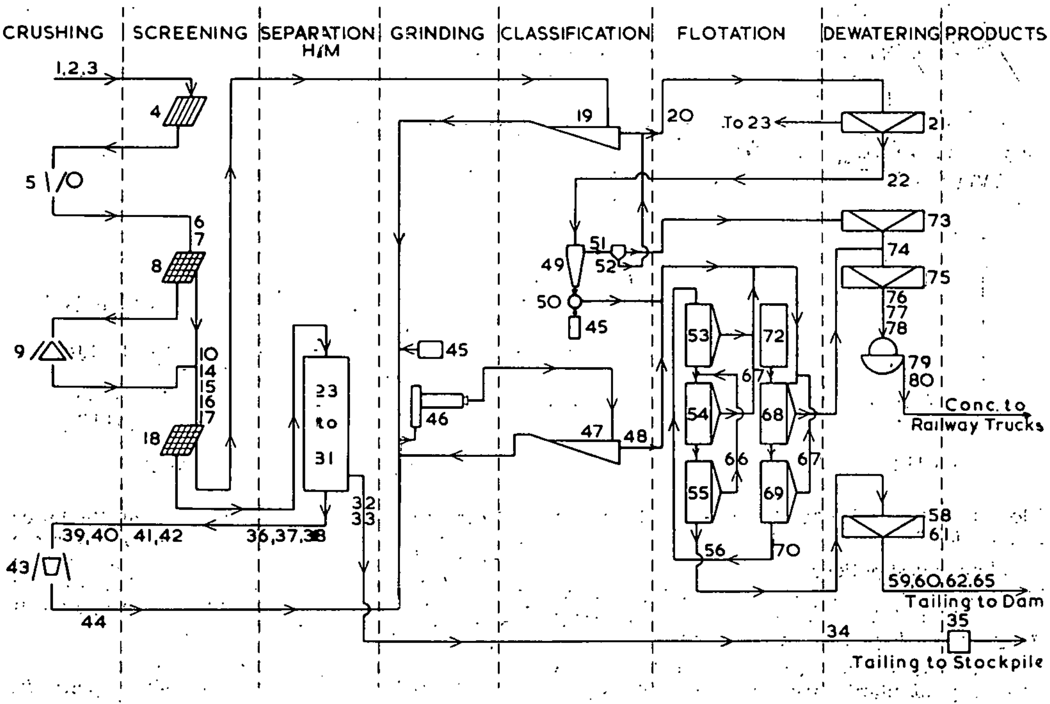

FIGURE 6:—Flow sheet of mineral dressing processes. See key for details.

A portion of the crushed feed is selected by washing and screening for immediate separation in an aqueous ferrosilicon suspension, having a specific gravity of 2.83. Provision is made for deep initial submergence of feed particles and separation is assisted by rotational agitation of the medium. At the optimum density of 2.83 approximately 91 per cent. of the uranium is recovered in the heavy material, which forms the concentrate from this initial separation. The ratio of concentration is 3.6:1.

Under normal operating conditions the sink materials contain very little uranium minerals.

Pre-washing and screening are necessary to avoid contamination of the suspension by the introduction of quantities of sub-colloidal material with consequent increase in viscosity and loss of particle velocity. Furthermore, small feed particles are not effectively separated because of their relatively low settling or rising velocities.

By controlling the density of make-up medium to the separation vessel a density differential is established between the top and bottom. This difference of 0.10 — 0.15 is used as a measure of the viscosity of the medium. Furthermore, particles which would otherwise tend to congregate in the "no man's land" between the well-defined sink and float products, respond to the changing density with a skipping action, which tends to accelerate their classification.

KEY TO PLANT UNIT NUMBERS

1	Coarse ore bin	Concrete, 100-ton capacity.
2.	Apron feeder	P & H Tools Ltd. 15ft. by 42in. wide, Mn - Steel flights, 5 h.p. speed range 3.6 to 15 f.p.m. through Reeves gear.
3.	Swing hammer regulator	Six hammers of 254lbs. each.
4.	Vibrating grizzly	Symons 42in. by 5ft., 7 bars, operature $2\frac{5}{8}$in.-$3\frac{7}{8}$in., 870 r.p.m., $7\frac{1}{2}$ h.p.

5.	Primary crusher	Traylor-Jaques Type "H" double toggle roller-bearing crusher, size 36in. by 24in. Set at $2\frac{3}{4}$in. (closed). Crusher r.p.m. 250, motor 80 h.p., squirrel-cage high-torque induction.
6.	Conveyor No. 1	24in. by 6-ply belt, 67ft. long C/C, 190 f.p.m., $18\frac{1}{2}$° slope, 7.5 h.p. motor.
7.	"Overband" magnetic separator	24in.; speed 200 f.p.m. motor 2 h.p., Magnet 2.8KW., 2-pole.
8.	Vibrating screens	Allis - Chalmers - Jacques "Low Head" 8ft. by 3ft. double-deck top plate punch plate, 1in. square aperture. Lower deck used as feed tray. Screen mechanism No. 2-D, 540 r.p.m., motor 7.5 h.p.
9.	Secondary crusher	Pegson-Telsmith "Gyrasphere" crusher, size 48in. $7\frac{1}{2}$in. receiving opening $\frac{3}{4}$in. set (closed). Speed: countershaft 550 r.p.m. Motor: 80 h.p. High-torque s.c. induction.
10.	Conveyor No. 2	18in. by 4-ply, 158 ft. centres, speed 240 f.p.m., slope 18°, motor 15 h.p. Delivering to Fine Ore Bin, Item 14.
11.	Exhaust fan	Sturtevant "Silentvane," 24in. impeller, 2250 r.p.m., motor 15 h.p.
12.	Dust collector	"Field" Wet Collector, Model 116, capacity 6000 c.f.m. at N.T.P.
13.	Collector underflow pump	$1\frac{1}{2}$in. Warman Standard Sand Pump, motor 5 h.p. Pumping to Dewatering Classifier, Item 19, Heavy Media Section.
14.	Fine ore bin	40ft. diameter by 42ft. reinforced concrete, 1400 tons live capacity. Five discharge openings.
15.	Feeders (2)	Jeffrey-Traylor, No. 3 HDTM, trolley-mounted, remote controlled.
16.	Conveyor No. 3	14in. by 4-ply belt, motor 3 h.p.
17.	Weigher	Blake-Denison Automatic.
18.	Feed preparation screen	Allis-Chalmers low head 8ft. by 4ft. with No. 2 1B mechanism, 950 r.p.m., $7\frac{1}{2}$ h.p. motor. Stainless steel screen cloth with 0.064in. by 0.352in. apertures.
19.	Dewatering classifier	Wemco SH 48in. by 23ft. 3in. single-pitch spiral, 5 h.p. motor.
20.	Overflow pump	Warman 2in. standard $7\frac{1}{2}$ h.p. motor.
21.	30ft. diameter thickener	2 h.p. motor.
22.	Pump to 6in cyclone	Warman 3/2 split casing. 7.5 h.p. motor.
23.	Heavy media circuit water tank	12,000 gallons.
24.	Water pumps	Two Harland Spiroglide 4/5, 40 h.p. motors.
25-31.	Wemco 3M. Mobil-Mill	(See flowsheet of heavy media unit.)
32.	Conveyor No. 8	14in. by 4-ply belt, 3 h.p. motor.
33.	Weigher	Blake-Denison continuous.
34.	Conveyor No. 8A	14in. by 4-ply belt, 5 h.p. motor.
35.	Tailing bin	250 tons, delivering to dump truck.
36.	Conveyor No. 4	14in. by 4-ply belt, 2 h.p. motor.
37.	Weigher	Blake-Denison continuous.
38.	Magnet	Permanent.
39.	H.M. concentrate bin	Reinforced concrete, 300 tons capacity, with three discharge openings.
40.	Feeder	Jeffrey-Traylor, as Item 15.
41.	Conveyor No. 5	14in. by 4-ply belt, 2 h.p. motor, magnet as Item 38.
42.	Weigher	Blake Denison continuous.
43.	Crusher	Symons Shorthead, set at $\frac{1}{4}$in., countershaft speed 580 r.p.m., 80 h.p. motor.
44.	Conveyor No. 6	14in. by 4-ply belt, 2 h.p. motor.
45.	Reagent feeder	Clarkson.
46.	Ball mill	Ruwolt 7ft. by 6ft. grate-discharge, with skiplap liners, charge 12 tons, make-up balls 3in. and 2in. 22 r.p.m., 175 h.p. motors.
47.	Classifier	Wemco SH, 36in. by 21ft. 6in. 2.9 h.p. motor.
48.	Overflow pumps	Two Warman 3/2 split-casing 5 h.p. motors.
49.	Cyclone	6in. cyclone with variable discharge.
50.	Agitator	5ft. 3in. by 3in. agitator 3 h.p. motor.

51.	Pump to cyclone	Warman 3 x 2 split-casing 5 h.p. motor.
52.	Multicyclone	Bank of 24 by 30 millimetre cyclones.
53.	Cells 1-4 of "A" Bank	Twelve No. 48 Agitair cells, 220 r.p.m., one 15 h.p. motor per 2 cells.
54.	Cells 5-8 of above	\
55.	Cells of 9-12 of above	
56.	Tailing pumps	Two Warman 4/3 split-casing 7½ h.p. motors.
57.	Splitter box	Delivering to 58 and 61.
58.	Thickener No. 3	25ft. diameter, 2 h.p. motor.
59.	Diaphragm pump	Dorrco 4 VM, 2.9 h.p. motor.
60.	Sand pump	Warman 1½in. standard 5 h.p. motor (delivering to 63).
61.	Thickener No. 5	30ft. diameter, 2 h.p. motor.
62.	Diaphragm pump	Dorrco 4 VM, 2.9 h.p. motor.
63.	Sampler	Geary-Jennings.
64.	Holding tanks (2)	11ft. diameter by 10ft., 5 h.p. motors.
65.	Tailing disposal pumps	One Warman 4/3 10 h.p. motor. One Warman 3/2 7½ h.p. motor pumping through 800ft. of 3in. pipe to tailing dam.
66.	"A" scavenger pumps	Two Warman 3/2 5 h.p. motors.
67.	Rougher pump	Two Warman 3/2 5 h.p. motors.
68.	Cells 5-8 "B" Bank	Details as for 53-55 (cleaners).
69.	Cells "B" Bank	As above; can be used as cleaners or roughers.
70.	"A" Bank feed pumps	Two Warman 3/2 5 h.p. motors used for pumping tailings from "B" Bank to "A" Bank.
71.	Concentrate pumps	Two Warman 3/2 5 h.p. motors.
72.	Cells —14 "B" Bank	Recleaners (when required).
73.	Thickener No. 1	15ft. Thickener for thickening slime concentrates.
74.	Slime concentrate pump	3 by 2 Warman 5 h.p. motor, pumping slime concentrates to concentrate holding tank.
75.	Concentrate thickener No. 4	25ft. diameter, 2 h.p. motor.
76.	Underflow pump	Warman 1½in. standard 5 h.p. motor.
77.	Sampler	Geary Jennings.
78.	Holding tanks (2)	11ft. diameter by 10ft. 5 h.p. motors.
79.	Filter feed pumps	Two Warman 1½in. standard 5 h.p. motors.
80.	Concentrate filter	Peterson 6ft. by 8-disc 2 h.p. motor.
81.	Weighbridge	50-ton capacity, makers Hawke & Co., S.A.
82.	Flotation blower	"Tornado" 30 h.p. motor.
83.	Filter blowers (3)	Roots type, 0.16 cu. ft. displacement, 2 h.p. motors.
84.	Vacuum pumps (3)	Chicago pneumatic type TVB 18in. by 7in., 350 r.p.m., 30 h.p. motors.
85.	Conveyor No. 7	Concentrate to trucks from filter. 14in. by 4-ply belt.
86.	Cooling water pump	Ajax 1¼in. 2 h.p. motor (vacuum pumps).
87.	Filtrate pumps (3)	Worthington 1½in. 2.9 h.p. motors. Three filtrate receivers, three moisture traps.
88.	Flotation circuit water tank	12,000 gallons.
89.	Water circulation pumps	Two Harland Spiroglide 5/6 20 h.p. motors.
90.	High pressure pump	Warman 3/2 15 h.p. motor.
91.	Flotation and grinding circuit water tank	4,800 gallons.
92.	Tailing dam water return pump	Ajax 1½in. pump.

In spite of washing under high pressure sprays, some fines are carried into the separation vessel, and new fines are produced by attrition. Continuous cleaning is effected by exposing the medium to a magnetic field. The slightly magnetic ferrosilicon is separated from non-magnetic contaminants, densified, demagnetised, and returned in a highly dispersed condition to the separation vessel.

Sizing analyses of the two grades of ferrosilicon used are:

	100-mesh grade	65-mesh grade
+ 65	0.1%	1.2%— 1.3%
— 65+100	0.1%	4.8%— 5.2%
—100+150	0.3%— 0.5%	10.5%—10.8%
—150+200	4.5%— 5.5%	13.0%—13.5%
—200+325	24.0%—26.0%	27.4%—27.8%
—325	71.0%—68.0%	43.1%—41.4%

The ground alloy has a specific gravity of 6.5 to 6.8, is resistant to corrosion and caking, and its silicon content is between 14 per cent. and 16 per cent. Consumption of ferrosilicon is 0.38lb. per ton of +10 mesh material treated.

Certain elements of the waste rock have a high ferro-magnesian content and reach a specific gravity higher than 2.83. This causes some dilution of the concentrates and where relatively large quantities are involved the average density of the suspension is temporarily adjusted.

Consideration is being given to the introduction of a further stage of dense medium separation with a view to increasing the grade of concentrate to a point sufficiently close to that of the final product to reduce the quantity of material requiring flotation treatment.

Fine grinding

Two uranium-rich products leave the dense medium separation section.

(i) Minus 10 mesh material from initial screening, and

(ii) Plus 10 mesh concentrate.

The minus 10 mesh material is deslimed in a spiral classifier and the granular portion joins the plus 10 mesh concentrate for fine grinding in ball mills prior to flotation. Closed circuit classification is provided by 36in. spiral type classifiers, and the ground product has the following average sizing analysis.

+60 mesh	1.0 per cent.
+100 mesh	8.5 per cent.
+150 mesh	12.0 per cent.
+200 mesh	13.8 per cent.
—200 mesh	65.7 per cent.

Two-inch and 3in. diameter cast iron balls in a 1 : 1 ratio are used within a manganese and chrome cast steel shell. Circulating load is of the order of 250 per cent.

To encourage their emulsification, flotation reagents are added at the ball mills.

Flotation

Flotation reagents developed for use with davidite ore are

TABLE OF FLOTATION REAGENTS

Flotation Reagent	Function	Point of addition to circuit	Quantity used per ton flotation feed	Landed cost per lb.	Remarks
Linseed fatty acid	Collector and emulsion stabilizer	Ball mill	2.5	1/3	By-product of linseed oil manufacture. Contains linoleic acid, linolinic acid, and oleic acid.
Peltogen	Emulsifier and secondary collector	Ball mill	1.5	2/1	Sulphonated sperm whale oil.
Cresylic acid	Coupling agent emulsion stabilizer	Ball mill	0.5	1/11	Used to stabilize emulsion and to couple collectors.
Diesel fuel oil	Froth modifier. Increases contact angle	Ball mill and flotation cells	10lb.	2/8	6lb. of fuel oil added to ball mill and 4lb. of fuel oil added to roughers and scavenger froths to control excess frothing.

Laboratory work suggested that the use of sorbitan monoleate and a polyoxyethylene derivative P100 would give closer control of frothing and lead to an overall reduction in reagent cost. Plant scale tests have not yet conclusively proved this to be the case.

Some research has been directed towards the differential flotation of davidite from the associated iron titanium minerals. No usable process has yet been developed.

Controlled aeration and mechanical agitation is provided in Agitair Cells Class 84. Impellor and guide vein wear is excessive unless stainless steel or neoprene is used. The presence of fuel oil prohibits the use of rubber protection.

Settling agents, such as Cyanamide S3000 and Dow Separan, are used to encourage flocculation in the dewatering stages which follow flotation. The quantity of flocculating agent used is related to ambient temperatures.

Recovery in this section is 92 per cent., with a ratio of concentration of 3 : 1.

Operational data are:

	Per ton of flotation feed
Power	10.4 kWl
Linseed Fatty Acids	2.5lb.
Fuel Oil	10.0lb.
Peltogen	1.5lb.
Settling Agent	0.01lb.
Cresylic Acid	0.5lb.

Classification of very fine material

Fine material over-flowing from the spiral dewatering classifier used to deslime minus ten mesh materials is thickened and delivered to a two-stage Dutch State Mine cyclone process. Initial separation takes place in a 6in. cyclone at a pressure of 30-35lb./sq. in. Underflow is conditioned with flotation reagents and is delivered to the flotation section. The fine overflow pulp is fed to a bank of neoprene lined 30mm. cyclones operated at a pressure of 20lb./sq. in. The fine overflow bypasses the flotation cells, is dewatered, and forms part of the final concentrate.

Disposal of products

Tailings from the dense medium separation section are stock-piled or loaded directly from conveyors into railway wagons for ballast purposes.

Tailings from the flotation circuit are delivered to a square holding dam 400ft. by 400ft., provided with a central discharge. Recovered water is re-cycled.

Final concentrate is thickened, filtered, loaded into open rail trucks, sampled for moisture and uranium content, and despatched to Port Pirie.

The Regional Distribution of Uranium Occurrences, Northern Territory, Australia

By B. P. Walpole*

Uranium deposits in the Northern Territory have been found in Precambrian rocks, ranging from Archaean to Upper Proterozoic in age. The important deposits are confined to a particular facies assemblage within the Lower Proterozoic sequence, and to rocks which belong to the lower part of the Upper Proterozoic (Lower Adelaidean) succession. None of the major deposits can be related to granic intrusions and all are Upper Proterozoic in age. Some can be related to Upper Proterozoic volcanic activity, but the remainder, and particularly those at Rum Jungle, are not associated with volcanic rocks.

Introduction

The first recorded discovery of radioactive mineralization in the Northern Territory was made in 1947, when samarskite was found in the pegmatite of the Butcher Bird mine in the Harts Range. The discovery of the Rum Jungle deposits in 1949 provided an impetus which has since led to the location of more than 70 radioactive prospects in the Territory. Only a few of them have developed to the stage of production. Very few have reserves which allow them to be classed as economic mining propositions, and all these latter occur in the Rum Jungle and South Alligator River districts of the Katherine-Darwin region. All the known prospects occur in Precambrian rocks.

This paper will not describe the different deposits in detail. They will be subdivided into geographical groups and, where necessary, into different types of deposits within any particular area.

General geology

The distribution of the Precambrian rocks of the Northern Territory is shown in Figure 1. It has long been recognised that these rocks can be considered in three major divisions, here referred to as Archaean, Lower Proterozoic and Upper Proterozoic. The divisions were based primarily on degree of metamorphism, the presence or absence of granite intrusions, and the degree of folding, and all data gained in recent years emphasise the correctness of these divisions, tentative subdivision of the Lower Proterozoic into Lower Proterozoic and Upper/Lower Proterozoic and of the Upper Proterozoic into Lower and Upper Adelaidean is now possible, but it is certain that such subdivision will be modified as more data become available. The salient features of the rocks in these major divisions, and their subdivisions and their areal distribution are summarized in Table 1.

Archaean

Joklik (1955) has described a suite of Archaean rocks from the Harts Ranges of Central Australia. These may be considered as typical. The sediments are very highly metamorphosed, the granite intrusions are gneissic, and granitization and migmatization are common features.

In direct contrast to the intense metamorphism which these rocks have undergone, structural deformation is not severe. This feature has not been widely recognised in the past, but there are very good examples in the Northern Territory which may be noted. One such area is the Harts Range. Here the main structural elements are fairly simple domes and basins, the flanks of which dip at angles averaging only about 40°. Dips of 20° are common. The writer is of the opinion that most of the major faults in the Harts Range area are not Archaean in age, but were developed late in Precambrian time. A second example is the Oenpelli area of the Katherine-Darwin region, where again the structural deformation is not intense, but the degree of regional metamorphism is high.

The Archaean rocks of the Northern Territory are singularly lacking in known economic deposits of base metals in general and of uranium in particular. There are a few minor occurrences of copper, lead and other minerals, but major discoveries of base metals have not yet been made in these rocks.

Uranium discoveries in the Archaean of the Northern Territory are confined to a few occurrences of samarskite in the mica-bearing pegmatites of the Harts Range, and to small veins of pegmatite containing betafite in the Mt. Cavenagh area on the Northern Territory—South Australian border.

Lower Proterozoic

The Lower Proterozoic rocks of the Northern Territory unconformably overlie the Archaean metamorphics, and crop out in the Tennant Creek area, and in the Katherine-Darwin-Victoria River region. The sediments are geosyn-

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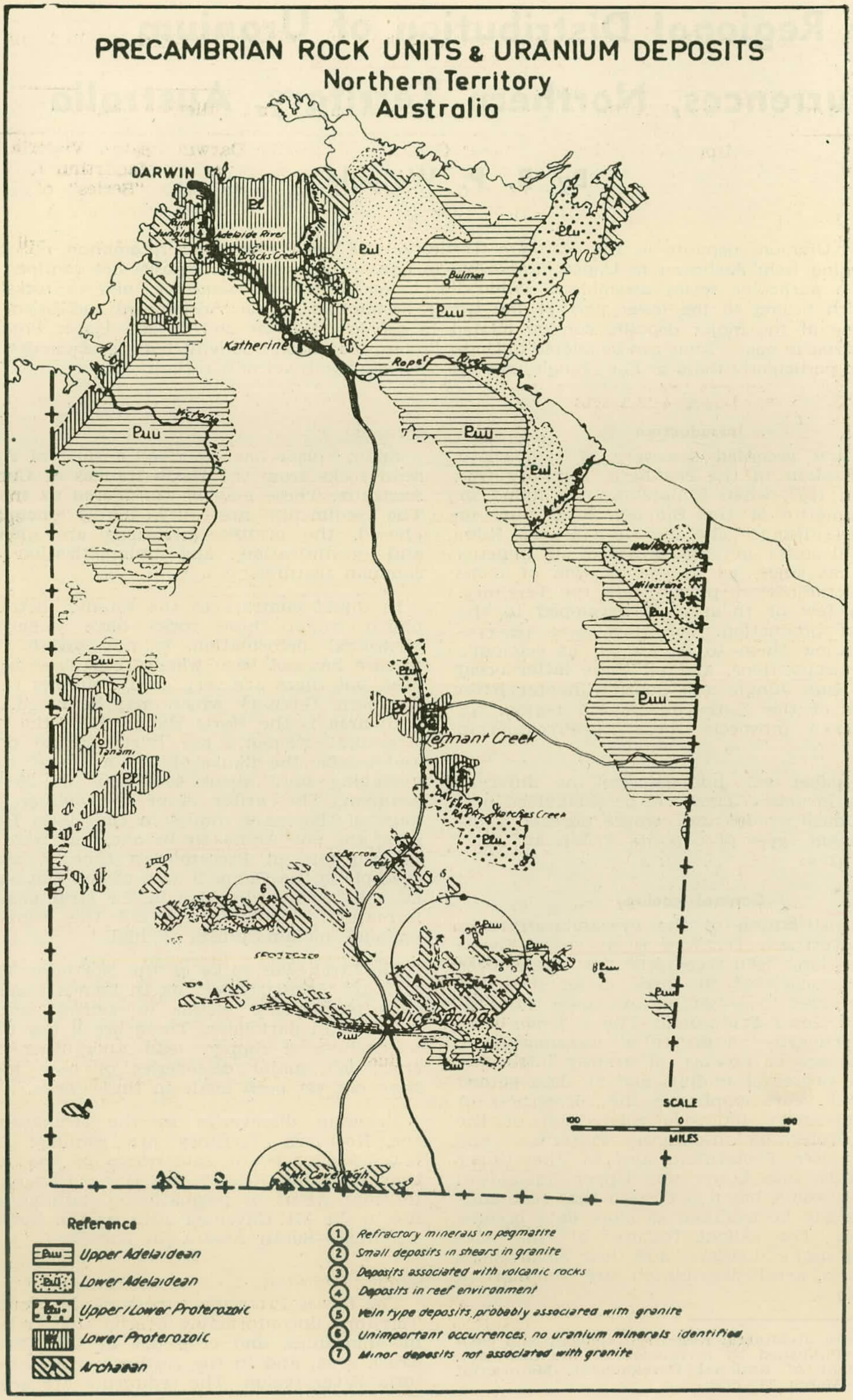
PRECAMBRIAN ROCK UNITS & URANIUM DEPOSITS
Northern Territory
Australia
DARWIN
Rum Jungle
Adelaide River
Brocks Creek
Katherine
Roper River
Bulman
Victoria River
Wollogorang
Milestone
Tanami
Tennant Creek
Hatches Creek
Barrow Creek
Mt Doreen
Harts Range
Alice Springs
Mt Cavenagh
Puu
Pul
Pl
A
SCALE
100 0 100
MILES
Reference
Upper Adelaidean
Lower Adelaidean
Upper / Lower Proterozoic
Lower Proterozoic
Archaean
① Refractory minerals in pegmatite
② Small deposits in shears in granite
③ Deposits associated with volcanic rocks
④ Deposits in reef environment
⑤ Vein type deposits probably associated with granite
⑥ Unimportant occurrences, no uranium minerals identified
⑦ Minor deposits not associated with granite

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TABLE 1: — NOMENCLATURE AND DISTRIBUTION OF PRECAMBRIAN ROCKS OF THE NORTHERN TERRITORY.

Main Division	Sub-Division	Distribution and Salient Features
UPPER PROTEROZOIC	Upper Adelaidean	Tolmer Group of Katherine-Darwin Region, Victoria River Group, rocks of Bulman area, part of MacArthur River and Constance Range areas, Pertatataka "Series" of Central Australia. Unmetamorphosed rocks. Both arenaceous and argillaceous sediments. Coarse clastics and volcanics subordinate. Algal dolomitic bioherms and biostromes common. Rocks deposited in broad intracratonic basins, folding commonly very gentle and marginal fracturing not pronounced. No granite intrusion. At least one unconformity within this subdivision.
	Lower Adelaidean	Katherine River group, of Katherine-Darwin Region, part of sequence in MacArthur River and Nicholson River areas, Pertaknurra "Series" of Central Australia. Unmetamorphosed rocks, commonly coarse clastic sediments with interbedded volcanics. Dolomitic sediments in some places, particularly MacArthur River area. Rocks deposited in intracratonic basins. Folding gentle but rocks strongly fractured on margins of basins where steep dips due to drag on faults or modified depositional features are common. At least one unconformity within this subdivision. No granite intrusion.
	Unconformity	
LOWER PROTEROZOIC	Upper/Lower Proterozoic	Rocks of east coast of Arnhem Land, Davenport Ranges and Ashburton Ranges. Typical eugeosynclinal sedimentation. Well developed volcanic component. Sediments are commonly clastic and include greywacke, conglomerate, coarse sandstone, etc. Only slightly affected by regional metamorphism. Folded, faulted and intruded by granite and hypabyssal rocks. Note: The literature contains many references to quartzite in the Davenport and Ashburton Ranges. These rocks are not quartzites of metamorphic derivation.
	Major Unconformity	
	Lower Proterozoic	Rocks of Pine Creek Geosyncline, Warramunga Group of Tennant Creek area, rocks of Tanami-Granites area. Geosynclinal sedimentation. Regional metamorphism is commonly low-grade but main bulk of sediments can be considered as unmetamorphosed. Mica and andalusite schists developed in regional shear zones in Katherine-Darwin and Hall's Creek regions. Main metamorphism in all areas is due to contact effects of granite intrusions. Sediments are folded and extensively faulted and intruded by acid and basic igneous rocks. Granites are commonly massive and unstressed discordant bodies.
	Unconformity	
ARCHAEAN		Includes Arunta Complex of Central Australia, Nanambu and Hermit Hill Complexes and Magela Creek Metamorphics, Katherine-Darwin Region, rocks of possible Archaean age in the Cape Arnhem area, Northern Territory. Features are high-grade regional metamorphism, granitization and migmatization, gneissic granites, etc. In most areas where structural studies have been made, structural deformation by folding is not particularly intense.
	Major Unconformity	

clinal, are folded and faulted, and intruded by acid and basic igneous rocks. They are commonly little affected by regional metamorphism, and such metamorphism as is present can in most places be attributed to the contact effects of granite intrusions or is localized in shear zones.

Most of the earlier workers loosely refer to these rocks as "metamorphics." However, all the Lower Proterozoic rocks seen by the writer in the Northern Territory are characterized by only a very low degree of regional metamorphism. The granite intrusions are commonly massive discordant bodies, with many siliceous contact aureoles. Some have been intruded into the cores of domes, but these too are discordant in detail.

TENNANT CREEK AREA

In the Tennant Creek area, the Lower Proterozoic sediments are moderately folded and intruded by granite and porphyry.

The main lithologies present are sandstone, ripple-marked sandstone, siltstone, sandy shale, greywacke, mudstone, and "tuffaceous" sandstone. The tuffaceous sandstone has not yet been positively identified as such, and may possibly be greywacke. Limey and carbonaceous rocks are noticeably absent.

The area is extensively mineralized. At Tennant Creek, gold and copper mineralization is associated with hematite and magnetite bodies. Tungsten and cobalt are minor accessories in some places. The mineralization is clearly of hydrothermal origin.

Only one occurrence of uranium mineralization has been found in the area. This is a minor deposit localised on a shear in granite, in the Mosquito Creek area, about 50 miles north-west of Hatches Creek. Three other radioactive occurrences have been reported from near Tennant Creek, but all are small, and no uranium minerals have yet been identified from them.

VICTORIA RIVER AREA

The Lower Proterozoic rocks in the Victoria River area are a southwesterly extension of those that crop out in the Katherine-Darwin region. In the Victoria River area they are mainly sandstone, and for all practical purposes are not mineralized.

KATHERINE-DARWIN REGION

The Lower Proterozoic rocks of the Katherine-Darwin region crop out over an area of about 14,000 square miles. The sediments were deposited in a composite trough known as the Pine Creek Geosyncline in which a number of episodes of sedimentation or facies assemblages, which overlap in space and in time, can be recognised. The first episode is miogeosynclinal, and arkose, conglomerate, sandstone and siltstone, and algal reef structures characterize the shelf environment. The transitional or slope environment is typified by quartz greywacke, sandstone, and pyritic carbonaceous siltstone. Pyritic carbonaceous siltstone, chert, and dolomitic rocks characterize the trough environment. The sediments were laid down in an asymmetrical trough with a narrow shelf area and a sharp fall away on the western side (Rum Jungle area) and a gentle slope on the eastern side. Reef structures are present on the western shelf edge at Rum Jungle, but are found on the eastern margin. There is no traditional zone on the western side and here the pyritic, carbonaceous, and dolomitic sediments of the trough environment directly overlie the shelf sediments. Most of the sediments are regarded as having been derived from the east and north.

The succeeding episodes are attributed to the influence of basement faults and the second phase of sedimentation was entirely from the west. The rocks are eugeosynclinal and comprise greywacke, sandstone, conglomerate, siltstone, greywacke siltstone, and volcanics. They commonly show well-developed sedimentary structures indicative of density-current deposition. In some areas they disconformably overlie the earlier sediments, but in the centre of the geosyncline the two assemblages are interfingered.

A third episode resulted from downfaulting of a secondary trough on the eastern side of the geosyncline, and the entrapment in this trough of later sediments of the miogeosynclinal episode. The western margin of this trough is along the South Alligator Valley, and is marked by strong faulting and by a discontinuous reef facies.

The last stage of sedimentation is in part platform and in part taphrogeosynclinal. The rocks are mainly sandstone, lapping back into the old source area on the western side of the geosyncline and also filling a graben structure which extends south-west to beyond the Victoria River area.

The rocks are extensively faulted, but in general the folding is not intense. Most of the faults can be related to a regional tensional stress condition. The sediments are intruded by basic sills and dykes, some of which were emplaced before the folding. Granite was intruded after the sediments were folded.

Mineralization is widely scattered within the Pine Creek Geosyncline, but the great majority of the deposits are small. Quartz-cassiterite, quartz-gold, and small epigenetic base-metal deposits associated with shears or quartz veins are common. Most of these are within the eugeosynclinal sediments of the second episode, and are clearly associated with the granite bodies of the area.

Uranium occurrences have been discovered in a number of places within the region. These will be discussed later.

Upper/Lower Proterozoic

These rocks crop out in the Davenport Range, Ashburton Ranges, and the eastern Arnhem Land area. They are eugeosynclinal, and consist mainly of greywacke, conglomerate, and sandstone with alternating volcanics in some places. They are moderately folded, and are intruded by granite and porphyry. Regional metamorphism is very weak. The rocks do not contain any known uranium deposits, although radioactive volcanic rocks have been discovered in a number of places. Quartz veins containing tungsten and gold are present in the Davenport Ranges.

Upper Proterozoic

Lower Adelaidean

The Lower Adelaidean rocks crop out in the Wollogorang and MacArthur River areas, and the western section of the Arnhem Land Plateau in the Katherine-Darwin region. In the Calvert Hills area and the Katherine-Darwin region, these rocks are mainly clastic sediments with intercalated volcanics. Some dolomitic rocks are present in the Wollogorang area. The rocks in the MacArthur River area are mainly dolomitic, and their correlation with the Lower Adelaidean rocks is tentative only.

The rocks were deposited in broad intracratonic basins. They are only very gently folded, are not metamorphosed, and are not intruded by granite. The Pertaknurra "series" of the Alice Springs area, composed of clastic sediments and algal carbonate rocks, is also probably part of the Lower Adelaidean sequence.

The Lower Adelaidean rocks are extensively fractured and in places are intruded by dolerite dykes. Minor lead, zinc, and copper mineralization occurs in these rocks in the MacArthur River area. Copper mineralization is associated with the volcanic rocks in the Wollogorang area.

Upper Adelaidean

Uranium mineralization has been found in both the Wollogorang and Katherine-Darwin areas.

The Upper Adelaidean rocks crop out in the shallow basin structures in the Katherine-Darwin, Victoria River, Bulman, MacArthur River, and Calvert Hills areas. The rocks consist mainly of fine to medium-grained arenites, dolomites, and lutites. Volcanics are present in some places, but are of local importance only. The rocks are not metamorphosed, and are only gently folded. Basic sills intrude the rocks in some places, but granite intrusions are not known. Minor lead, zinc, and copper mineralization occurs in these rocks in the Bulman area, and iron ore deposits in the Roper River area, but in most places the rocks are not mineralized. The Pertatataka "series" of the Alice Springs area can be equated to the Sturtian and Marinoan Series of the Adelaidean System, and is therefore also Upper Adelaidean. No uranium discoveries have yet been made in the Upper Adelaidean rocks of the Northern Territory.

Uranium deposits

Of the five main stratigraphic subdivisions recognised in the Precambrian rocks of the Northern Territory, important uranium deposits have been discovered in only two. The distribution of the occurrences is summarized in Table 2.

TABLE 2:—SUMMARY OF URANIUM OCCURRENCES

Stratigraphic Division	Types of Deposit	Remarks
Upper Adelaidean	No deposits yet located.	Apparent association with volcanics
Lower Adelaidean	Small vein-type deposits in volcanics and sediments of Wollogorang area; and in shears at contact of Lower Proterozoic and lower Adelaidean rocks of South Alligator River area. Secondary deposits in volcanic rocks in Katherine area.	
Upper/Lower Proterozoic	No deposits yet located.	Eugeosynclinal sediments
Lower Proterozoic	No deposits in eugeosynclinal sediments of Tennant Creek area. A few small vein-type deposits in eugeosynclinal sediments of Katherine-Darwin region and in granites. All major deposits in Northern Territory are in carbonaceous rocks or mixed carbonaceous and dolomitic sediments (miogeosynclinal sediments) of the Katherine-Darwin region.	Significant lack of economic deposits in eugeosynclinal sediments.
Archaean	Small deposits of refractory minerals in pegmatites.	No deposits of economic significance yet located.

Deposits in Archaean rocks

These deposits consist of small occurrences of refractory minerals, such as betafite and samarskite, in pegmatite veins. Deposits of this type have been found in the Harts and Strangways Ranges, where samarskite occurs as a minor accessory mineral in pegmatite bodies which have been or are being mined for their mica content. In the Mt. Cavenagh area, very small pegmatite veins containing betafite have been discovered.

None of these occurrences are of economic interest. There are no known davidite deposits such as those in the Archaean rocks of South Australia.

Deposits in Lower Proterozoic rocks

The deposits which have been found in the Lower Proterozoic rocks can be grouped into three main types. These are as follows:

(i) Deposits in granitic rocks: Some small uranium deposits have been found in the Edith River area of the Katherine-Darwin region, and at one locality in the Mosquito Creek area of Central Australia. They are localized on fractures within granite. They are all small, and testing has not indicated any deposit of economic importance. Most are low grade, and contain surface or near-surface showings of secondary minerals.

(ii) Deposits in eugeosynclinal sediments: These have been found in only a few places in the Katherine-Darwin region. They include deposits such as those at Adelaide River and George Creek. They are all small, and of doubtful economic importance. All are vein-type deposits localized on shears. The Adelaide River and George Creek deposits contain pitchblende. They occur in rocks which contain a large number of quartz veins bearing gold, cassiterite, copper, lead, and other minerals. These are clearly associated with granitic intrusions, and in the writer's opinion the uranium deposits have a similar origin.

(iii) Deposits in miogeosynclinal sediments: Miogeosynclinal rocks of Lower Proterozoic age in the Northern Territory occur only in the Katherine-Darwin region. Uranium deposits within these rocks include the Rum Jungle and South Alligator deposits, which are of known economic importance, and a few minor deposits of no economic significance in the Brocks Creek area. All occur in carbonaceous rocks, commonly with associated dolomitic sediments. At Rum Jungle the main deposits are localized by a system of faults, but are also stratigraphically controlled by the contact of the Coomalie Dolomite and the overlying Golden Dyke Formation. The Coomalie Dolomite is a reef facies containing algal bioherms and dolomitic sediments. The Golden Dyke Formation consists mainly of pyritic carbonaceous and dolomitic rocks. The deposits occur within the Golden Dyke Formation both at Rum Jungle and at Brocks Creek.

In the South Alligator River area, the deposits are localized in shears which cut the Lower Proterozoic Koolpin Formation and the overlying Upper Proterozoic (Lower Adelaidean) rocks. The mineralzation occurs in both the Lower and Upper Proterozoic rocks, but the main deposits are in the Lower Proterozoic sediments. The occurrences are commonly marked at the surface by spectacular splashes of secondary uranium minerals. Most pass into pitchblende, but none have yet been proved to extend beyond a fairly shallow depth. At the Palette mine, pitchblende occurs at the surface. The rocks of the Koolpin Formation belong to the miogeosynclinal sequence of the Pine Creek Geosyncline. They are mainly carbonaceous sediments, with a prominent member consisting of pyritic and carbonaceous siltstone with bands, lenses and nodules of silicified dolomite. Discontinuous algal bioherms occur in places at or near the base of the Formation. The overall composition of the rocks is therefore very similar to that at Rum Jungle.

Deposits in Lower Adelaidean rocks

Uranium deposits in the Lower Adelaidean rocks of the Northern Territory have been found in the South Alligator River, Katherine, and Wollogorang areas. The South Alligator deposits have already been briefly discussed. The deposit near Katherine consists of a small shoot of secondary minerals in volcanics. The Wollogorang deposits are commonly small vein-type deposits, which occur in minor shears in volcanic rocks or in shears at the contact between volcanic rocks and clastic sediments. Some of these occurrences contain pitchblende, but none have yet proved to be sufficiently large to support a profitable mining operation.

Summary of regional pattern of distribution of uranium occurrences

Any attempt to generalize regarding the distribution of mineral occurrences is necessarily subjective, because it can only take into account the known deposits. Such generalizations can be useful or misleading, depending entirely on how they are applied, and the pattern indicated here must therefore be considered in that light.

The pattern which emerges from a study of the distribution of the known occurrences is as follows. Very few deposits are known in the Archaean metamorphics, Lower Proterozoic eugeosynclinal sediments, and Upper Lower Proterozoic rocks. Such deposits as are known are of doubtful or no economic significance. None are known in the Upper Adelaidean rocks.

None of the deposits in granitic rocks is of economic importance. Of the deposits which occur in the Lower Adelaidean rocks, only those in the South Alligator River area have yet been shown to have economic dimensions, and here the main mineralization is in the underlying Lower Proterozoic sediments.

All the major deposits in the Northern Territory occur in miogeosynclinal carbonaceous and dolomitic sediments of the Golden Dyke or Koolpin Formations in the Katherine-Darwin region. The important deposits occur in the Rum Jungle and South Alligator River areas.

Origin of deposits

Only the salient features which bear on the origin of the main uranium occurrences of the Northern Territory will be noted here. The Archaean and Upper/Lower Proterozoic occurrences will not be discussed.

Although very little has been published on the uranium deposits of the Northern Territory, and in particular on their origin, the matter has been a constant topic of discussion amongst geologists who have examined these deposits. It is also true to say that most of the deposits have been tacitly considered by most geologists as having been derived from the granitic rocks of the different areas where the deposits occur, or where such intrusions are absent, to volcanic activity. In the Katherine-Darwin region, the high radioactivity of the numerous granitic intrusions has been used as an argument for association of these intrusions with the uraniferous bodies which occur nearby. This has been the case particularly with the Rum Jungle deposits. The contribution of radioactivity in granite from sources other than uranium has been to a large extent ignored.

The writer does not question the origin of deposits such as those in the eugeosynclinal sediments of the Pine Creek geosyncline. These are in a mineral environment marked by very large numbers of similarly disposed occurrences of other minerals, the majority of which are clearly associated with granite intrusion. Nor does he question the association of volcanic rocks and uranium occurrences in the Northern Territory section of the Wollogorang area. Granite crops out south of the main group of deposits—those at Milestone—in the Wollogorang area, but does not intrude the sediments in which the Milestone occurrences are localized.

In the Katherine-Darwin region the deposits in the Rum Jungle and Alligator River areas and the minor deposit at Brocks Creek cannot be related to the granite intrusions which crop out in close proximity to the mineralzed occurrences in each case.

At Rum Jungle, the deposits occur in the Lower Proterozoic sediments. These are intruded by the Rum Jungle and Waterhouse granites, both of which are markedly radioactive. The main deposits, however, are localized by a fracture system—known as the Giants Fault System — which displaces the granite laterally about 2½ miles. The Giants Fault itself extends over 100 miles south-west from Rum Jungle, and is obviously due to an Upper Proterozoic movement. The Rum Jungle and Waterhouse granites are comagmatic. Both have been dated, and preliminary determinations based on the strontium/rubidium method carried out by Professor P. Hurley of the Massachusetts Institute of Technology place their ages at between 1700 and 1800 million years.

According to Holmes' time scale, this would place the granite intrusions and the intruded sediments as Archaean in age, but as the question of Precambrian subdivisions is at present under serious review throughout the world, it is preferred here to use Lower Proterozoic until some finality is reached in this matter.

Preliminary chemical analyses of Rum Jungle pitchblende indicate an age of about 650 million years. The age of the pitchblende has yet to be checked by determination of the lead isotope ratios, but it is confidently predicted that the error will not be greater than 20 per cent. The Rum Jungle ore cannot, therefore, be related to the granite intrusions.

At Brocks Creek the same features apply. Pitchblende from the Fleur de Lys mine has been dated by Professor Birkett-Clewes (University of Western Australia) at 700 million years. The nearby granite masses have all been dated at between 1,700 and 1,800 million years.

In the South Alligator River area, the granite has been dated at 1,700 to 1,800 million years, and the uranium ores at between 550 and 650 million years. Here there is also clear evidence of an unconformity between the granite and the mineralized Upper Proterozoic rocks.

The South Alligator deposits occur in a volcanic environment, and it can be argued that the presence of the volcanics indicate that a hydrothermal source is therefore available for the uranium ores. This argument, however, cannot be applied to either the Rum Jungle or Brocks Creek deposits. The presence of a granite at depth to provide a "telemagmatic" source for the Rum Jungle and Brocks Creek deposits is at best a misty hypothesis, and is not considered to be a tenable argument to support a theory of epigenetic origin of the uranium ores in either the Rum Jungle or Brocks Creek areas.

The deposits in the Rum Jungle, Brocks Creek and South Alligator areas have some points in common. They all occur in similar sediments—carbonaceous rocks with associated dolomitic sediments. Pyrite of syngenetic origin is widely distributed throughout these rocks. The Rum Jungle and South Alligator deposits both occur in areas marked by algal bioherms. Few of the bioherms are coincident with or close to the actual deposits, but the overall sedimentation is nevertheless clearly that of a reef environment. The implication of such an environment has been discussed by Condon and Walpole (1955). Although some of the regional geological data presented in that paper have since been modified, and the interpretation of local detail—particularly in regard to the origin and age of some breccias—has been shown by the writer to be erroneous, the general principles outlined are still considered here to be tenable.

It is suggested here that a clue on the origin of these deposits lies in the fact that they occur in an environment which, by all considerations, must be regarded as favourable for the deposition of syngenetic metals. Most geologists accept without question the presence of syngenetic pyrite, which is widespread in the areas under consideration, and yet find it difficult to accept syngenesis of other base metals. If sulphate-reducing bacteria are suggested as a precipitating agent for such metals, a common opinion is that such bacteria did not necessarily exist in Precambrian time. Yet there is good evidence of life in the form of algal and other fossil forms, and surely the reducing processes which must apply in the precipitation of pyrite can also apply to other metals. Current research being carried out in conjunction with the Bureau of Mineral Resources by Professor Baas-Becking of C.S.I.R.O. (personal communication) has shown that very great concentration of base metals can be produced by bacterial agencies. Perhaps other mechanisms are also possible.

It is true that the main uranium ore occurrences in the Northern Territory occur in the Pine Creek geosyncline and in a shelf environment, and therefore in a depositional area which can be favourable for the deposition of syngenetic metals.

The majority of the uranium occurrences in the Northern Territory are Upper Proterozoic in age, and are unrelated to granitic intrusions. An empirical association of uranium and volcanic rocks is present in the Wollogorang and South Alligator River areas, but the close similarity of the South Alligator Lower Proterozoic host rocks and those at Rum Jungle—where there is no volcanic association—might perhaps be significant.

Most ore bodies require a localizing structure, and the problem of how ore-bearing solutions or fluids penetrate—apparently through miles of country rocks—to enter these structures has, in most cases, not been satisfactorily answered. The fact remains however, that a mechanism whereby ore minerals can migrate must exist. At Rum Jungle a large volume of base metals in mineable concentrations is present without an obvious hydrothermal source, and in rocks which have been little affected by regional metamorphism. Only very small concentrations occur at Brocks Creek, but here again there is no obvious hydrothermal source and little or no regional metamorphism.

In regard to the emplacement of the Rum Jungle deposits, the Giants Fault system can be considered to have constituted a zone of low potential energy in contrast to the high potential energy environment of the surrounding rocks at the time of faulting. This may have caused migration of metals into the fault zone. Whether the metals were introduced into the sediments by the Rum Jungle granite (which to the writer seems unlikely) or whether they were of syngenetic origin, is a debatable point. It is perhaps significant that similar, favourable rock types, intruded by highly radioactive granites, with similar favourable structures, but present in deeper sections of the geosyncline away from the shelf edge, have yet to be shown to contain economic deposits of uranium ore.

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The Flotation of Uranium Minerals, with Particular Reference to Uraninite

By John S. Carr*

Introduction

A major difficulty in the treatment of uranium-bearing materials is the very low uranium content, usually only a few tenths of one per cent, and often in the form of several minerals. The mineral beneficiation problem presented by such materials is largely concerned with the attainment of a satisfactory minimum ratio of concentration, and at the same time a low assay product for rejection. This problem is complicated by the wide variation in character and geologic environment of uranium deposits (Everhart 1954). In terms of beneficiation characteristics, the associated minerals may be largely siliceous or mainly calcite, and appreciable amounts of sulphide minerals or organic materials may be present. Such constituents may be present in economic quantities or in amounts sufficient to interfere with the separation of the uranium minerals. Some of these features will undoubtedly continue to be regarded as adverse until more fundamental information is available concerning the beneficiation characteristics of the uranium minerals in particular.

As has been emphasized elsewhere (Frondel and Fleischer 1955; Gaudin 1956), uranium minerals fall into three classes, viz., simple oxides—uraninite (U^{4+}) and pitchblende (U^{4+} and U^{6+}); mixed oxides (U^{4+})—brannerite and davidite; and compound salts (U^{6+})—phosphates (autunite, parsonsite, saléeite, torbernite), vanadates (carnotite), and silicates (coffinite).

This discussion is concerned principally with the flotation behaviour of the simple oxide minerals, uraninite and the colloform variety, pitchblende (Carr 1954; C.S.I.R.O. 1954). Even for these minerals, a complication may arise when uraninite and pitchblende samples from any one deposit, and especially from different deposits, are considered, because of the known variation in reactivity of the oxides with their method and conditions of formation (Katz and Rabinowitch 1951).

Various techniques have been employed in some of the investigations reported, viz., contact angle determinations on uraninite and associated minerals (Talbot and Spinks 1953; Korchinski et al. 1954; Van Cleave 1955; Tinker et al. 1956 (2)), and batch flotation of uraninite from synthetic mixtures (Murray 1947), and from natural ores, particularly low-grade pegmatite ores (Talbot and Spinks 1953; Korchinski et al. 1954; Van Cleave 1955; Lord and Light 1956; Tinker et al. 1956 (1), 1956 (2)).

While the effectiveness of both anionic and cationic type collectors has been investigated, the most successful results, from the viewpoint of recovery but not necessarily of ratio of concentration, have been obtained with anionic type reagents. These include fatty acids and their salts, sulphonated crude fatty acid, and octyl orthophosphoric acid. The modifying ions most commonly used are silicates and phosphates. Frequently optimum conditions have been established over a restricted pH range, as for example with a saponified fatty acid and silicate, 7·0–8·0 (Tinker et al. 1956 (1)), and phosphates, 7·6–7·9 (Tinker et al. 1956 (2)); oleic acid and silicate, 6·0–6·9 (Murray 1947); myristate and hexametaphosphate, 8·5–9·5 (Van Cleave 1955; Crawford et al. 1957); oleic acid, 6·4–7·0 (Ginnocchio 1956); fatty acid and petroleum sulphonate, 6·0 (Mariacher 1956); and octyl orthophosphoric acid, 5·8 (Breymann et al. 1953).

In the present studies the action of both an anionic type (diethyldithiocarbamate, $(C_2H_5)_2NCSS^-$), and a cationic type (Armeen CD, largely dodecylamine, $CH_3(CH_2)_{11}NH_3^+$) collector on the flotation of uraninite was investigated.

Experimental methods

The flotation behaviour of small particles of uraninite was investigated by means of the bubble pick-up technique (Cooke 1949; Cooke and Digre 1949). After a specified solution was conditioned for 15 minutes, an air bubble was pressed against the particles for 15 seconds, and the number which adhered to the bubble when lifted was determined. Ten determinations were made for each solution. The temperature of the conditioning solution was maintained at 20° C unless stated otherwise.

The particulate sample was produced from a massive specimen of pitchblende, obtained from White's Mine, Rum Jungle, N.T. Coarse particles (approximately 3 mesh Tyler) were boiled in sodium hydroxide and in water. This material was reduced in size under water with a porcelain pestle and mortar (previously cleaned with chromic-sulphuric acid, sodium hydroxide, and distilled water). The size fraction, —65 +100 mesh Tyler, was separated under water, and was stored under conductivity water.

Amine hydrochloride was prepared by neutralizing

TABLE 1.—EFFECT OF COLLECTOR CONCENTRATION ON FLOATABILITY.

Collector concentration (mg./litre)	Floatability	
	Amine	Dithiocarbamate
0·01	4	—
0·05	10	—
0·1	23	—
0·5	52	—
1	85	—
5	110	3
10	120	n.d.*
25	n.d.	18
100	120	34
300	120	68
500	4	80
1000	—	100

* n.d.—not determined.

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the purified amine, Armeen CD (48 per cent $C_{12}H_{25}^-$, 18 per cent $C_{14}H_{29}^-$) with the appropriate quantity of A.R. hydrochloric acid (Armour and Company 1948). Except where indicated otherwise, sulphuric acid and sodium hydroxide were used for pH adjustments. Conductivity water was used in all the experiments. All glassware used was Pyrex. It was cleaned with chromic-sulphuric acid solution and rinsed with distilled water before use.

Results

The experimental data are presented in terms of the variables studied; all involve the effect of selected variables on the flotability, i.e., the average number of uraninite particles picked up in the ten determinations for each solution after treatment under specified conditions.

Collector concentration

Typical floatability results obtained for amine and dithiocarbamate at various concentrations are given in Table 1. (In both instances the pH of the solutions varied with concentration, but no attempt was made to restrict this variation.)

Temperature

The data obtained on varying the temperature for certain concentrations of both collectors are expressed relative to the observed floatability at 10° C (Table 2).

TABLE 2.—EFFECT OF TEMPERATURE ON FLOATABILITY.

Temperature (°C)	Relative floatability				
	Amine (mg./litre)		Dithiocarbamate (mg./litre)		
	0·1	0·5	15	25	35
10	1·0	1·0	1·0	1·0	1·0
20	2·7	2·4	1·4	1·5	1·3
30	4·5	3·4	1·8	1·9	1·9
40	0·2	1·7	2·2	2·2	2·3

Collector adhesion

The results of experiments to determine the extent to which floatability may be decreased by reducing the concentration of the collector in solution (and thereby possibly allowing absorption to occur) are given in Table 3.

TABLE 3.—EFFECT OF COLLECTOR ADHESION ON FLOATABILITY.

Concentration of conditioning solutions (mg./litre) Initial*	Final*	Total time of conditioning (min.)	Floatability
Amine			
1·0		15	80
	0·01	15	20
		30	6
1·0		15	80
	Distilled water	15	24
		30	6
		105	6
10		15	105
	0·1	15	53
		30	24
Dithiocarbamate			
500		15	75
	15	15	73
		30	75
		45	72
		60	71

* After the floatability was determined the initial conditioning solution was decanted off; the sample was conditioned for various times in the final solution prior to the floatability determinations.

Time

The time taken to reach constant floatability of uraninite was determined for both collectors (Table 4). The observed results are expressed as a fraction of the constant value attained.

TABLE 4.—EFFECT OF TIME ON FLOATABILITY.

Conditioning time (min.)	Floatability (fraction of final constant value).	
	Amine (0·5 mg./litre)	Dithiocarbamate (25 mg./litre)
5	1·0	n.d.
15	1·0	0·33
30	1·0	0·58
60	1·0	0·83
90	1·0	1·00
120	1·0	1·00

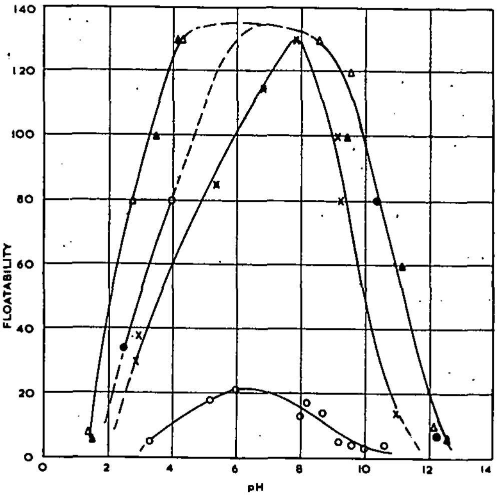

FIGURE 1:—Effect of pH on floatability for amine. Legend: o, 0.1 mg/litre; x, 1.0 mg/litre; ●, 10 mg/litre; △, 100 mg/litre; ▲, 300 mg/litre.

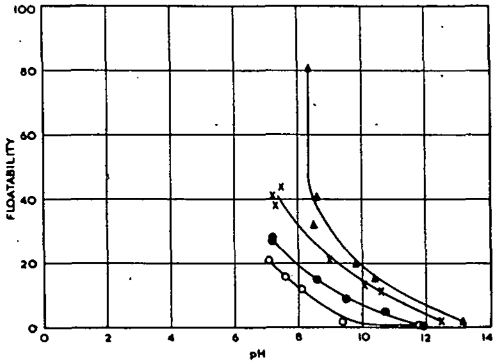

FIGURE 2:—Effect of pH on floatability for dithiocarbamate. Legend: o, 25 mg/litre; ●, 50 mg/litre; x, 200 mg/litre; ▲, 500 mg/litre.

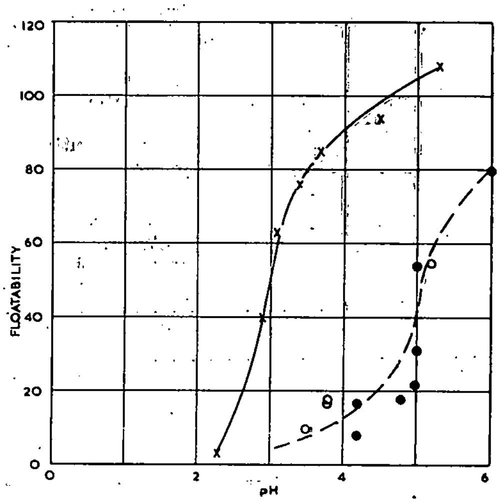

FIGURE 3:—Effect of adjusting pH with different acids on floatability with amine. Legend: o, hydrochloric acid; ● sulphuric acid; x, phosphoric acid.

Agitation

The effect of varying the conditions of agitation from moderate (the standard procedure) to intense (as produced by a mechanical shaker) on the adsorption of dithiocarbamate by uraninite was investigated. For solutions containing 25 mg./litre of collector, the floatability observed after intense agitation was about twice that for moderate conditions.

pH effect

Typical results for amine are shown in Figure 1, and for dithiocarbamate in Figure 2.

In addition, the effect of adjusting the pH using phosphoric, hydrochloric, and sulphuric acids respectively on the floatability with amine (0·5 mg./litre) is shown in Figure 3. Also, it was observed for amine as collector that the floatability in alkaline solutions was not affected by the presence of sulphate ions.

The variation in pH of the collector solutions produces effects which are important in this investigation; one of these is the pronounced decrease in concentration of the amine ion in alkaline solutions, particularly in the pH range above 8·5. This has been studied elsewhere, and the results discussed by Plante (1947).

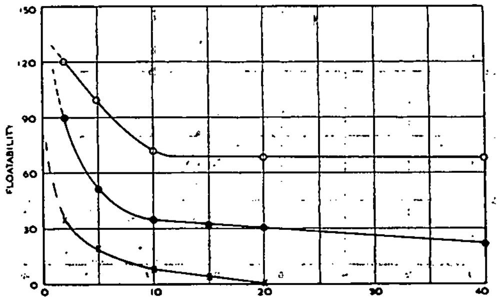

FIGURE 4:—Effect of ageing air bubble on floatability with amine. Legend: o, 300 mg/litre; ●, 400 mg/litre; x, 500 mg/litre.

The other effect is the instability of dithiocarbamate on addition of acid to solutions. This limits the concentration of dithiocarbamate ions in such solutions. The decomposition was investigated by the following procedure. An aliquot of acid was added to the collector solution, it was stirred under standard conditions, the pH was determined, and the residual dithiocarbamate ion concentration estimated by titration with iodine. The floatabilities as determined both for a natural solution containing 50 mg./litre, and for another one of the same concentration, but obtained by adding the appropriate amount of acid to a natural solution containing 500 mg./litre, were identical.

Bubble-armouring

The sudden decrease in floatability observed at the higher amine concentrations (Table 1) was studied in greater detail. The technique adopted involved "ageing" the air bubble for specified times before bringing it in contact with the conditioned particles for 15 seconds. Ageing of the air bubble for as long as 60 seconds had no effect on the floatability for an amine solution containing 100 mg./litre. The results obtained for solutions of higher concentrations are shown in Figure 4.

Discussion

The process occurring at the solid solution interface in the systems investigated is quite complex, because of the possibility of competition between solute species, and between the latter and the solvent for sites in the interface. No attempt has been made, however, to elucidate completely the many aspects of this process as, for example, determination of the amount of collector species absorbed per unit area of interface under various conditions. Instead, an attempt has been made to discover how certain factors affect the flotation behaviour of uraninite particles.

For collectors to function as such, they must form what may be regarded as an interfacial complex (Carr in press) or a localized two-dimensional absorption compound (Biltz, 1904).

Although the adsorption compound so formed is not necessarily identical with any interaction compound formed in solution, chemical interaction between collector species and soluble salts containing the mineral ions may prove a guide to the adsorption of collector at the solid-solution interface.

With respect to the collectors used in this investigation, viz., amine and diethyldithiocarbamate, it is interesting to note that the uranyl ion, $UO_2^{2}+$, which is remarkably stable, has a strong tendency to increase its covalency by complex formation as, for example, either by:

(i) adding amine and phosphate to form uranyl amine compounds (Bailey, 1948; Bearse et al., 1948) — $(CH_3NH_3)UO_2PO_4$, $(C_2H_5NH_3)UO_2PO_4$; or

(ii) forming a complex salt with alkaline salts —$UO_2(SSCN(C_2H_5)_2)_2$, a yellow orange compound existing in the pH range 2.5-7.0, and $UO_2(SSCN(C_2H_5)_2)_2 \cdot NaSSCN(C_2H_5)_2$, a red compound formed at a pH higher than 7.0 (Paley, 1956; Zingaro, 1956).

The identity of the reactive species in the mineral surface is not known. In fact, little is known about the real nature of the surface of the uraninite particles. Although both $U^{4}+$ and $U^{6}+$ are considered to be present in the mineral, it is expected that the uranium present in the surface will be oxidized to $U^{5}+$ or $U^{6}+$ (McConnell and Roberts, 1957). In other words, the surface species may be principally hexavalent, probably hydrated. In this connection, it is interesting to recall that the ion $U^{6}+$ does not exist in aqueous solution, but instead what may be regarded as a hydrolysed state, $UO_2^{2}+$, is formed (Ahrland, 1949).

Also, interaction of $U^{4}+$ with oxygen and water results in the formation of the uranyl ion (Halpern and Smith, 1956). The significance of the collector species—uranyl ion interactions quoted will be apparent in relation to the present investigation.

The conditions under which adsorption compounds are formed with amine and dithiocarbamate respectively, have been investigated by determining the floatability of the uraninite particles. The actual relationship between floatability and the concentration of collector species in the uraninite-solution interface is not known. At this stage the assumption is made that floatabilty varies in some way with the degree of surface coverage by the collector species. Moreover, it has been shown elsewhere (Wark and Cox, 1934; Gaudin and Vincent, 1942), that the contact angle, Θ, changes in a continuous way with collector concentration. The floatability with amine in particular is not solely a function of surface coverage, however, because the interfacial energy of the air-water interface, γ_{aw}, a factor determining the suspending force in pickup tests, also varies with the concentration of collector. From the data given in Table 1, it may be concluded that the amine species is more readily adsorbed at the uraninite-solution interface than is the dithiocarbamate.

The fact that the floatability increased steadily with increasing temperature, 10°-40°C, for dithiocarbamate (Table 2), may be taken to indicate activated adsorption of the collector species. It is suggested that this interpretation receives some support from the fact that a plot of ln (floatability) against 1/T gives a linear relationship ef. Arrhenius plot of ln (velocity constant) against 1/T. For the purposes of this plot, floatability, as determined after a constant conditioning time, is assumed quite arbitrarily to be some measure of the amount of collector species adsorbed in that time. The data for amine is quite reminiscent of that for activated adsorption of a gas in that floatability decreases (i.e., desorption occurs) at 40°C because the distribution coefficient for collector species, $C_{surface}/C_{solution}$ decreases with increasing temperature.

The floatability may be reduced by causing desorption of collector to occur in another way, viz., by reducing the concentration of the collector in solution. While the data in Table 1 indicate this possibility, only for amine is there virtually complete desorption of the "excess" collector after conditioning for 30 min. (Table 3). The adhesion of the adsorbed dithiocarbamate is so strong that it was not possible to reverse the process.

Not only is desorption of dithiocarbamate extremely slow, but also absorption is relatively slow (Table 4). On the other hand, the interaction of amine is fairly rapid. Other factors shown to affect the adsorption of dithiocarbamate are temperature (already discussed), and movement of the collector species through the diffusion layer considered to surround each particle. This is inferred from the observed effect of type of agitation employed during conditioning.

It has already been emphasized that the collector species have to compete with other species, both solute and solvent, for surface sites. From the floatability data given in Figures 1 and 2, it may be inferred that H+ competes with the amine ion, $CH_3(CH_2)_{11}NH_3+$, and OH− with the dithiocarbamate ion, $(C_2H_5)_2NCSS-$. The fact that the hydroxyl ion competes with dithiocarbamate is not surprising in view of the hydrolytic reactions which take place between uranyl and hydroxyl ions with the formation of polymeric complexes (Ahrland, 1949; Sutton, 1949), or the reactions between uranyl ions and sodium hydroxide to form alkali metal polyuranates (Sutton, 1955). A real assessment of the effect of pH on floatability is not possible, however, because whereas the pH of the bulk solution is determined, it is the pH of the interfacial phase which should be known. These are probably not identical.

Variation in pH also affects floatability by altering the concentrations of amine and dithiocarbamate ions in solution. First, the decrease in floatability in alkaline solutions of amine is probably caused by the marked decrease in concentration of the ion. This decrease results from the fact that the ratio free base/ion depends on the pH, according to the relationship $RNH_2 + H+ \rightleftharpoons RNH_3+$, and the free base exhibits restricted solubility in the pH range above 8.5 (Plante, 1947). Second, the decrease in floatability with dithiocarbamate solutions to which acid has been added can be ascribed to the corresponding reduction in the concentration of dithiocarbamate ions in solution. Apparently the products resulting from the decomposition of dithiocarbamate, viz., CS_2 and diethylamine (Lopatecki and Newton, 1952), do not affect the floatability. In general, the floatability (or surface coverage by collector species) is a function of the concentration of the collector ion in solution.

The pronounced effect observed when the pH was adjusted with phosphoric acid is undoubtedly caused by the known strong interaction between uranyl and phosphate groups, as for example with H_3PO_4 to form complexes such as $UO_2H_2PO_4+$, $UO_2H_3PO_4++$ (Baes, 1956), with phosphate and amine to form precipitates of dodecylammonium uranyl phosphate (Bailey, 1948; Bearse et al., 1948), and with insoluble octadecyl phosphoric acid monolayers (Parreira and Pethica, 1957). Uranyl ions likewise form complexes with sulphate and chloride ions, but the interaction is not as strong as for phosphate. Thus it may be deduced that in particular phosphate, and possibly sulphate, and chloride ions act as activators in promoting the formation of adsorption compounds containing amine. From this it follows that phosphate may also act as a depressant by preventing the formation of adsorption compounds containing anionic-type collectors.

Floatability not only decreases in amine solutions when the concentration of the ion is reduced, but also when bubble armouring occurs, i.e., when it is impossible for interpenetration of the amine species adsorbed at the air-water interface and the two-dimensional array immobilized at the solid-solution interface (Leja and Schulman, 1954) to occur. The effect observed when the bubbles were aged (Figure 4) is indicative of the occurrence of so-called armouring, more especially since it has been observed elsewhere (Hoffman et al., 1942) that γ_{aw} for dodecylamine hydrochloride solutions decreases slowly with time.

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Treatment of Radium Hill Concentrates for the Recovery of Uranium

By J. N. Almond*

The paper describes the Port Pirie Treatment Plant for recovery of uranium from the refractory Radium Hill concentrates, using the sulphuric acid-ion exchange processes. The plant is characterised by the counter current decantation washing system and four column ion exchange operation.

Introduction

The recovery of uranium from ores has been the basis of considerable research and industrial effort for more than a decade following recognition of the military importance of uranium and its potentiality as a fuel in nuclear power plants.

Prior to the Second World War, most of the world's uranium was produced as a by-product in the recovery of radium from the rich deposits of Canada and the Belgian Congo. The greatly increased military demand stimulated intensive prospecting, which revealed uranium deposits, mostly of low grades, in many countries of the world. The large-scale treatment of such deposits is now a feature of the uranium industry.

The extraction of uranium follows the general pattern of hydro-metallurgical recovery of other metals, as in the recovery of gold, vanadium, copper, etc. Three stages are involved, the dissolution of the metal from the ore, the separation of the solution containing the metal from the spent solids, and the recovery of the metal from the solution. In many hydro-metallurgical processes the extraction is carried out under alkaline or neutral conditions. Whilst in the large-scale recovery of uranium in plants in South Africa, Canada and the U.S.A., dilute sulphuric acid leaching is employed, requiring acid resisting equipment. In some sections corrosive and highly abrasive conditions are met with.

The early technique of uranium extraction involved mechanical separation of a uranium rich fraction, which was leached with sulphuric acid to take the uranium into solution. After filtering off the spent solids, the uranium was recovered from the liquor by one of four standard procedures:

(i) The addition of lime, magnesia or ammonia to obtain the uranate or $UO_3 2H_2O$, by one or more precipitation stages.

(ii) Reduction of the leach liquor with metallic iron, then the precipitation of uranous phosphate, followed by caustic treatment of the filter cake to remove impurities. Finally, the cake is dissolved in acid and uranium precipitated by addition of ammonia.

(iii) Addition of peroxide under controlled conditions to precipitate the oxide.

(iv) The precipitation of impurities by addition of carbonate, followed by the addition of ammonia to precipitate uranium as uranate.

The small quantities of uranium-rich ores treated were readily soluble in weak acid. In the case of the more refractory uranium minerals, alkali fusion or alkali leaching was used.

All of these processes have certain difficulties, the filtration and washing of the precipitates which are formed, resulting in low recoveries and undesirable contamination of the product.

Research over the last 15 years has been directed towards the development of more efficient methods of extraction as the older processes were unattractive for large-scale exploitation.

Investigations were carried out, mainly at the Division of Industrial Chemistry, C.S.I.R.O., Melbourne, into the recovery of uranium from the ore by the established processes; but all were found to be limited and costly. At the same time an investigation into the metallurgical beneficiation of the ore was conducted and this resulted in a relatively high-grade concentrate being made available for chemical processing.

In 1951 a process was developed in which the concentrates were leached with nitric acid and the uranyl nitrate extracted by solvent extraction with hexone. Water extraction of the hexone recovered the uranyl nitrate in the aqueous layer and addition of ammonia precipitated pure ammonium uranate. This method appeared promising if the major portion of the nitric acid could be regenerated from the nitrate formed during the leaching.

In 1952 a technical mission from the Combined Development Agency put forward the basic principle of the sulphuric acid-ion exchange process, and suggested its application for the extraction of the uranium from the Radium Hill concentrates. After comparing this process with the nitric acid-solvent extraction method, it was apparent that it was more economical and overcame the raw materials supply problem. The Division of Industrial Chemistry, C.S.I.R.O., investigated this new process until laboratories and a pilot plant were established in South Australia late in 1952 to continue the work and to finalise details of the method of treatment of the concentrate.

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*Uranium treatment plant, Port Pirie, S.A. Manuscript received March 17, 1958.

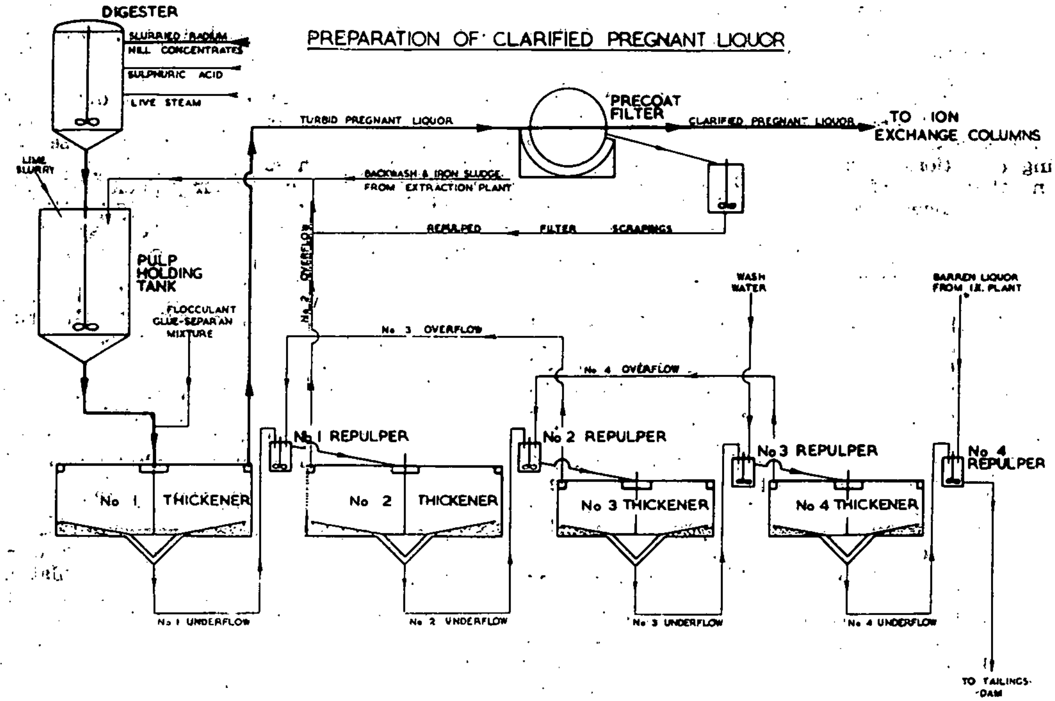

FIGURE 1:—System for leaching uranium. Separation of dissolved uranium from spent solids.

Uranium Production at Port Pirie

The feed to the treatment plant is railed 230 miles from Radium Hill as a black powder of minus 200 mesh and containing 10-15% moisture. The uranium is carried in the mineral davidite, an ilmenite-haematite complex. The mineral composition of the concentrate is as follows:—

Component	Percentage by Weight Average	Range
Rutile	26	19-37
Haematite	22	14-28
Biotite and Chlorite . .	13	6-20
Ilmenite	12	8-16
Davidite	10.5	7-14
Quartz and Felspar . . .	6	2-15
Magnetite	5.5	2-8
Other constituents. . .	5%	—

The main raw materials are:

(a) Sulphuric Acid—98%.
(b) Precipitated calcium carbonate containing a small amount of caustic soda.
(c) Sodium Chloride—commercial grade.
(d) Animal bone glue.
(e) Diatomaceous earth.
(f) Magnesia.

(i) Dissolution of uranium

Figures 1, 2 and 5 outline the flowsheet of the process. The concentrates are discharged from rail trucks on to the stockpile by a transporter crane. This crane also loads concentrates from the stockpile into a feed hopper. The concentrate is discharged from the base of the hopper, and fed by a 12-inch belt conveyor over a Blake-Denison weightometer to record the plant input. It then discharges over the end via a sample cutter, into one of two mild steel concentrate slurry tanks, 8 ft. high x 8 ft. diameter, each of which is fitted with a turbine type agitator driven by a 20 h.p. motor. These tanks, like all other agitator tanks in the plant, are fitted with 4 baffle plates to ensure efficient agitation of the charge. Each tank holds ten tons of concentrate as a 60-62% w/w slurry in water, representing one charge for a digester.

The slurry is pumped into one of the digesters on the top floor of the leaching plant building. This building is an asbestos covered, steel-framed structure with two working levels. On the top level, digesters are set in the framework of the building. The mild steel digester shell, 11 ft. 6 in. high and approximately 8 ft. diameter, having a conical base, is lined internally with lead which, in turn, is covered by a double layer of acid-resisting ceramic to a total thickness of 4½ inches. Internal baffles made up from special sections of the ceramic are built integrally into the lining. The dished lid, which is bolted to the digester body, has a manhole, a sight glass and concentrate slurry, acid, and steam inlet points, and also a vent connection.

The drive for the 5-blade turbine type stirrer, fitted with a stabilising ring, is mounted centrally on the lid. The 3-inch diameter, solid stainless steel shaft entering the digester via a water-sealed stuffing box is approximately 13 ft. long, rotates at 56 r.p.m., is driven by a 20 h.p. motor and has no bearing inside the digester.

The lids are lead lined internally, but more recently some have been coated with an Epoxy resin preparation. Lead lining has not been completely satisfactory due to creep and corrosion. As an alternative, an Epoxy resin filled with solid leach residues applied to the mild steel surface has shown promise under operating conditions. This resin has also been used in other parts of the plant where severe abrasion is encountered, combined with corrosive conditions.

The slurry is agitated continuously and concentrated sulphuric acid (98%) is added from an acid charge-head tank, supplied from one of two 350-ton storage tanks.

In leaching the concentrate, 750 pounds of sulphuric acid (100%) is used per short ton of concentrates and is added over a period of one hour. The charge is brought to the boil by the heat generated by the dilution of the acid and the heat of the reaction, and this boiling is maintained for 10 hours by the admission of live steam. 92-94% of the uranium is dissolved from the concentrate by this treatment and the acid strength reduced from approximately 37.5% to 2-5% free sulphuric acid. These are the most severe conditions of corrosion and abrasion found in the plant.

Stainless steel, type 316, has been found to be the most suitable for the agitator and its shaft, but it is only satisfactory if the chloride content of the liquors remains at a low level.

The mains water supply at Port Pirie is from the Morgan-Whyalla pipe-line and two reservoirs in the nearby Flinders Ranges. This water contains 300 parts per million chloride and 1,000 p.p.m. TDS. In addition, the concentrates contain up to 0.06% chloride. These two sources raise the chlorida concentration in the leach liquor to a level which would cause excessive corrosion of the stainless steel.

To overcome this, a small ion exchange plant has been installed to produce sufficient low chloride water for use in the concentrate slurry made-up. The ion exchange unit has 4 columns, each 10 ft. high and 12 in. diameter, charged with Deacidite FF resin. The columns operate in parallel and the resin selectively removes chloride ions and, when saturated, is regenerated by eluting with a 2N. sulphuric acid solution. Provided the soluble chloride in concentrates is less than 0.06% and the chloride level in the water used in making the concentrate slurry is less than five p.p.m., the corrosion of the stainless steel is checked.

The hot leach pulp is discharged through the bottom outlet of the digester into a pulp-holding tank located below the digester. These tanks, 13 ft. diameter x 19 ft. 6in. high, with a conical base, are fabricated from milk steel and are lead lined. The turbine type agitator fitted to each tank is fabricated from type 316 stainless steel and driven at 60 r.p.m. through a reduction gearbox by a 60 h.p. motor. The quill shaft construction, heavy duty gearbox supports the 4 inch diameter by 16 ft. long agitator shaft and dispenses with a bearing in the pulp.

Prior to receiving the leach pulp, the pulp-holding tank is filled with approximately 10,000 gallons of thickener overflow from No. 2 thickener in the C.C.D. section. This liquor cools the charge, reduces the pulp density to 12-14% solids, decreases the sulphate content of the leach liquor and tends to hydrolyse any titanium present in solution. Whilst agitating the charge, a 50% "lime" slurry is slowly added to reduce the acidity of the pulp to pH 1.6, to produce a liquor with suitable characteristics for subsequent ion exchange operation.

A typical liquor analysis is:

Total Iron	20-30 gm/litre
Ferris Iron	4-6 gm/litre
TiO_2	0.05-0.25 gm/litre
Sulphate	65-70 gm/litre
Chloride	0.5-0.7 gm/litre
pH	1.6

(ii) Separation of uranium solution from pulp

Pilot plant experimental work revealed that the poor filtration and washing characteristics of leach pulp led to high soluble losses, even when the pulp is treated with flocculating agents. Bench tests indicated that a four stage counter current decantation (C.C.D.) washing system provided the best means of separating the pregnant liquor from the solids and reduced this loss to less than 1%.

The C.C.D. Section consists of four thickeners in series. the first two measuring 100 ft. diameter by 15 ft. deep, and the second two 80 ft. diameter by 14 ft. deep. The steel tanks and the internal steelwork, including mechanisms, are rubber covered, as are the diaphragm pumps, pipes, etc., in the section. Where rubber cannot be used, type 316 stainless steel or rigid P.V.C. plastic is used to withstand the corrosive conditions. The underflow lines, slurry pumps and other auxiliary equipment have been duplicated to provide against breakdown or blockages of pipe lines.

The centrally located control room has a comprehensive control board fitted with flowmeters, level controllers and ammeters indicating the current drawn by all electric motors operating in the section. Failure of any of the equipment, including the thickener rake arms, causes an alarm to sound and warning lights flash on the panel to indicate the location of the breakdown. This protection, together with a continuous routine inspection of the plant, sampling of the pulp, and pulp density checks every 30 minutes, has ensured the continuous operation of the section.

The pulp in the pulp-holding tanks is pumped to a launder and gravitates to No. 1 thickener (see Figure 1). A glue-"Separan" solution is added to the pulp in the launder at the rate of 2 lbs of glue and 0.02 lbs. of "Separan" per ton of solids. to flocculate the solids and induce better settling conditions.

At the other end of the C.C.D. system, water is fed into a repulper on No. 4 thickener, mixed with the underflow pulp from No. 3 thickener and fed into No. 4 thickener. This water

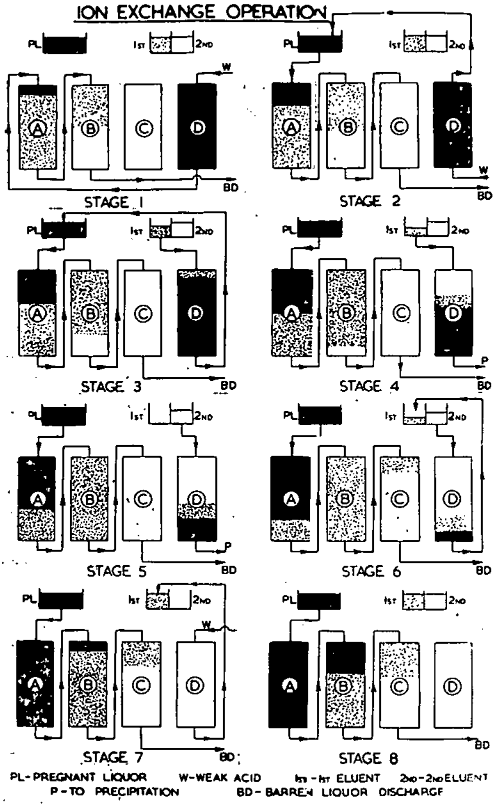

FIGURE 2:—Uranium recovery by ion exchange.

passes progressively through thickeners Nos. 4, 3 and 2 and overflows from No. 2 thickener into a storage tank and is pumped, as required, to the pulp-holding tanks in the leaching plant to be used as diluting liquor.

The solids, fed as a 12-14% pulp, settle and are discharged from the thickener cone as a pulp containing 50% solids at an average sp. gr. of 1.60. This pulp is transferred by diaphragm pump to a small repulper tank where it is mixed with the overflow from No. 3 thickener to make a 12-14% slurry which feeds to a launder and gravitates into No. 2 thickener. This sequence of operations continues from Nos. 3 and 4 thickeners where the solids, washed free of entrained pregnant liquor, are discharged as a 50% pulp into a pump box and mixed with other waste liquors before pumping to the tailings dam.

Over the period of its operation this section has given little trouble and has borne out in practice the calculated washing efficiency of a four-stage C.C.D. system where a fine, slow settling solid with poor filtration characteristics had to be separated from the liquid fraction.

The following table shows the average uranium oxide content of the feed pulp liquor and in the overflows from each thickener:

	U_3O_8 gm/litre
Feed	1.50
No. 1 Thickener	1.50
No. 2 Thickener	0.20
No. 3 Thickener	0.05
No. 4 Thickener	0.012

Recovery of uranium from solution

The following two main operations are necessary to recover the uranium as a relatively pure concentrate from the pregnant liquor:—

(a) The separation of the uranium from the major impurities and its concentration in solution.

(b) The precipitation from this solution of relatively pure yellow uranium oxide.

(a) URANIUM SEPARATION FROM IMPURITIES

The turbid overflow from No. 1 thickener is a relatively weak, cloudy, impure solution of uranium of the same composition as the liquor in the feed pulp. Uranium with some iron, in addition to other minor impurities, are present as complex basic sulphate ions. The uranium complexes include the following ions:—

$UO_2(SO_4)_3\ {----}\qquad UO_2(SO_4)_2\ {--}$

These complex ions are absorbed on certain strongly basic resins and some of these resins exhibit a preference for these complexes over other anions. If the pregnant solution is passed through the resin, the solution yields up its uranium to the resin in what can be considered a double decomposition reaction, written as follows:

$UO_2(SO_4)_3\ {----} + 4RCl \qquad R_4UO_2(SO_4)_3$

With the exception of some iron, the impurities remain in the solution and pass out to waste in the barren liquor.

To recover the uranium from the resin, an acid chloride solution is passed over the resin, stripping the uranium from the resin into solution and at the same time regenerating the resin to its original chloride form. A 1N solution of sodium chloride in O.1N sulphuric acid is used in practice.

The ferric sulphate anions behave similarly to the uranium ion complexes. To ensure optimum conditions for unanium adsorption, leaching conditions are controlled so that the ferric iron-uranium ratio in the leach liquor is less than 5 and preferably less than 3. The acidity of the leach liquor is adjusted to pH 1.6-1.7, as low acidity favours iron adsorption. Adsorbed iron occupies resin sites which could be taken up by the uranium complex ions, so reducing the effective uranium loading of the resin. Eluent is also wasted in stripping the iron impurities, so increasing the overall costs of eluting the uranium.

The turbid pregnant liquor is clarified by filtering through a precoat vacuum drum filter, using diatomaceous earth as the precoat medium and unbleached calico sheeting as the filter cloth. The filter is 6 ft. diameter and 10 ft. face, fabricated from type 316 stainless

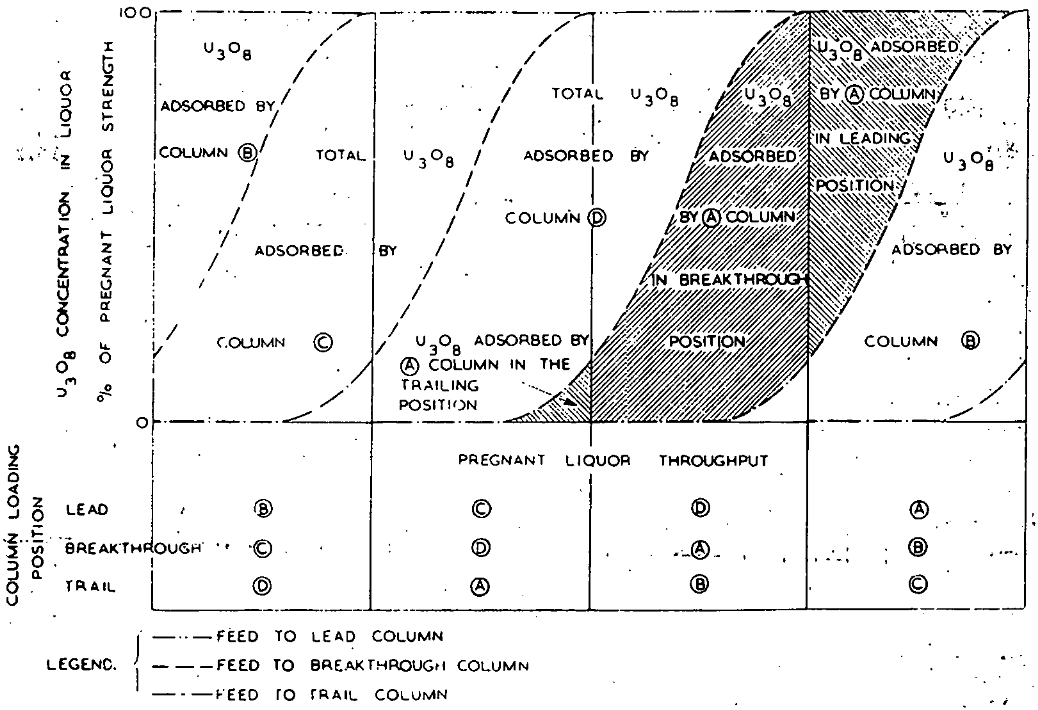

FIGURE 3.

steel and rubber lined mild steel. It is considered that the most suitable liquors for ion exchange treatment should contain less than 1 p.p.m. suspended solids. The clarified pregnant liquor is stored pending pumping to the ion exchange plant.

Two ion exchange sets, each with four columns, have been installed, each set operating independently either automatically or manually controlled. A set consists of four columns or drums, each 7 ft. diameter by 12 ft. high, made of rubber lined mild steel. The operating mechanism is so arranged that any two columns or any three columns can be used in series at one time for adsorption, whilst one or two columns in series can be eluted.

In the pilot plant experiments, the leach liquor characteristics were different from those now generally obtained and were less suitable for ion exchange treatment, due to the high ferric iron content. These early liquors had the following composition:

U_3O_8	1.8—2.2 gm/litre.
Total Iron	28 gm/litre.
Ferric Iron	10-12 gm/litre.
TiO_2	2-8 gm/litre.

Titanium has an adverse effect on the resin by either hydrolysing on or around the resin beads, resulting in the "poisoning" of the resin and, over a relatively few cycles of operation, reducing the resin loading capacity from 55-60 gms. uranium per litre of resin to 20-25 gms. per litre. The high ferric iron-uranium ratio adversely affected uranium adsorption on the resin, reducing the uranium resin loading and producing excessive iron adsorption on the resin and in the eluate.

It is possible, however, to reduce the amount of iron adsorbed by continuing the adsorption cycle after the resin is saturated, when the uranium will slowly replace the iron complex on the resin. Pilot plant ion exchange operation showed that with the pregnant liquors used, the standard two column loading cycle resulted in breakthrough of uranium from the second column before saturation of the resin by uranium had occurred on the first or lead column. As well as inefficient use of resin, an excessive amount of iron complex was adsorbed on to the resin. Thus, three loading columns in series adopted to ensure uranium saturation of the resin in the lead column, whilst the trailing column acts as a scavenger, reducing uranium losses in the out-going barren liquor to a minimum.

With the lower ferric iron content pregnant liquor now encountered at Port Pirie, the three column adsorption is still used, as it protects against uranium losses in the barren liquor. Uranium breakthrough is determined on liquor from the outlet of the second or breakthrough column on the adsorption cycle. When uranium breakthrough is indicated, the loading cycle is continued for a further 5 hours to ensure saturation of the leading column and replacement of some of the adsorbed iron by uranium.

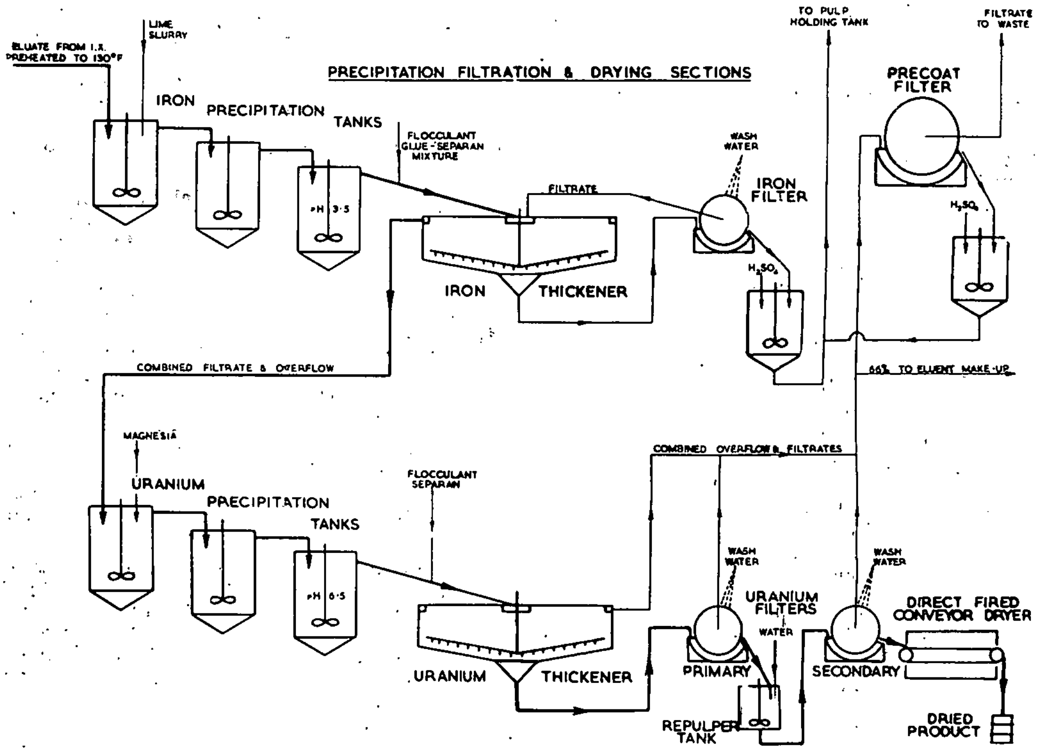

FIGURE 4:—Uranium recovery from ion exchange eluate.

One complete cycle for loading and eluting the resin involves eight operating stages and these are shown in Figures 2 and 3. Column D has just completed loading and is to be eluted, whilst columns A, B and C are commencing the adsorption cycle with column C being brought on stream as the trailing column, as follows:

STAGE 1—Column D has just been completely loaded and taken off the loading cycle as the first or leading column when columns D, A and B were on adsorption. Column D is given a downwards weak acid wash to displace the barren liquor via columns A and B to waste. Any soluble uranium from the displacement wash is adsorbed on columns A and B.

STAGE 2—Columns A, B and C are on adsorption.

Column D is given a backwash with dilute sulphuric acid to remove any solids which have settled out on the top of the resin bed, i.e. this is a cleaning stage for the resin, the backwash liquor finding its way eventually via the leaching plant to the pregnant liquor storage.

STAGE 3—Columns A, B and C on adsorption.

Column D. The weak acid remaining in the column is removed by a downwards displacement wash of first eluent. The displaced liquor is returned to pregnant liquor storage via the Leaching Plant.

STAGE 4—Columns A, B and C on adsorption.

Column D. First eluent continues to pass through Column D and the outgoing liquor or eluate pumped to the eluate tank for subsequent treatment for uranium recovery. At this stage a considerable amount of the uranium on the resin is being stripped and the eluate is quite concentrated with respect to uranium. Figure 4 indicates how the uranium is eluted, the graph indicating that the major portion is stripped during this stage.

STAGE 5—Columns A, B and C on adsorption. Breakthrough of uranium from Column B occurs.

Column D: All the first eluent has been used and second eluent is now introduced. One bed volume is passed through Column D, displacing first eluent and the outgoing liquor pumped to the eluate tank.

STAGE 6—Columns A, B and C on adsorption. The leading column A is nearing saturation. Some uranium is now being adsorbed on column C.

Column D: Second eluent continues to pass through column D and is returned to the first eluent tank as it contains only a small amount of uranium. This liquor becomes first eluent to be used in stages 3 and 4 as first eluent for stripping the next loaded column, i.e. column A.

STAGE 7—Columns A, B and C on adsorption.

Column D: All the second eluent has passed through column D and the uranium stripped from the resin. A weak acid wash of three displacement volumes is passed through column D to displace the uranium liquor into the first eluent tank.

STAGE 8—Columns A, B and C on adsorption.

Column A is now fully loaded and is about to be taken off the loading cycle.

Column D is at standby, the resin having been eluted and regenerated to the chloride form. It is ready to return to the loading cycle as the trailing column.

The eluate from Stages 4-5 contains uranium with some impurities, chiefly iron.

Figure 3 shows how uranium is adsorbed by each column on the loading cycle from the feed to each column. It will be seen that uranium is adsorbed by the resin on each column when they are in the trailing, breakthrough (middle) or leading positions.

(b) PRECIPITATION OF URANIUM

Uranium is precipitated as a yellow oxide from the eluate by a continuous precipitation process, using magnesia, after the iron has been removed by precipitation as a hydroxide by the addition of "lime." Figure 5 shows the general arrangement in this section.

The eluate received from the ion exchange plant (stages 4 and 5 of operation) is stored in a rubber-lined, 10,000 gallons storage tank, 13ft. 6in. deep by 12ft. 6in. diameter. It is mixed and heated to 130°F to provide optimum conditions for the subsequent precipitation stages. The eluate is pumped to the first tank of the continuous iron precipitation unit, consisting of three conical base tanks in series, each 5ft. 6in. high by 4ft. 6in. diameter, fitted with an agitator and baffles. A 5% "lime" slurry is fed at the same time to the first tank, the amount being determined by an automatic pH controller, so set as to ensure sufficient "lime" addition to raise the pH of the liquor in the slurry discharging from the third precipitation tank to pH 3.5.

Ferric hydroxide and calcium sulphate are precipitated and the pulp, after dosing at the discharge point with glue, to flocculate the solids, gravitates to a 16ft. diameter by 7ft. deep thickener. The ferric hydroxide and calcium sulphate underflow is pumped to a 3ft. diameter and 1ft. face vacuum drum filter, the resulting filter cake, after washing, is slurried with weak sulphuric acid and returned to the leaching plant for recovery of the entrained uranium.

The filtrate and washings are combined with the thickener overflow and pumped to the first tank of the continuous uranium precipitation unit. This unit is identical with the continuous iron precipitation arrangement.

An automatic pH controller determines the amount of a 3% magnesia slurry run into the first tank to raise the pH of the liquor to 6.5 at the point of discharge from the third precipitation tank. A bright yellow mixture of uranium hydroxide and magnesium uranate forms in the three tanks, and at pH 6.5 uranium precipitation is complete. The slurry gravitates to a rubber covered thickener, 16ft. diameter by 7ft. deep, after dosing with flocculent. The underflow is pumped by diaphragm pump to the primary vacuum drum filter, 3ft. diameter and 1ft. face. The yellow filter cake is washed with water, and drops from the filter into a small repulper tank and is slurried with water.

The yellow cake slurry is pumped to the secondary filter, which is a rotary vacuum drum type, 2ft. diameter by 4ft. face. The slurry is

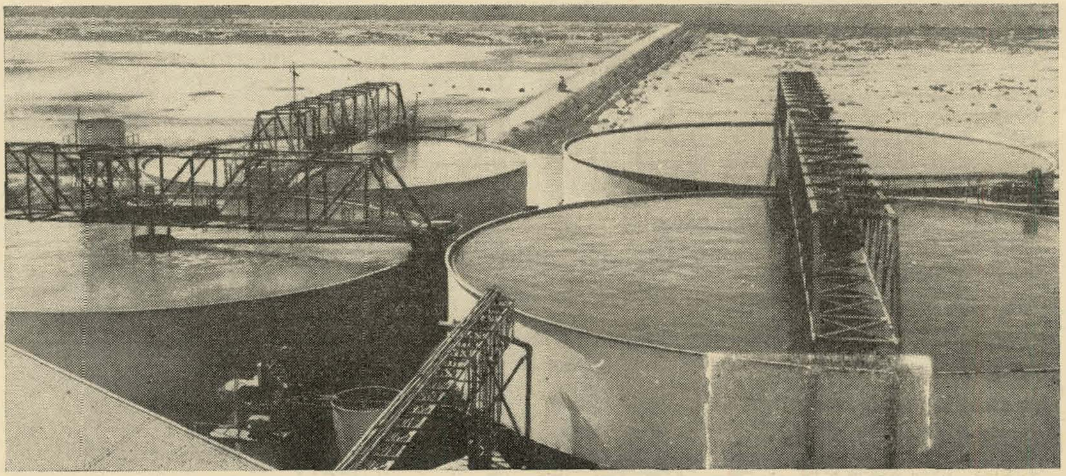

FIGURE 5:—General view of counter current decantation washing section. No. 1 thickener overflowing pregnant liquor at rear right. Tailings dam for washed spent solids shown in upper left hand background.

filtered, washed with water, discharged on to a conveyor belt, and transported to the dryer. The filtrate and washings from the primary and secondary uranium filters are combined with the thickener overflow and pumped to a rubber-lined storage tank of 10,000 gallons capacity. Approximately one-third of this eluate is pumped to a precoat vacuum drum filter for recovery of the finely suspended uranium and the filtrate rejected to waste. The remaining two-thirds is pumped to a rubber-lined make-up tank of 10,000 gallons capacity, where brine and sulphuric acid are added to form the second eluent used in stages 5 and 6 in the ion exchange plant.

The wet yellow cake falls from the conveyor belt into a hopper of the pelleter, which consists of a stainless steel plate rolled into an arc and drilled with ¼ inch diameter holes placed at 2 inch centres.

Two rubber covered rollers reciprocate over the length of the plate, extruding the yellow cake on to the stainless steel belt of the dryer. The drying belt is operated through a variable speed drive reduction unit, at an average speed of 30ft. per hour. Drying is accomplished by combustion gases from diesel fuel oil circulating through the dryer and passing over the wet pellets. The average temperature in the dryer near the belt is approximately 600°F. The dryer is kept under slight negative pressure to minimise the escape of uranium dust. A portion of the moisture saturated vapours is bled off and passed through an electrostatic precipitator to trap any dust prior to discharging to the atmosphere.

The dried product drops into a hopper and discharges into open headed 44 gallon capacity steel drums. After sampling, the drums are sealed, weighed, suitably marked and are then ready for despatch.

The Occurrence of Uranium in Queensland

By J. H. Brooks*

All deposits of uranium of possible economic significance in Queensland are confined to rocks of Proterozoic age. With the exception of the Westmoreland prospect, they are all located in the Cloncurry-Mount Isa area. There is no close association between uranium and other metallic mineral deposits in that area. The Mary Kathleen is the only major economic deposit proved to date. The Counter and Skal could provide moderate tonnages of ore, and thirteen other occurrences may have a small quantity of economic grade ore.

The deposits are considered to be of epigenetic origin and may be grouped into four main types. The Mary Kathleen type occurs in highly altered, calcareous and arenaceous sediments, and is characterised by the presence of zones of garnetisation and the occurrence of fine-grained uraninite as inclusions in allanite. The Counter type of deposit occurs in a succession of interbedded, metamorphosed, arenaceous sediments and basic volcanics. The uranium occurs as disseminated uraninite or brannerite. In the torbernite-ironstone type of deposit low grade, disseminated uranium mineralisation occurs in massive ironstone lodes and small concentrations of torbernite occur in the zone of oxidation. Davidite-type minerals have been discovered, practically all of them in a sequence of dominantly calcareous metasediments, mostly associated with pegmatite or granite. Extensive secondary uranium mineralisation occurs in beds of sandstone in the Westmoreland Conglomerate. There is a small torbernite occurrence at Percyville, North Queensland, and several other occurrences of mineralogical interest in the Herberton, Charters Towers and Mount Perry districts.

Introduction

The first discovery of significant deposits of uranium in Queensland was in March, 1954.

Mary Kathleen is the only deposit to which a close field and laboratory study has been directed. Few of the other occurrences have been subjected to anything more than reconnaissance geological mapping, and knowledge of their behaviour in depth is limited because of the small amount of underground exploration which has been carried out to date. Much valuable mineragraphic work has been undertaken, but such is the complexity of the mineralogy that a great deal remains to be done. In this paper an attempt has been made to bring together in summary form the available data.

Distribution

Practically all the deposits of economic significance discovered to date are confined to the Lower Proterozoic rocks of the Cloncurry-Mt. Isa mineral fields. The only deposit of possible economic significance elsewhere is in the Upper Proterozoic rocks of the Westmoreland area in the far north-western corner of the State. Other small, isolated occurrences have been found at Percyville, in the Etheridge Mineral Field, North Queensland, and at Targinnie, near Gladstone, in Central Queensland. The Percyville occurrence is in Proterozoic rocks, but that at Targinnie is in granite, which is intrusive into late Palaeozoic sediments. It is the only case where appreciable uranium mineralisation is found in rocks younger than Proterozoic.

Several occurrences of mineralogical interest have been located in the Palaeozoic rocks of the Herberton, Charters Towers and Mt. Perry districts. The presence of uranium has also been confirmed in a ferruginous sandstone of Mesozoic age west of Rockhampton. Figure 1 shows the distribution of uranium occurrences in Queensland.

Cloncurry-Mount Isa area

Some hundreds of claims for discoveries of uranium have been made over an area of about 10,000 square miles. They are distributed in a belt 160 miles long and 60 miles wide, trending north-north-west. The geosynclinal sediments and volcanics within this area are highly folded and faulted, moderately metamorphosed, and intruded by numerous major bodies of granite and minor basic dykes. With two exceptions, uranium has not been found in the less deformed and relatively unmetamorphosed epicontinental Precambrian sediments which occur principally to the north-west of the Cloncurry-Mt. Isa area, the main exception being the Westmoreland deposit. The other is an occurrence of weak uranium mineralisation in a ferruginous caprock over flatly dipping Upper Proterozoic or Lower Cambrian sandstones and shales in the Mt. Birnie area, 68 miles south-south-east of Mt. Isa.

Occurrences have been found in most Lower Proterozoic rock formations deposited prior to the major Proterozoic orogeny and the accompanying granitic intrusions. They are most abundant in two formations—The Eastern Creek Formation† of interbedded, metamorphosed basic volcanics and dominantly arenaceous sediments, and the Corella Formation† of metamorphosed, impure limestone and dolomite, with subordinate arenaceous sediments and basic volcanics. The granites of the area which have been tested are slightly radioactive, exhibiting two to five times background radioactivity.

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*Geological Survey of Queensland, Department of Mines, Brisbane. Manuscript received March 10, 1958.

† Formal names proposed by the Bureau of Mineral Resources for adoption by the Stratigraphic Nomenclature Committee of the Geological Society of Australia.

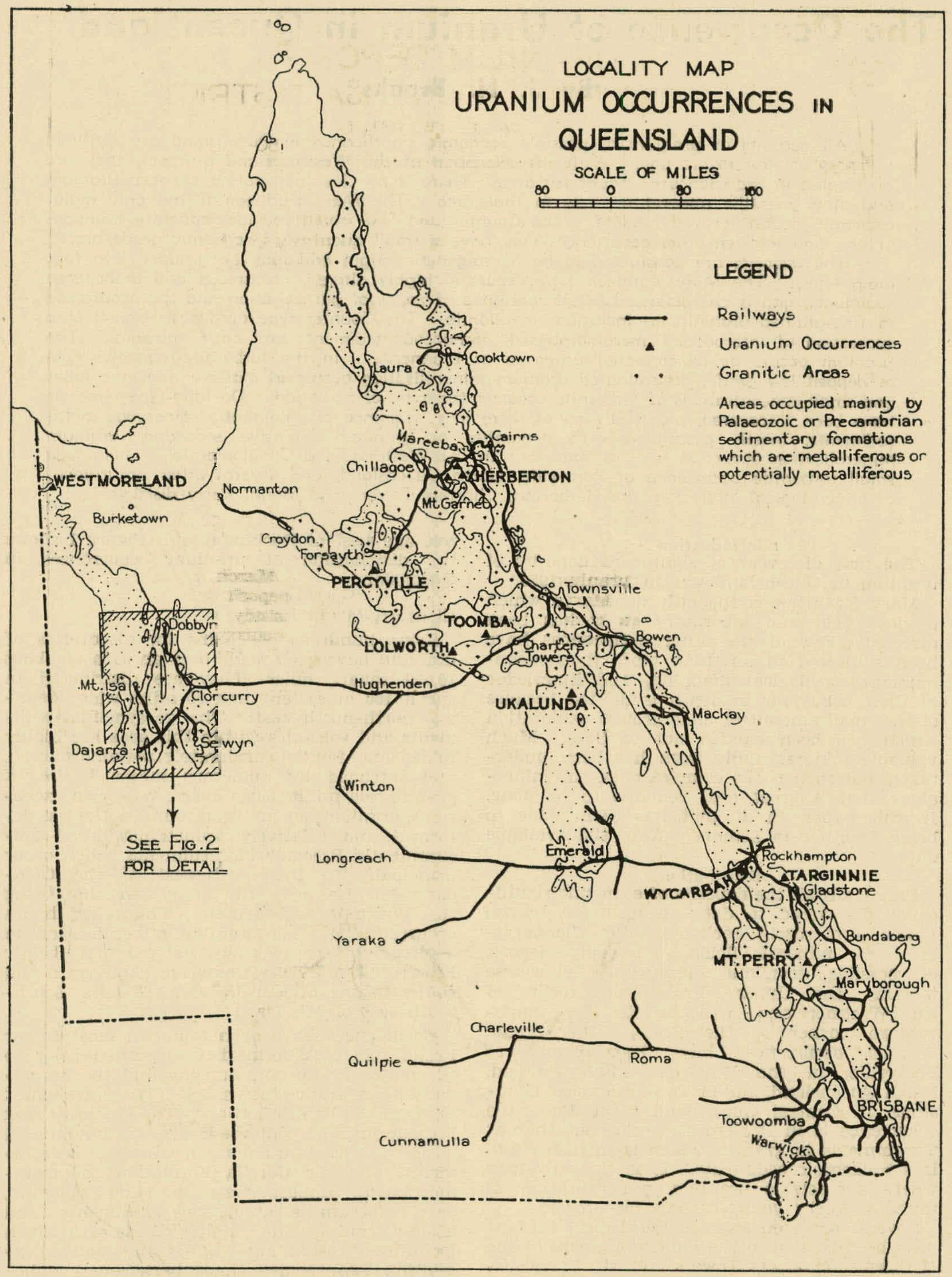
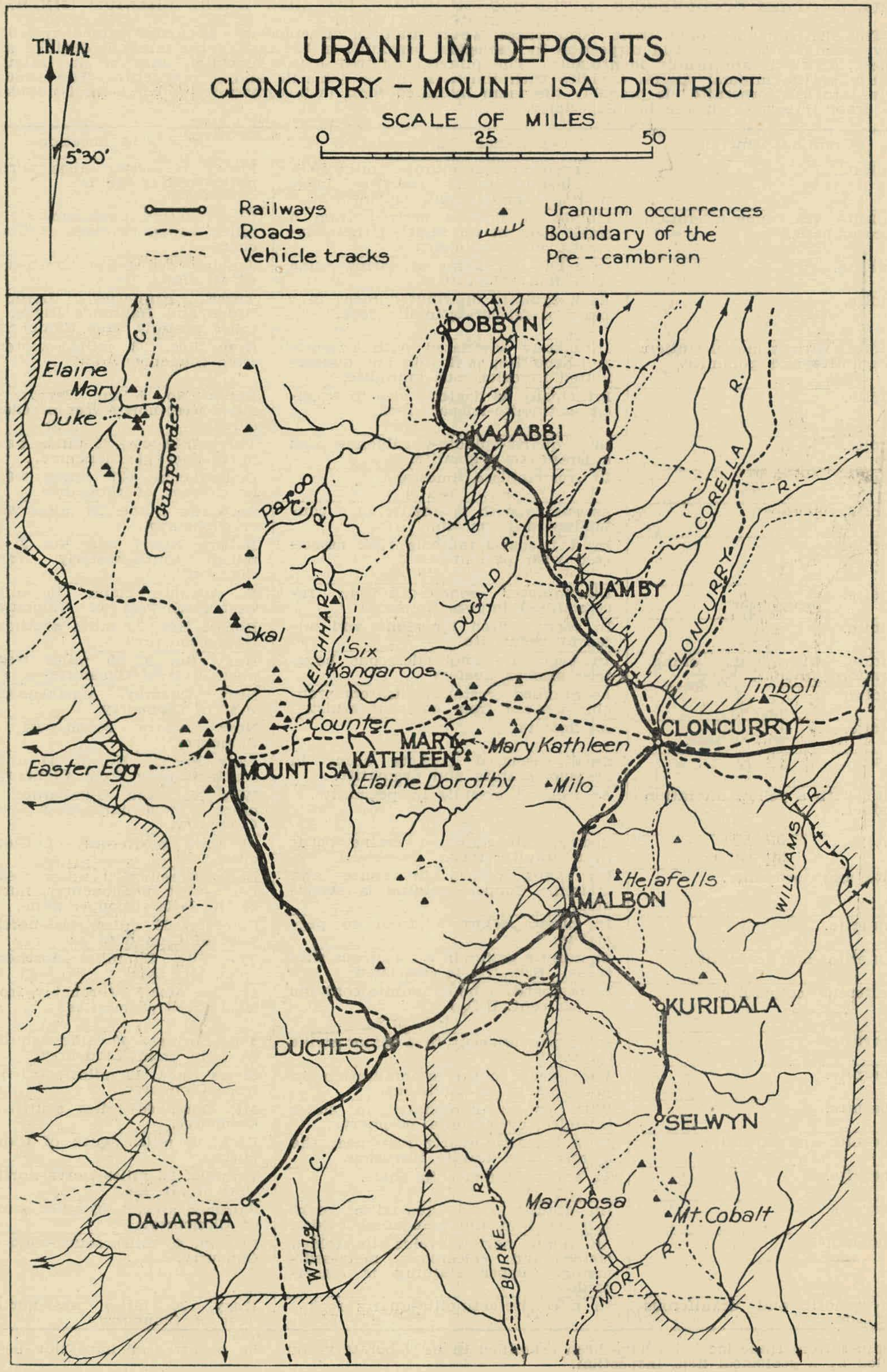


FIGURE 1:—Locality Plan; uranium occurrences in Queensland.



URANIUM DEPOSITS
CLONCURRY – MOUNT ISA DISTRICT

TABLE 1.—OTHER OCCURRENCES IN THE CLONCURRY-MT. ISA AREA WHERE URANIUM MINERALS HAVE BEEN IDENTIFIED

This list includes only those occurrences where uranium minerals have been determined by laboratory examination, or by field inspection by geological personnel in the case of the minerals davidite and torbernite, which are readily identifiable. The mineral referred to as "davidite" may be an intimate mixture of the mineral species davidite with ilmenite and/or hematite and/or magnetite. The mineral referred to as "torbernite" is probably the more hydrated form meta-torbernite, which is indistinguishable from torbernite in the hand-specimen.

Uranium Minerals	Occurrence and Association	Lease and Location
Uraninite.	As aggregates of minute grains in a hornblende schist carrying magnetite, hematite and sphene.	Flat Tyre, seven miles north-north-west of Mt. Isa.
Uraninite with some alteration to uranophane.	Mainly as floaters derived from a small lode in an albite-hornblende-calcite meta-sediment.	Clear Waters, approximately 30 miles west-north-west of Cloncurry.
Uraninite.	In a lode consisting of narrow veins in a fold structure.	Janelle, five miles south-west of Mt. Isa.
Davidite.	As irregularly distributed blebs in a band of felspar-actinolite rock.	Three Brunettes, Leydon's Ridge and Hannan's Ridge, 24 miles westerly from Cloncurry.
Davidite, with some alteration to carnotite and gummite.	Intimately associated with ilmenite and hematite; as floaters and disseminated in calcareous granulite.	Rosy Bee, 38 miles westerly from Cloncurry.
Davidite.	Principally as floaters; also in a calcite-actinolite-felspar rock.	Hobby No. 2 and Verpat, 26 miles west-north-west of Cloncurry.
Davidite.	In a quartz-epidote-hornblende rock adjacent to pegmatite.	Three Inkspots, 25 miles west-north-west of Cloncurry.
Davidite.	In recrystallised limestone.	Dodge-Loane, 21 miles west-north-west of Cloncurry.
Davidite.	Associated with magnetite in a contaminated pegmatite.	Amy and Diane, 28 miles west of Cloncurry.
Davidite.	In a felspar-amphibole rock associated with pegmatite.	Ballara No. 1 and No. 2, 39 miles south-westerly from Cloncurry.
Davidite.	Disseminated sporadically in recrystallised impure limestone.	P. Aittola's claim, 15 miles west-north-west of Cloncurry.
Davidite.	In recrystallised limestone and adjacent pegmatite.	Robert Heg, 35 miles southerly from Cloncurry.
Davidite, with some orange alteration product.	In pegmatite and calcareous hornfels; also as floaters.	Huxy No. 3, 30 miles west-north-west of Cloncurry.
Davidite.	As rounded crystalline floaters probably derived from adjacent schists.	Near Quamby, north-north-west of Cloncurry.
Davidite.	As floaters.	Near Dajarra, south-west of Cloncurry.
Absite.	Small, isolated pockets occur in graphic, granite and pegmatite.	Lady Rachel, No. 6, 44 miles south-westerly from Cloncurry.
Absite, with a yellow alteration product.	As isolated crystals in pegmatite.	Why Not and Gumbardo, 24 miles west-north-west of Cloncurry.
Absite.	Sparsely disseminated grains occur in a granitic rock.	28 miles north-west of Cloncurry.*
Absite and torbernite.	On joint planes in aplite and greisen. Copper staining is associated.	Doris Norelle, 22 miles west-north-west of Cloncurry, north of the Federal copper mine.
Brannerite.	In biotite schist adjacent to pegmatite.	Comfort, 28 miles west-north-west of Cloncurry
Brannerite and carnotite.	As minute grains in a calcereous siltstone. Hematite is associated.	The Pile, 22 miles northerly from Mt. Isa.
Uraniferous columbite.	In pegmatite (the columbite contains 0.7 per cent. U_3O_8.)	Mt. Isa area.* (Probably from Mica Creek pegmatites.)
Torbernite.	In jasper adjacent to a major strike fault.	The Springs, 40 miles south-westerly from Cloncurry.
Torbernite.	With weak copper mineralisation on joint planes in sandstone.	Sierra Rada, 19 miles southerly from Cloncurry.
Torbernite.	With copper carbonates in slates intruded by rhyolite porphyry.	Mt. Dore, 66 miles south of Cloncurry.
Torbernite and autunite.	In a hematite-quartz rock near the margin of a granite intrusion.	Utah, 63 miles south of Cloncurry.
Torbernite.	As scattered flakes in shale.	Valhalla, 26 miles north-north-west of Mt. Isa.
Torbernite.	In granitic and quartzitic rocks carrying iron and manganese oxides.	Kuridala area, * 40 miles south of Cloncurry.
Uranophane.	As small narrow veins in folded slates and calcareous meta-sediments. Copper staining is associated.	Pelican, 43 miles south-west of Concurry.
Beta-uranotil and uraniferous magnetite.	In a calcite-actinolite-quartz rock.	Huxy, 30 miles west-north-west of Cloncurry.

*Samples from these localities have been examined inthe laboratory, but the occurrences have not been verified by a geological field inspection.

The distribution of uranium deposits in the area is similar to that of the copper deposits, although the former are somewhat more restricted and are less numerous. Uranium mineralisation has been found in only two deposits which were previously worked for copper, but minor amounts of copper are of common occurrence with the uranium. Other points of interest are the occurrence of small amounts of torbernite in association with cobalt and copper minerals at Mt. Cobalt, 70 miles south of Cloncurry, and the occurrence in the Mary Kathleen deposit of small, irregular bodies of pyrite, pyrrhotite, and chalcopyrite, which carry uraninite. Apart from these isolated minor instances, there is no close association between the occurrence of uranium and that of other metallic mineral deposits in the Cloncurry-Mount Isa area. Figure 2 shows the distribution of uranium occurrences in the area.

Development and reserves

Fourteen discoveries have been tested by diamond drilling, and a further seven by prospecting shafts and crosscuts. Of these, only Mary Kathleen has been proved a major deposit. Two other deposits have indicated reserves in excess of 100,000 tons. These are the Counter, with indicated reserves of 210,000 tons of 0.20 per cent. U_3O_8, and the Skal with 300,000 tons of 0.16 per cent. U_3O_8. Six of the remaining deposits have some indicated ore of a grade in excess of 0.25 per cent. U_3O_8. and may be worked by small scale, selective mining if the ore is amenable to treatment. Among these are the Milo, Easter Egg-Midnight and Duke deposits. At least seven others (including the Elaine Mary), which have had little or no underground exploration, may have to be added to this number (Brooks, 1956).

Several deposits, such as the Milo, have not been tested to sufficient depth for an adequate appraisal. With the anticipated commencement of production at the Mary Kathleen treatment plant in 1958, interest in exploration for new deposits and the development of known deposits should be renewed.

Uranium deposits of the Cloncurry-Mount Isa area

There are some interesting features which may be noted regarding these deposits. Only in the "davidite-type" occurrence is the primary uranium mineral visible in the hand-specimen. Where the primary mineral is uraninite it occurs as disseminated grains of microscopic size. With few exceptions, very little leaching has taken place, and the outcropping ore usually consists of primary minerals with subordinate secondary minerals, and the grade does not differ greatly from that of the ore at depth. Only at Milo has an appreciable amount of secondary uranium ore been located. No pitchblende fissure-vein type of deposit with accompanying base metal sulphides, such as commonly occur in some other uranium fields of the world, have been found. Nor do the deposits bear many similarities to those of the Northern Territory or South Australia.

Although the uranium deposits differ widely in mode of occurrence, they may be grouped into four main types. These are described in turn below and brief descriptions of a selection of the more important or typical deposits are given for each type. Other occurrences in the Cloncurry-Mount Isa area where uranium minerals have been identified are given in Table 1.

Mary Kathleen type

The occurrence of this type of deposit is confined to a meridional belt of highly metamorphosed and metasomatised calcareous and arenaceous sediments, five miles long and two miles wide lying between two major concordant intrusions of granite. These meta-sediments are characterised by the presence of zones of intensely garnetised rock and by the occurrence of uraninite, principally as inclusions in allanite, and other rare earth minerals which have replaced the garnet. The allanite, in itself, is practically non-uraniferous. The deposits are highly irregular in outline and grade, and it is only at Mary Kathleen, where mineralisation has been of great intensity, that a deposit of economic grade ore has been found. Four other deposits of this type were tested, but in none was a significant amount of economic grade ore found.

MARY KATHLEEN (Matheson and Searl 1956)

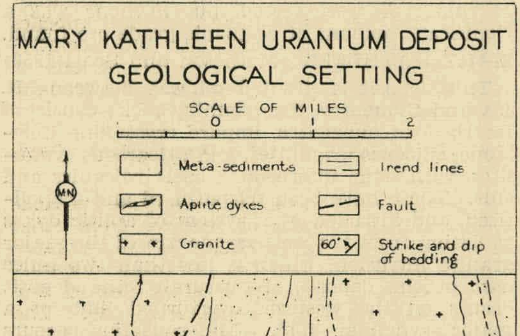
This deposit is situated midway between Mt. Isa and Cloncurry. The country rocks consist of interbedded quartzites, impure crystalline limestone, siliceous granulites, and calcareous granulites, with some interbedded basic volcanics and sills. These have been felspathised and scapolitised, and intruded by a system of aplitic dykes radiating from the western margin of the major granite intrusion, which is less than two miles east of the deposit. Structurally, the deposit occurs on the western, overturned limb of a major synclinal fold. Mineralisation occurs in an irregularly shaped, garnetised zone between two north-north-westerly trending shears.

The ore consists of dark brown to black allanite-rich rock partially or completely replacing reddish-brown garnet-diopside granulite. The uranium occurs as minute, disseminated, discrete grains of uraninite, intimately associated with the allanite, and to a lesser extent with the rare earth minerals stillwellite, caryocerite and rinkite. Gummite, uranophane and beta-uranotil occur together with uraninite in the zone of oxidation to a depth of 50ft., and incipient oxidation continues to a depth of 120ft. The uranium is in equilibrium throughout the deposit. The control of mineralisation appears to have been governed principally by the favourable host rock, the fault and joint system, and the distribution of garnetisation. Mary Kathleen is regarded by Matheson and Searl as a pyrometasomatic type of deposit derived from a late-stage emanation from granite intrusion.

The orebody consists of irregular, high-grade bodies of ore surrounded by low-grade material and interspersed with unmineralised areas. By selection in open-cut mining the material is classified into stockpiles of ore, marginal ore and waste. The reserves and grade of the deposit have not been made public, but the ore body has a maximum length of about 500ft. and a vertical extent as indicated by diamond drilling of approximately 400 feet. To date, exploratory diamond drillling has failed to locate any major extensions or repetitions of the deposit.

ELAINE DOROTHY

This deposit is similar in many ways to the Mary Kathleen deposit, but the intensity of mineralisation is much weaker. It occurs four miles to the south, near the closure of the major syncline with which Mary Kathleen is also associated. An irregular, discontinuous zone of garnetisation occurs over a length of 4,500ft. in calcareous and siliceous granulite and amphibolite. The localisation of mineralisation within the zone of garnetisation appears to be related to cross-faults and zones of fracturing. No mineragraphic reports on the ore are available, but the uranium occurs in a dense black rock comparable with the Mary Kathleen ore-rock. Two of the eleven drill holes on the deposit intersected economic grade ore, but the overall grade is very low. Figure 3 shows the geological setting of the Mary Kathleen and Elaine Dorothy deposits.



Counter type

This is a large, loosely defined group of deposits in which sub-types can be distinguished. All the deposits occur in the north-western section of the mineralised Precambrian belt of the Cloncurry-Mount Isa area in a succession of metamorphosed basic volcanics and interbedded arenaceous and calcareous sediments. The uranium usually occurs in sheared, impure, arenaceous meta-sediments, but in several cases it occurs in the interbedded meta-basalts or hornblende schists. The Skal group of deposits, north of Mount Isa, occurs within the same stratigraphic horizon and the Easter Egg group of deposits, north-west of Mount Isa, also occupies a single stratigraphic zone (Carter, 1955). The author agrees with Carter that the disposition of the occurrences is to be attributed to lithological and structural controls of epigenetic mineralisation, rather than to a syngenetic origin.

Very fine-grained uraninite has been identified in three deposits and brannerite in two other deposits. Elsewhere, the primary uranium mineral has not been identified, but in some cases it is intimately associated with magnetite or hematite. Calcite, hematite, magnetite, and quartz are the principal gangue minerals. In some cases the usual gangue minerals are absent, and the limits of uranium mineralisation can only be defined by a detection instrument due to the similarity of host rock and country rock. Five of the more important variants of this group are briefly described below.

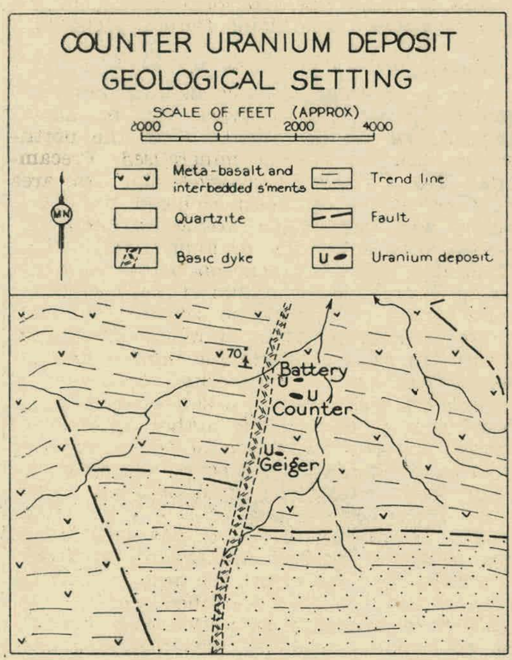
COUNTER

The deposit is situated nine miles north-west of Mount Isa. The country rocks consist of chloritised basalt, quartzite and greywacke intruded by basic dykes. The host rock is a lens of altered magnetite greywacke, and significant mineralisation is confined strictly to this rock type. Strike shearing along the margin of the lens of greywacke may also have been responsible to some extent for ore localisation.

Extremely fine grains of a mineral which is probably uraninite constitute the primary uranium mineralisation, and some uranophane occurs in the zone of weathering (McAndrew and Edwards, 1954). A minor basic dyke exhibiting weak radioactivity traverses the deposit and divides it into two sections. The main deposit is 105ft. long and 52ft. wide (420 tons per vertical foot), and the grade is 0.20 per cent. U_3O_8. Drill intersections to a depth of 500ft. indicate that the lode is reasonably uniform in depth. The principal impediment to the development of this deposit has been the difficult metallurgical problems associated with extraction of the uranium content from the ore. Figure 4 shows the geological setting of the Counter and adjacent uranium deposits.

SKAL

The discovery of this deposit, which is situated 20 miles north of Mount Isa, set off the prospecting boom of 1954. The uranium



mineralisation is associated with a zone of strike shearing. Much quartz has been introduced along this line of shearing, but the uranium is located principally in beds of quartzitic slate and calcareous quartzite. Adjacant chlorite schists are unmineralised. Where visible, the primary uranium mineralisation consists of blebs of brannerite with a little chalcopyrite and galena, in a gangue of calcite, magnetite and hematite. Generally, however, the primary uranium mineral occurs as very fine, disseminated grains associated with magnetite and hematite. A little carnotite is visible on fracture planes in the outcrop.

The deposit consists of two ore-shoots. The southern ore-shoot has a length of 370ft. and a width of 60ft. The surface grade is only 0.06 per cent. U_3O_8. but in an enriched zone, indicated by the 100ft. drill intersections, the grade is 0.16 per cent. U_3O_8. The northern ore-shoot is 300ft. long and 76ft. wide, and has a surface grade of 0.04 per cent. U_3O_8. The grade indicated by the 100ft. drill intersections is 0.17 per cent. U_3O_8. (Mount Isa Mines Ltd. data). The finely divided state of the uranium mineral and the presence of calcite would present problems in extracting the uranium content of the ore.

EASTER EGG-MIDNIGHT

Easter Egg and Midnight are adjacent leases on the same deposit, five miles north-west of Mount Isa. The deposit occurs in a horizon of allanite-quartz-hornblende schist, interbedded with mica schist and quartzite. The margin of the major Sybella Granite occurs two miles to the west. Minor amounts of quartz and calcite have been introduced in association with the uranium mineralisation. A little carnotite occurs in the outcrop, and the primary uranium mineral is probably uraninite, although positive identification has not been possible. Uraninite, occurring as aggregates of very small grains, has been identified at the Flat Tyre, a similar deposit two miles to the north (Campbell et al, unpublished data, 1955). In outcrop the main ore-shoot is 100 ft. long and 5½ ft. wide, and the grade is 0.18 per cent. U_3O_8.

Diamond drilling has indicated the continuity of values to a depth of over 100 ft., but the outline of the ore-shoot is highly irregular, and the distribution of uranium values is quite erratic.

DUKE

This is representative of a number of small deposits in the Calton Hills area, 55 miles north of Mount Isa. The uranium mineralisation occurs where faulting has shattered the meta-basalts against the more competent, lenticular quartzite beds. A notable feature of the Calton Hills area is the absence of outcropping granitic intrusions.

The highest grade ore in the Duke deposit consists of a dense, hematite-rich jasperoid rock with yellow secondary uranium mineral along joint cracks. The primary uranium mineral has not been identified, but appears to be associated with magnetite and hematite in extremely fine grains. The largest lens of ore is 40 ft. long and 9 ft. wide, and the grade is of the order of 0.30 per cent. U_3O_8.

ELAINE MARY (Carter, Unpublished Data, 1955)

A description of this deposit is included because it differs in a number of features from other occurrences in the Calton Hills area. It is situated 60 miles north of Mount Isa. Uranium mineralisation occurs in strongly fracture-cleaved and altered basalt, which is vesicular in part. The area is extensively faulted, and cross-faults displace the lode in two places. Small quartz-tourmaline veins occur at these points. Hematite, magnetite, calcite and a little chalcopyrite occur, and the uranium appears to be intimately associated with the iron oxide minerals. Sampling indicates that a small quantity of ore of a grade in excess of 0.25 per cent. U_3O_8 is present.

Torbernite-ironstone type

The occurrences grouped in this type are characterised by very low grade, disseminated uranium mineralisation in massive ironstone lodes, and by the presence of small concentrations of meta-torbernite ore in the zone of weathering. Low-grade copper mineralisation is commonly associated with the lodes, all of which occur in an area extending from Cloncurry to the southern limit of the mineralised Precambrian. The primary uranium mineral has not been definitely determined but, as with most Type 2 deposits, it occurs in a very finely divided state in association with iron oxide minerals. The deposits are considered to be of hydrothermal origin, although the possibility that they are of sedimentary origin with subse-

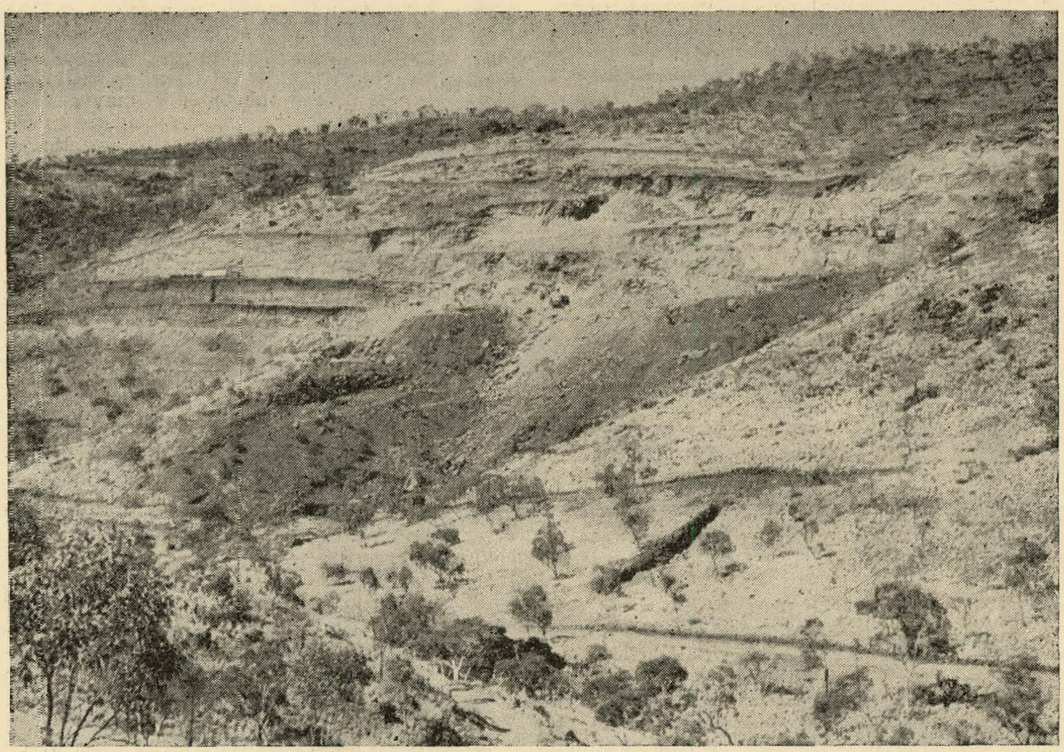


FIGURE 2:—General view Mary Kathleen open cut (K. W. Wolff photo).

quent metamorphism is not denied. Shearing and drag-folding were chiefly responsible for ore localisation.

TINBOLL (MONAKOFF)

This deposit previously produced a small quantity of copper carbonate ore. It is situated 13 miles north-east of Cloncurry, almost at the eastern limit of the outcropping Precambrian. The country rocks consist of amphibolite (metamorphosed basalt), with subordinate mica schist and quartzite. The deposit occurs in a sheared and drag-folded horizon of altered, ferruginous sandstone. Meta-torbernite and copper carbonates are associated with a variety of gangue minerals, including hematite, limonite, fluorite, barytes, psilomelane, quartz and garnet. Disseminated chalcopyrite, pyrite and magnetite, with purple fluorite, quartz and calcite are present in the primary ore (Campbell et al., unpublished data, 1955). Minute inclusions, which may be uraninite, occur in the fluorite. The surface grade is less than 0.05 per cent. U_3O_8. Diamond drilling tested the deposit to a depth of 250 ft. and showed no increase in the grade of primary ore over that at the surface. The main deposit has roughly 400 tons of ore per vertical foot, but even though the value of the deposit is enhanced by the presence of low-grade copper ore, the grade is too low to warrant development under present conditions.

MILO

The Milo prospect has the only appreciable concentration of secondary uranium ore located in the Cloncurry-Mount Isa area. It is situated 13 miles east-south-east of Mary Kathleen. The country rocks consist of slates, calc-silicate rocks and calcareous granulite. Meta-torbernite mineralisation occurs in a montmorillonite clay which appears to represent the alteration product of an acid igneous rock, possibly a rhyolite or rhyolitic tuff. The deposit occurs on the side of a prominent ridge which is over one mile in length. The ridge is composed of silicified slates which are jasperoid and gossanous in part. Weak radioactivity, due to small amounts of meta-torbernite, is associated with the ferruginous gossan material. The meta-torbernite is a supergene concentration in a favourable structural and/or chemical environment, and may have been derived from a disseminated primary source in the surrounding slates. A small tonnage of economic grade ore has been exposed. The higher grade section of torbernite ore is 110 ft. long and 10 ft. wide and has a grade of 0.135 per cent. U_3O_8. The deposit has been explored to a maximum depth of 25 ft. Further underground exploration is necessary, in particular to determine the significance of the primary mineralisation.

HELAFELLS

The deposit occurs in a succession of interbedded slates and quartzites, 20 miles south of Cloncurry. Several small lenses of radioactive ironstone occur, the largest being 50 ft. long and 5½ ft. wide. The lenses are conformable with the surrounding meta-sediments, and appear to occur along a line of shearing and drag-folding. Stains of malachite are widespread in the ironstone lenses. No systematic sampling has been carried out, but the surface grade, judged on the intensity of radioactivity, would be quite low.

Underground exploration of this deposit is being undertaken on the assumption that the torbernite-bearing ironstone bodies at the surface represent leached outcrops, and that the primary mineralisation may be of economic significance. However, exploration of comparable deposits elsewhere in the Cloncurry Field has given negative results.

MARIPOSA

The Mariposa prospect is situated towards the southern limit of the Precambrian mineral belt, 70 miles south of Cloncurry. The main occurrence of torbernite is along a sheared contact between quartzite and slate. Elsewhere, uranium occurs disseminated in a massive ironstone lode similar to the ironstone bodies on the Helafells. Only a very small amount of high-grade meta-torbernite ore has been exposed, and although no underground exploration has been carried out, it is doubtful if the primary mineralisation will prove to be of economic significance.

Davidite type

Thirteen occurrences of davidite-type minerals have been recorded in the central and eastern portions of the Cloncurry-Mount Isa area. What is referred to as davidite in the field usually consists of a mixture of davidite and ilmenite. Hematite may also be closely associated (Lawrence et al. 1957). In some cases it appears that the ilmenite may also be uraniferous. Elsewhere, uraniferous columbite and magnetite have been reported. Practically all the occurrences of davidite are in the Corella Formation, in which the characteristic rock type is re-crystallised, impure limestone. This formation outcrops extensively in the eastern half of the Precambrian in the Cloncurry-Mt. Isa area. The davidite is present as blebs or crystals up to three inches in length, and is associated in all but a few instances with granitic or pegmatitic intrusions. Brannerite and absite have been found with similar associations, but are of less frequent occurrence (see Table 1). Both thorium and uranium minerals occur in association with the Mica Creek pegmatites, south-south-west of Mount Isa.

Most of the deposits of this type show convincing evidence of a pegmatitic origin, although Lawrence (1957) considers that the uranium was deposited during geosynclinal sedimentation and was subsequently concentrated by regional metamorphism.

Little economic significance can be attached to the davidite-type deposits in view of their size and the erratic distribution of the uranium minerals.

SIX KANGAROOS

At the Six Kangaroos, which is 27 miles west-north-west of Cloncurry, davidite occurs as medium to very coarse-grained crystals and blebs in a lens of crystalline calcite. The calcite is interbedded with a calcite-actinolite-

FIGURE 3:—Open-cut bench, Mary Kathleen.

TABLE 2.—OTHER OCCURRENCES OF URANIUM OUTSIDE THE CLONCURRY-MT. ISA AREA.

Uranium Minerals	Occurrence and Association	Lease or Mine and Location.
Torbernite.	With wolfram, molybdenite and bismutite in a quartz-topaz-hematite lode in granite.	Lappa wolfram mine, 34 miles west of Herberton.
Torbernite, with traces of pitchblende.	With cassiterite and fluorite.	Gladstone tin mine, Stannary Hills, 13 miles north-west of Herberton.
Torbernite and a greenish-yellow uranium mineral.	As small flakes on joint faces in silicified granite containing arsenopyrite, chalcopyrite, etc.	Peacemaker tin-copper mine, Bakerville, seven miles north-west of Herberton.
Torbernite.	With segregations of biotite in a quartz, mica, topaz, and fluorite lode carrying wolfram and cassiterite.	Pine Log mine, Bakerville, seven miles north-west of Herberton.
Unidentified uranium mineral.	Associated with wolfram and tungstic ochre in a quartz lode in greisenised granite.	Tommy Burns prospect, Sunny Mount, 37 miles west of Herberton.
Torbernite.	——	King George mine, Fingertown, 26 miles south-west of Herberton.
Torbernite.	Found in mine dumps.	Anniversary copper mine, Herberton.
Unidentified mineral.	In an auriferous fissure vein consisting of quartz with sheared granitic material.	Cobar claim, six miles west-north-west of Georgetown.
Torbernite.	In oxidised gold-copper ore in granite.	Lucky Hit mine, Lolworth, 80 miles west of Charters Towers.
Torbernite.	Found in mine dumps.	Toomba gold mine, 40 miles west-north-west of Charters Towers.
Unidentified mineral.	With wolfram in quartz lode in aplite.	Wyarra, Ukalunda, 80 miles south-east of Charters Towers.
Unidentified mineral.	In a fissure vein in granite associated with chalcopyrite and molybdenite.	Old Mt. Dingle workings, Harper's Hill, three miles west of Wolca, on the Mt. Perry railway line.
Unidentified mineral.	With chalcopyrite, molybdenite, and scheelite in a quartz-carbonate-chlorite fissure vein in granite.	Globe copper mine,* six miles north-north-west of Mt. Perry.
Absite.	In pegmatite.	Boolboonda,* 12 miles north-north-east of Mt. Perry.
Pitchblende.	In granitic lode with pyrite, chalcopyrite, and molybdenite.	Mt. Perry district.*
Autunite and uranium ochre.	In vugs with white quartz.	Monsildale Creek,* north-north-east of Linville, 66 miles north-west of Brisbane.
Unidentified mineral.	In ferruginous sandstone of Mesozoic age.	Sapphire prospect, one mile south-west of Wycarbah railway station, Central Queensland.

*Samples from these localities have been examined in the laboratory, but the occurrences have not been verified by a geological field inspection.

scapolite rock and pegmatite and aplitic granite occur within a few hundred feet of the deposit. The davidite is irregularly and sparsely distributed through the calcite, and most of the surface radioactivity results from detrital davidite in the soil surrounding the deposit. A small amount of davidite is also reported from within a granite area to the south-east of the lease.

Other uranium deposits

WESTMORELAND (REDTREE) PROSPECT

This is the most recent discovery of uranium in Queensland. It was located late in 1956 by Mount Isa Mines Ltd. prospectors, following its discovery by an airborne scintillometer survey by the Bureau of Mineral Resources. It is situated six miles east of the Queensland-Northern Territory border and 95 miles west-north-west of Burketown. Secondary uranium mineralisation occurs in beds of coarse, felspathic sandstone towards the top of the flatly dipping Westmoreland Conglomerate. The uranium minerals identified are carnotite, meta-torbernite, renardite, soddyite, phosphuranylite, zennerite and uranospinite.†

They occur as coatings on quartz and other mineral grains, as concentrations related to joint planes, and as disseminated interstitial grains. Radioactivity occurs in several sandstone horizons over an extensive area, the largest continuous area of radioactivity measuring over 200,000 square feet. The grade is highly erratic and, overall, it is probably quite low. The deposit is sedimentary in type, and may be either syngenetic or a supergene concentration from an eroded source. Pitchblende occurs in the volcanic sequence which overlies the Westmoreland Conglomerate in deposits across the Northern Territory border from the Westmoreland area, and this provides a possible source for the secondary uranium minerals.

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† South Australia Department of Mines and Commonwealth Bureau of Mineral Resources determinations.

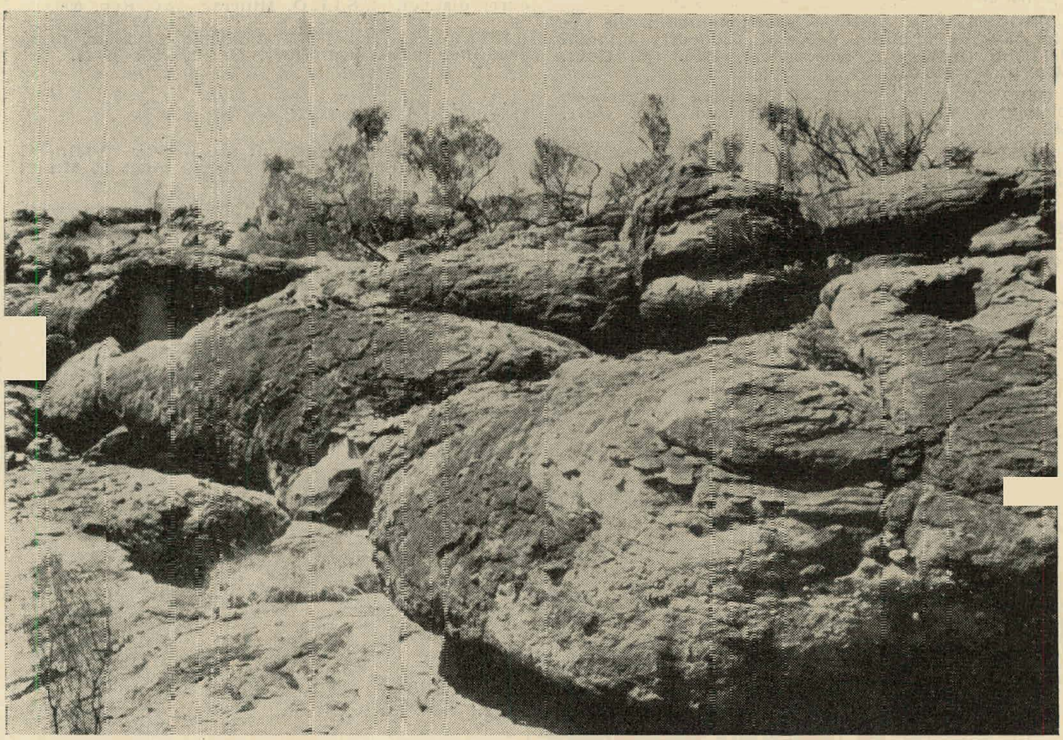

FIGURE 4:—Outcrop of Westmoreland Conglomerate, showing secondary uranium mineralisation.

LIMKIN'S PROSPECT, PERCYVILLE
(Wyatt, 1957)

This is situated five miles east-north-east of Percyville, in the Etheridge Mineral Field, North Queensland. It occurs in an area of coarse-grained granite with remnants of hornblende gneiss and granitised sediments. Uranium mineralisation is localised in two sub-parallel fractures in a large quartz vein and the adjacent kaolinised granite. The uranium minerals are torbernite, with a little autunite. Ten tons of ore with an estimated U_3O_8 content of over two per cent. have been raised from shallow workings, but the reserves are negligible and the prospects of finding further reserves are poor.

TARGINNIE PROSPECT

Targinnie is an old gold-mining centre, 14 miles north-west of Gladstone. Torbernite occurs in a narrow cupriferous quartz vein in granite. Recent workings are confined to shallow trenches along which the mineralised vein is four to six inches wide. Reserves are negligible, and there is little prospect of finding mineable deposits

Table 2 lists the other occurrences of uranium outside the Cloncurry-Mount Isa area. Most of them are in mines which have been previously worked for tungsten, tin, copper, molybdenum or bismuth. They are largely confined to the Herberton and Charters Towers districts of North-Eastern Queensland and the Mt. Perry district of South-Eastern Queensland. It is interesting to note the occurrence of an unidentified uranium mineral in intimate association with wolfram and tungsten ochre in two widely separated localities.

Acknowledgements

This paper is based on the author's field observations in North-Western Queensland, which extended over much of the period between August, 1954, and May, 1957, but frequent use has been made of published references and of unpublished reports by T. H. Connah, District Geologist, K. R. Levingston, Geologist, D. S. Carruthers, Assistant Geologist, and D. R. Davies, Assistant Geologist to the Chief Geologist, Geological Survey of Queensland. Mineralogical investigations have been carried out by the Commonwealth Scientific and Industrial Research Organisation, the South Australian Department of Mines, the United Kingdom Geological Survey, the Commonwealth Bureau of Mineral Resources, the University of Queensland Department of Mining Engineering, the N.S.W. University of Technology Department of Mining and Applied Geology, and the Geological Survey of Queensland.

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The Production of Uranium Ore Concentrates at Rum Jungle

By F. H. Baker*

Ore from White's deposit is crushed to minus half-inch by a jaw crusher, followed by a vibrating screen in closed circuit with a Symon's cone crusher. Milling is accomplished by a 7 ft. x 6 ft. Marcy ball mill in closed circuit with 60-inch Akins Classifier. Pulp overflows from the classifier at 30% solids. The fineness of grind is controlled at 65-70% minus 200 B.S.S.

Classifier overflow pulp is thickened to approximately 50% solids then pumped to pachucas for batch leaching under the following conditions:

(i) Pyrolusite is added to give 45 lb. of mangareal manganese dioxide per ton of ore.

(ii) Sulphuric Acid (98%) is added continuously to maintain the pH at 1.2 for the first four hours, then the pH is allowed to rise to 1.6 over the next four hours.

Liquid-solid separation is carried out by one stage of counter-current decantation, followed by two stages of filtration. The uranium-bearing liquor is treated by ion exchange, using anion exchange resin.

Complex uranyl sulphates are eluted from the resin, using a dilute solution of sodium chloride and sulphuric acid. Uranium is precipitated from rich eluate with magnesia slurry by neutralisation to pH 7.0. Uranium precipitate is thickened, then filtered on two stages of filtration. The second stage filter cake is dried to 5% H_2O on a continuous stainless steel belt passing through an oil-fired oven.

Introduction

In 1949, Mr. J. White discovered uranium mineralisation out-cropping at Rum Jungle, some 40 miles by air (70 by road) south of Darwin. The Commonwealth Government, through the Bureau of Mineral Resources, carried out the early development of the prospect to the stage of proving it to the production stage. Consolidated Zinc Proprietary agreed, for a nominal fee, to undertake uranium production at Rum Jungle on behalf of the Government, acting through the Australian Atomic Energy Commission. Consolidated Zinc Proprietary formed a new subsidiary, Territory Enterprises Pty. Ltd., to carry out these operations.

Underground mining was started in 1953, but for technical and economic reasons a change to open cut mining was made in 1954. Ore was stockpiled prior to processing in the treatment plant, which started operations in September, 1954. Output of uranium oxide in the latest financial year (1956/57) was 541,652 lb., which exceeded the rated capacity of the treatment plant.

The most important uranium mineral in the ore is uraninite. Other uranium minerals include torbernite and autunite. The copper minerals most common in the ore are chalcopyrite, chalcocite, and bornite. This paper deals only with the production of uranium concentrate, although copper is recovered by both the flotation and cementation processes.

Uranium mineralisation occurs in black slate and rocks, and the flaky nature of this rock causes trouble in crushing and grinding operations.

Ore dressing

Flowsheet No. 1 shows the ore dressing procedure.

Crushing and Sampling

A conventional crushing process reduces the minus 12 inch ore produced during mining operations to minus half inch.

The ore is delivered by five-ton trucks from the stockpile to the coarse ore bin, over which there is a 12in. x 18in. fixed grizzly. An apron feeder 30in. wide feeds the ore to a fixed bar grizzly with 1½in. bar spacings. The oversize is crushed to minus 1½in. in a 24in.x14in. Ruwolt jaw crusher, and joins the grizzly undersize for screening, which is carried out by an 8ft.x4ft. double deck cascade screen. The top and bottom decks of the screen are $1\frac{1}{16}$in. and ½in. woven wire mesh respectively. The plus ½in. material is reduced by a 3ft. Symons cone crusher set at ¼in. in closed circuit with the screen. The circulating load varies up to 150% of the new feed. The minus ½in. ore is stored in two bins, each of 500 tons capacity. The conveyor belts used in the crushing and grinding sections are 18in. wide.

Sampling of the ore is carried out on the minus ½in. material by a Vezin sampler, in three stages, each cut being 10% of the ore fed to the stage. After the first cut, a set of rolls 18in. dia. by 9in. reduces the large particles of ore to minus ¼in. mesh. This results in a final sample of 0.1% of the original tonnage.

Procedure for handling the Vezin cut is to apply Monsieur P. Gy's theory of ore sampling, and by this means the calculated sampling error in the final sample for assaying has been reduced to a minimum.

When operations started in 1954, trouble was experienced with the crushing equipment, which was not to handle wet ore containing more than 5% water. This problem was overcome by storing dry ore under cover for the wet season.

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* Treatment Plant, Rum Jungle. Manuscript received, March 10, 1958.

Ore grinding

A 7ft. diameter by 6ft. Marcy overflow ball mill operated in closed circuit with a 60in. diameter Akins spiral classifier grinds the ore to 65-70% minus 200 mesh B.S.S. The feed to the mill from the fine ore storage bins is regulated by a gate, which allows the ore to fall on to a reciprocating plate feeder 16in. wide, with a stroke adjustable up to six inches. A Blake Denison weightometer is used. This is maintained at an accuracy of the order $\lesssim$ 1%.

The sample for the head assay to the plant is taken at the classifier overflow by bulking half-hourly cuts of the pulp stream for 24 hours. The weight of dry ore milled is determined from the weightometer readings, and moisture determinations carried out on feed to the mill.

Mesh analyses are carried out daily on samples of mill feed and classifier overflow. A typical mesh analysis of ground ore is:

Mesh Size		Per Cent.
+ 60		3
− 60	+ 85	6
− 85	+100	6
−100	+120	5
−120	+150	3
−150	+200	11
−200		66
		100

Operating conditions during milling are:

Ball mill discharge: 75% solids approx.
Classifier overflow: 30% " "
New feed rate : Up to 25 t.p.h.

Some difficulty has been encountered from time to time with the variation in the grinding characteristics of the ore. The highly oxidised ore first mined from the open cut was very easy to grind. The harder subsurface ore is more difficult to grind, and the cause of the reduction in grinding rate is attributed to the hardness and "flaky" nature of the ore. Some variation in the size of grinding balls has been tried, and it has been found that 3in. diameter forged steel balls give the best results. Other sizes tried were 2in. and 4in. diameter. Highly siliceous purchased ores are difficult to grind in the mill, in which the optimum conditions have been set for White's ore.

The classifier overflow is pumped by a 6 by 4 split case Warman pump to the ore thickener.

Ore slurry thickening

The slurry from the classifier overflow is delivered to a 75ft. diameter Dorr torque thickener type "S." The slurry is flocculated with a 1% solution of a 50/50 mixture of gum guar-pearl glue with sodium pentachlorphenate ("Santobrite") added as a preservative. The addition of flocculant is 0.5 lb. total per ton of ore. Various mixtures of the two flocculants and other types of flocculant have been tried, but on an economic basis the mixture described above has given the best results. This mixture is used throughout the plant for aiding thickening and filtration of the ore.

The ore is thickened to a pulp containing approximately 50% solids, and is pumped via VM4 Dorrco Duplex pumps and 3 by 2 Warman pumps to the leaching pachucas. The excess water overflowing from the thickener is returned to the mill water pond for re-use.

Ore is fed to the thickener for about 11 hours a day, but the underflow is drawn off for 24 hours a day.

This requires careful control in operation, and the problem has been solved by keeping the thickener well loaded with pulp. Operating difficulties usually occur when the pulp load is allowed to fall.

Early operating difficulties encountered with the thickener were concerned with the accumulation of sand in the discharge cone. This was before the development of a technique of operating with a heavily loaded thickener, when a slightly coarser grind than usual would cause prolonged blockages in the underflow line.

Another early problem was the slow settling of the weathered surface ore. At that time little was known about flocculants, which have since been found to be very helpful in accelerating settling rates of finely divided solids. Oxidised ores gave an acid pulp of pH as low as 4.5 after grinding and it was necessary to add lime to the circuit to control the pH at 7. This is no longer found necessary.

Uranium extraction

Flowsheet No. 2 shows the process of extracting the uranium from the ore to make a final uranium oxide product. This is one of the most vital steps in the process. Leaching is carried out in rubber-lined pachucas 15ft. in diameter and 39ft. high, with a 60° conical bottom. Agitation is by compressed air, which enters at a pressure of 40 lb./sq. in. through a stainless steel sparger in the bottom of the pachuca. A stand pipe 18in. in diameter and 11ft. long assists in particle circulation.

Batches of approximately 100 tons of ore at approximately 50% solids to which pyrolusite (grade 60% manganese dioxide) equivalent to 45 lb. manganese dioxide per ton of ore has been added as a slurry, are leached with sulphuric acid at pH 1.1 for four hours. Then over the next four hours the pH is allowed to rise and is controlled at pH 1.6. The acid consumption under these conditions varies with the nature of the ore being treated but an average of approximately 250 lb. of 98.5% sulphuric acid is consumed per ton of ore. Leaching efficiency at present is of the order of 90-92%. Pyrolusite is necessary to oxidise uranium of valency 4 to uranium of valency 6, in which state it readily dissolves in dilute acid.

The catalytic action of iron present in solution is necessary to complete the reactions which may be written simply as follows:

$$MnO_2 + 4H^+ + 2e = Mn^{++} + 2H_2O + 1230 \text{ m v.}$$

This converts the iron in solution to the ferric state:

$$Fe^{++} = Fe^{+++} + e - 770 \text{ m v.}$$

which in turn oxidises the uranous salt to uranyl salt:

$$U^{++++} + 2H_2O = UO_2^{++} + 4H^+ + 2e \; 410 \text{ m. v.}$$

Much experimental work has been carried

TABLE 1

Factor		Level 1	Level 2
Acid Addition	A	200 lb. H_2SO_4/ton	250 lb. H_2SO_4/ton
*Pyrolusite Addition	P	60 lb./ton	90 lb./ton
Pulp Density	D	50% solids	55% solids
Leaching Time	T	8 hr.	16 hr.

*Grade of pyrolusite was 60% MnO_2.

out to determine optimum leaching conditions, which for the ore currently being treated are those set out above. Laboratory and pilot plant work carried out on samples of ore obtained during the early phases of mining indicated that a batch acid addition of 200lb. of sulphuric acid per ton of ore, followed by conditioning with limestone to pH 1.7, would give satisfactory U_3O_8 extractions. When operations started, the batch acid addition gave final pH values in excess of 1.7, and conditioning with limestone was not necessary. It was necessary to increase the acid addition to prevent reprecipitation of the U_3O_8 by keeping the pH down to 1.7.

In July, 1955, a laboratory scale leaching test factorially designed with four factors each at two levels was carried out with single replications, and yielded the information set out in Table 1.

The percentage of uranium leached from the ore and the final pH measurements were examined statistically and the following conclusions reached:

(i) Experimental and analytical errors, as shown by metallurgical balances, masked the effect of varying the levels of the factors. The pH measurements resulted in clear cut conclusions from the analysis of variance, probably because of the ease with which such measurements can be made.

(ii) By using a 16-hour leaching time instead of 8 hours, the extraction of U_3O_8 was increased by 3.3%. This was significant at the 1% level.

(iii) The final pH after 16 hours' leaching was 0.82, as against 0.66 after 8 hours' leaching. This was significant at the 5% level.

(iv) The effects of varying the levels of factors other than leaching time were not significant, and there were no significant interactions between factors for U_3O_8 extractions, although some minor interactions were found significant for the pH measurements.

Following this work it was decided to investigate leaching at constant pH, at the same time making note of e.m.f. readings. This work was carried out in the plant, and it was found that maximum U_3O_8 extraction from the ore being treated at that time was reached after four hours at pH 1.2. Since then, leaching has been carried out at constant pH, although conditions have been varied from time to time to meet the changing characteristics of the ore. It has not been found possible to find any relation between e.m.f. measurement and leaching efficiency.

Liquid-solids separation

After the uranium is in solution, the next step is to separate the uranium-bearing liquor from the leached ore as efficiently as possible. Using one stage of countercurrent decantation and two stages of filtration, the soluble loss is kept at about 3-4% of the total uranium input.

Countercurrent decantation

After leaching, the pulp is pumped by a 3 by 2 Warman pump to a 100-gallon mixing tank, where the 50% solids pulp is diluted with weak filtrate (i.e., liquor low in U_3O_8 content) to approximately 25% solids. Flocculant is added at the rate of 0.5lb. flocculant per ton of ore and the slurry is thickened in a 100ft. diameter rubber-lined Dorr torque thickener. The thickener overflow is pumped to the ion exchange section. The thickener underflow is pumped to the first stage filter storage pachuca.

The C.C.D. section was commissioned in May, 1957, and at the same time continuous leaching was introduced, using two pachucas in series. Because of the low leaching efficiencies caused by short circuiting of the pulp through the system, three pachucas in series were tried, but with little improvement in U_3O_8 extraction. The acid consumption was also high in both cases, and with three pachucas in series there were difficulties in keeping the pulp flowing between pachucas. It was necessary to operate with less than a 12in. margin in the first pachuca. Batch leaching was then resumed, with an all-round improvement in operating efficiency.

Filtration

The pulp from the first stage filter feed storage pachuca is pumped to a flocculating tank, where flocculant is added at the rate of 0.5lb. flocculant per ton of ore. The pulp flows by gravity to the first stage filters, which consist of one 11ft. 6in. diameter by 12ft. Oliver and one 12ft. diameter by 16ft. FEinc filter. Both filters are of the rotary drum type, and vacuum is created by Thompson vacuum pumps. The cake is washed with water acidified to pH 1.6 with sulphuric acid.

The filtrate is pumped to the weak filtrate holding tank, thence to the 100ft. diameter thickener, as mentioned previously. The cake is repulped to 50% solids with acidified water of pH 1.6, and pumped to the second stage filter feed storage pachuca.

The second stage filtering system is identical to the first stage, except that the cake is repulped to 40% solids with ordinary water, then neutralised with lime ready for the copper flotation section. The filtrate from the second stage joins the filtrate from the first stage.

The object in using acidified water is to prevent hydrolysis, which would cause precipitation of uranium, particularly if phosphate ions are present.

At the beginning of operations at Rum Jungle, only the Oliver filters were available for duty, and single stage filtration was carried out. With this system, the soluble U_3O_8 losses were high, being in the order of 10 to 15% of the total U_3O_8 input. After the FEinc filters were installed, two stage filtration became possible. The soluble losses fell to approximately 6% of the total input and, since the commissioning of C.C.D., this has been reduced to about 3-4%.

The coupling of the filtrate extraction pumps direct to the sand pressure clarifiers caused trouble in the early stages of operations. This arrangement was unsatisfactory, and an intermediate storage tank with a separate pump for pumping the liquor through the clarifiers was provided.

Blinding of the filter cloths because of an excessive amount of flocculant in the pulp was common and caused cloth life to be short. Rejuvenation of cloths by scrubbing and steam cleaning was not successful, but the research metallurgists found that by soaking a "blinded" cloth in a 5% solution of caustic soda for two hours, then lightly scrubbing it, and then soaking it for two hours in 5% sulphuric acid, followed by further scrubbing, the cloth was rejuvenated considerably. When applied in the plant the results were very pleasing. Now each filter cloth is treated once every two weeks.

Ion exchange

The overflow from the 100ft. diameter thickener is delivered to a settling cone, where a 1% solution of Separan flocculating reagent is added at the rate of approximately 3 c.c. per 100 gallons of liquor to help settle any solids, which are then returned to the filter section. The settling cone overflows into a storage tank from which the liquor is drawn off and pumped through either of two sand pressure clarifiers each 6ft. in diameter. The clarified liquor is delivered to storage tanks ready for ion exchange.

Prior to the installation of the settling cone, trouble was experienced with a quick build up in pressure across the clarifier, necessitating backwashing each clarifier several times a shift. With the present system it is necessary to backwash only one clarifier each shift. The importance of having clear liquor for the ion exchange columns cannot be over-emphasised.

There are four ion exchange columns of rubber-lined mild steel, each 7ft. in diameter by 12ft. overall height, with domed top and bottom. Each column contains 200 cu. ft. of wet settled resin (W.S.R.) resting on a bed of —⅜in. +¼in. silex. Except during backwashing to remove foreign solids from the bed, the liquor flows in at the top and out at the bottom. The pregnant liquor is pumped through three columns in series. The columns and resin content are designed so that when the effluent from the second column on line reaches a certain predetermined standard, approximately 0.05 gms. U_3O_8/litre (called the breakthrough level), the first column on line will be loaded to saturation. The third column in series is provided to take care of any U_3O_8 leakage past the second column. With four columns it is possible to take a column out of service for overhauling by carrying out adsorption on only two columns in series. When breakthrough is reached the first column in series is taken off line and the fourth column, previously on standby, becomes the third column in series.

Elution of the loaded column then proceeds, the solution used being 0.9 M sodium chloride, 0.09 M sulphuric acid containing 55-60 gm. of $SO_4^{=}/1$ tro when freshly prepared. The three-split elution technique is used whereby the first third of the eluate, being the richest in U_3O_8, is delivered to two of four precipitation tanks; the second third is returned to the circuit to become the first third in the next elution, and the

TABLE 2

Stage	Flow Rate g.p.m.	Time min.	Bed Vol.
1. Flush	53	22	0.94
2. Backwash	120	20	
3. First eluent to pregnant liquor	53	13	0.55
4. First eluent to precipitation	53	159	6.74
5. Second eluent to precipitation	53	13	0.55
6. (a) Second eluent to first eluent	53	172	7.29
(b) Third eluent to second eluent	53	163	6.90
7. Flush to second eluent	53	22	0.94
		584 = 9 hr. 44 min.	

One bed volume = 200 cu. ft.

Retention time for eluting solution = 9.44 min.

last third, being freshly prepared eluent, is returned to the circuit to become the second third in the next elution cycle. The flow rates and times used in each stage of the elution cycle are set out below:

Water, acidified to pH 1.6 with sulphuric acid, is used for stages 1, 2 and 7 to prevent hydrolysis in the columns. After uranium is precipitated from the rich eluate, the clear liquor is returned to the circuit to make up fresh eluent, except for 20% which is bled off to control the level of magnesium and sulphate ions in the eluting solution. The consumptions of sulphuric acid and sodium chloride in the eluent are 1.6lb. sulphuric acid and 4.7lb. sodium chloride per lb. of U_3O_8 produced.

The principle of the ion exchange process is that resin in the "chloride" form will absorb the complex uranium ions ($UO_2(SO_4)_2{=}$) and ($UO_2(SO_4)_3{}^{4}{-}$) in preference to other ions present in the pregnant liquor by replacement of the chloride ions which go out of the circuit with the barren liquor. The eluting solution removes the complex uranium ions from the resin, replacing them with chloride ions. Thus the eluate is a solution rich in uranium and contains very little of the impurities found in pregnant liquor. The resin used at Rum Jungle is De-acidite FF and loadings average 3.5lb. U_3O_8/cu. ft. W.S.R. Care is taken to see that the resin does not become poisoned by such substances as polythionates, titanium dioxide, silica, alumina.

At Rum Jungle resin poisoning has occurred by silica and alumina simultaneously, due probably to dirty pregnant liquor. After thoroughly cleaning the resin bed mechanically the loadings improved, and this emphasised the need for pregnant liquor that was as clear as could reasonably be attained.

A problem which arose in operations was the deposition of bismuth oxychloride on the walls of the pipes, resulting in restricted flow rates. This coincided with the treatment of certain purchased ores. At the time ordinary water was being used for stages 1, 2 and 7 of the elution cycle, and probably resulted in hydrolysis during those stages. Acidified water is now used during elution, and the pH of the system does not exceed 1.7 at any stage. Consequently no further trouble is expected from this cause.

Prior to April, 1957, two-split elution was practised, and the samples at the end of elution showed uranium contents of about 0.15 gm. U_3O_8/litre. Thus very little uranium was lost in the chloride wave which travels down the trailing column when a fresh loading cycle is started. Return of any of this liquor to the circuit was unnecessary. Since the introduction of three-split elution, the uranium content of the samples taken at the end of elution has been high at approximately 1.0 gm. U_3O_8/litre, and it is now necessary to return the barren liquor to the pregnant liquor circuit for the first 20 minutes of the loading cycle. This is called a "royal barren."

Regular maintenance of the automatic valves controlling the liquor flows is necessary to prevent liquor leaking from the pregnant-barren circuit to the elution circuit, and vice-versa.

Product precipitation and thickening

Uranium is precipitated from the rich eluate as magnesium diuranate by neutralising to pH 6.8-7.2 with a slurry of magnesia. The magnesia slurry contains approximately 25% solids, and the magnesia is 90% MgO. The precipitation is carried out in two agitated tanks, and a recording pH meter is used for accurate control. After precipitation, the product is allowed to settle in the tanks and the clear liquor is siphoned off, 80% being returned to the circuit for fresh eluent make-up and 20% being sent to waste for control of sulphate and magnesium ion level in the fresh eluent. Water is then added to the tank and, after agitation, the slurry is pumped to a 20ft. diameter thickener. A 1% solution of Separan is used to aid product settling and thickening. The underflow discharges from the thickener at approximately 20% solids and is pumped to the product filter.

Product filtering, drying and packing

The product thickener underflow is delivered to a surge tank, common to both first and second stage product filters, from which the slurry is continuously circulated to the first stage product filter, a 3ft. diameter by 4ft. rotary drum string discharge filter of monel metal construction, with a rubber-lined mild steel bowl. The cake is water washed, and then repulped with water and returned to the surge tank. By this means the chloride content of the product is kept within the maximum specification.

Slurry is fed from the surge tank to the second stage product filter, but no recirculation takes place. This filter is of similar dimensions and construction to the first stage filter. The filter cake is delivered into an extruder, which consists of a roller oscillating over a punched plate. All filtrate from the product filter is returned to the precipitation tanks.

During early operations it was found difficult to keep the chloride within specifications because of the absence of the first stage product filter. A 3ft. diameter by 1ft. rotary drum filter was installed in September, 1956, and used until October, 1957, but had insufficient capacity to keep the chloride content of the product within specification. Its action was supplemented by washing the product in the precipitation tanks and this was carried out after the barren eluate had been siphoned off. Water was added to the tank, and after agitation and settling of the product the liquor was siphoned off to waste. Two water washes were given to the product before pumping the slurry to the thickener. With the installation of the larger first stage filter this washing became unnecessary.

Product drying

The cake from the second stage filter is extruded through half-inch diameter holes on to an endless stainless steel conveyor belt. The product contains approximately 55% moisture as it comes off the filter, and is dried to 5% moisture by hot gases from an oil-fired burner. The gases circulate over the product, and return to the combustion chamber where portion is bled off to atmosphere to control the moisture content of the circulating gases. Any dust in the gases bled off is collected in an electrostatic precipitator. The product from the drying belt contains lumps of plus 1in. mesh, and to obtain a product of greater uniformity the product discharges from the belt to a set of toothed rolls 8in. in diameter by 6in., which crushes the product to minus ¾in.

The rolls deliver the product into a steam-jacketed screw conveyor, where some further drying takes place, accompanied by some reduction in particle size. The screw conveyor delivers the product to 44-gallon drums through a hood, connected to which is a cyclone integrated with the oven gas exhaust system. Large particles are trapped by the cyclone, and the fine dust is extracted by the electrostatic precipitator. A vibrator is used to increase the loading of the drums, which normally hold between 500 and 600lb. of product containing over 50% U_3O_8.

Product sampling

When operations first started, drums had an 8in. diameter hole in the top, and centre point auger sampling was practised. Investigations showed that by using a full open top drum and random sampling a more accurate sample would be obtained, the auger sampling method being quite satisfactory. Thus full open top drums and random sampling were introduced, and gave results which agreed closely with those of the buyers of the product using the same method. The introduction of toothed rolls and screw conveyor improved the results.

With the reduction in the size of particles of product a hypothesis was put forward that centre point sampling would be just as accurate as random sampling. Recent investigations have failed to substantiate this hypothesis, and random sampling is still practised. Results obtained by random sampling agree closely with results obtained by coning and quartering, whereas statistically significant differences have been found between random sampling and other suggested methods.

The laboratory

The laboratory staff analyse samples taken in the treatment plant for process control, most of these being daily composite samples. Uranium content is determined by radiometric, fluorimetric, gravimetric or colourimetric methods, depending on the type of sample. In addition, samples are also analysed for the geology, mining, pilot plant, engineering, and personnel departments.

Sample preparation is under the control of the Chief Chemist. The number of determinations made each week is approximately 1,000. Maintenance of instruments is carried out by the instrument section under the control of the Chief Chemist.

The pilot plant staff investigate current plant problems, as well as new techniques and processes for treating uranium-bearing ores.

Acknowledgement

The author wishes to thank the management of Territory Enterprises Pty. Ltd. and the Australian Atomic Energy Commission for permission to publish this paper.

Neutron Diffraction on HIFAR

By H. M. Sabine*

A brief account of the use of neutron diffraction as a complementary technique to X-ray diffraction in crystallographic analysis is given. The spectrometers to be installed on HIFAR are described and the experimental limitations of the technique discussed.

Introduction

Neutron diffraction has become an established technique for the study of solids but it will probably always remain a complementary technique to X-ray diffraction. In all investigations of atomic arrangement it is essential to obtain first all the information which can be obtained by X-ray methods.

This paper discusses the particular cases where neutron diffraction gives information unobtainable by X-rays and describes the neutron diffraction apparatus to be installed on HIFAR. No attempt is made to review the literature or to discuss the theory of neutron diffraction as these have been fully covered in a recent book (Bacon 1955) and in many publications, e.g., Wollan and Shull (1956), Bacon and Lonsdale (1953).

Uses of neutron diffraction

In investigating the structure of solids by X-ray or neutron beams the angular positions and intensities of the diffracted spectra are determined experimentally and the atomic arrangements deduced from these measurements. The positions of the spectra are determined by the unit cell dimensions and their intensity by the positions of the atoms in the unit cell and by the manner in which the incident radiation interacts with the individual atoms. It is the process of scattering radiation by atoms which is very different for neutrons and X-rays and gives rise to the special uses of neutron diffraction.

X-rays are scattered by the electronic cloud around the nucleus while in most cases the neutrons are scattered by the nucleus alone. The important exception to this is the case of the magnetic ions which give additional scattering because of the interaction between their unbalanced electron spins and the spin of the neutron. This exception will be discussed later.

The atomic scattering factor for X-rays is markedly angularly dependant, falling off according to a form factor with increasing angle between the incident and scattered radiation. This occurs because the X-ray wavelength is about the same as the linear dimensions of the electron cloud with the result that radiation scattered from different parts of the atom is in phase only in the forward direction. The neutron wavelength, however, is very much greater than the linear dimensions of the nucleus so the nucleus may be regarded as a point scatterer and the neutrons are scattered isotropically.

The freedom of neutron diffraction from the necessity to make assumptions about the form factor gives it an advantage over X-ray diffraction in some applications particularly in investigtions of the degree of ordering in alloys and in the study of gases and liquids.

There are three main applications for which neutron diffraction has decided advantages.

(i) Detection of light atoms in the presence of heavy atoms

The atomic scattering factor for X-rays increases regularly throughout the periodic table because of the continual increase in the number of extra-nuclear electrons. The ratio of the scattering factors for heavy atoms and light atoms is generally of the order of thirty or forty and since the diffracted intensity is roughly proportional to the square of the scattering factor, the observed spectra are very insensitive to the positions of the light atoms.

The scattering factor for neutrons, however, while it has a small steady increase due to increase in nuclear size, depends mainly on resonance effects which seem to vary in an arbitrary manner from atom to atom. As a result the neutron scattering factors for all atoms are roughly the same within a factor of 2 or 3 and the intensities of the diffraction spectra are sensitive to all the contributing atoms.

The heavy metal hydrides and carbides have been examined by neutrons and the positions of the light atoms, which cannot be located by X-rays, found. Studies of hydrogen bonds have been made in many substances, for example, sodium sesquicarbonate, α-resorcinol and benzene, using neutrons to locate the hydrogen atoms.

(ii) Differentiation between atoms of neighbouring atomic number

Because of the apparently random variation of neutron scattering factors with atomic number, ordering of alloys of elements close together in the periodic table can be detected. Thus ordering in alloys like FeCo and Mn_3Ni, undetectable by X-rays, can be easily found. However, ordering in Cu_3Au, which is easily shown by X-rays, cannot be found by neutrons since copper and gold happen to have identical scattering factors.

(iii) Magnetic scattering

The interaction between the atomic magnetic moment and the spin of the neutrons provides the only direct method of determining the magnetic state of a material. In a

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*Australian Atomic Energy Commission, Research Establishment. Manuscript received March 17, 1958.

paramagnetic material the atomic magnetic moments are randomly orientated and completely uncoupled so the magnetic scattering is completely incoherent and contributes only to the background.

However, in ferromagnetic, antiferromagnetic and ferrimagnetic substances coherence appears between amplitudes scattered from the ordered magnetic moments giving rise to diffraction peaks.

If the magnetic unit cell is the same size as the structure unit cell the magnetic diffraction peaks will coincide with the nuclear peaks and the total intensity will be the sum of the nuclear and magnetic intensities. In some substances the magnetic cell has twice the dimensions of the nuclear cell and the magnetic reflections occur at superlattice positions. This occurs, for example, in the antiferromagnetic oxides MnO, NiO and CoO.

The magnetic intensities depend on the orientation of the magnetic moments relative to the incident and scattered beams so it is possible in principle to determine the orientations of the magnetic moments relative to the crystallographic axes. In practice this determination is complicated by domain effects.

Since the interaction is between the electron and the neutron there is a magnetic form factor expressing the fall off in magnetic scattering factor with angle. However, unlike the X-ray case where the form factor is due to all the electrons in the atom, the magnetic form factor comes only from the electrons which contribute to the atomic magnetic moment. Therefore the magnetic form factor can be inverted to give the radial distribution of the magnetic electrons in the atom.

Limitations of neutron diffraction

The limitations on neutron diffraction arise almost completely from the relatively low fluxes available. Similar difficulties do not occur in X-ray diffraction since the number of quanta per unit area in a normal X-ray beam is greater than that in a neutron beam by a factor of 10^5. A compromise has always to be made between an incident beam intense enough to give observable diffraction spectra and resolution good enough to prevent considerable overlapping of the spectra.

Some of the more serious limitations are listed below. The numerical values given apply to reactors with a maximum thermal flux of 10^{12} neutrons cm^{-2} $sec.^{-1}$.

An attempt is made to assess the improvement which will be gained by using HIFAR where the thermal flux in the holes used for neutron diffraction is 7×10^{13} neutrons and cm^{-2} $sec.^{-1}$.

(i) Specimen Size

It is not possible to use very small crystals. Single crystals must be of the order of 2 by 2 by 5 mm. and powder specimens of about 7 cc. On HIFAR it is hoped to reduce the size of single specimens to ¾ by ¾ by 2 mm.

(ii) Resolution

Because of the low flux the monochromatisation cannot be very strict. At present specimens of lower symmetry than tetragonal cannot be examined as powders because of overlapping of the lines. Again it is hoped that the higher intensity from HIFAR will make more vigorous collimation possible and resolution equal to that of X-ray methods obtainable.

(iii) Experimental methods

Neutron diffraction data is much more laborious to collect than X-ray data. Photographic techniques are at present unsatisfactory and all information must be collected by counters. The weight of the counter and shielding (~ 2 cwt.) means that the counter must be restricted to movement in the horizontal plane. This restricts examination to one layer line unless the crystal can be adjusted to bring each layer in turn into the plane of the counter.

Because only one reflection can be examined at a time the collection of single crystal data is a laborious process. A reasonable estimate of the time taken is one hour for each reflection. It is unlikely that this time will be reduced.

The counter covers the same region in one traverse as an X-ray powder pattern but since the diffracted spectrum is only detected while the counter passes through the Debye-Scherrer ring only a small fraction of the diffracted neutrons are recorded. To compensate for this long exposure times are necessary. The average time is about 12 hours and in many cases it is necessary to average over several patterns.

(iv) Background intensity

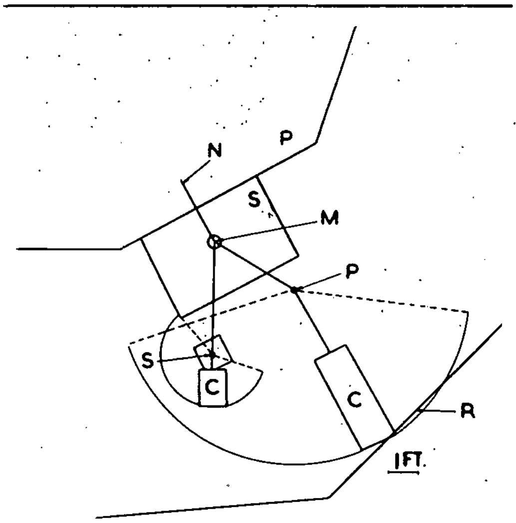
When a crystal contains an element having nuclear spin or a mixture of isotopes then there is a different scattering cross-section for each spin state and for each isotope. This mixture of scattering cross-sections gives rise to a high incoherent background which can be very troublesome. The incoherent scattering is particularly high in hydrogenous materials and in many cases single crystal methods must be used or the hydrogen replaced by deuterium.

Facilities on HIFAR

Thermal neutrons from a pile have a Maxwellian energy distribution with the peak at approximately 1.5A°. The experimental arrangement for neutron diffraction consists of a collimator to extract a parallel beam from the pile, a monochromator to select a beam of monochromatic neutrons and a spectrometer carrying the specimen and counter.

The collimator size is chosen to give the best compromise between intensity and resolution. It is essentially a steel tube 5 feet long with a cross-section of about 2 by 1 inches. With these collimator dimensions the angular spread in the incident beam is about ⅜°.

The monochromator consists of a large single crystal orientated so that the wavelength reflected is near the peak of the distribution curve. The crystals used are copper, lead and calcium fluoride, giving wavelengths of 0.81A°, 1.09A°



N	NEUTRON BEAM
P	PILE
S	SHIELDING
M	MONOCHROMATING CRYSTAL
P	POWDER SPECIMEN
S	SINGLE CRYSTAL SPECIMEN
C	COUNTER
R	RAILING ON FUEL ELEMENT STORAGE BLOCK

FIGURE 1: Plan view of spectrometers installed on pile face.

and 1.20A° respectively. Wavelengths on the short wavelength side of the peak are chosen to keep the second order contamination in the monochromatic beam to a minimum. In practice the second order component is about 2 per cent.

Two spectrometers will be installed on HIFAR. The first is a powder spectrometer. To obtain adequate resolution and low background the specimen-to-counter distance must be large and the counter well shielded. The specimen-to-counter distance is about 5 feet and the background should be about 30 counts per minute. This instrument cannot be used for single crystal work.

The second spectrometer is a single crystal instrument with a two to one ratio between counter rotation and crystal rotation. Since the counting rates for single crystals are much higher than for powders and the line width is governed by the specimen and not by the resolution of the instrument, a much smaller counter can be used and a specimen-to-counter distance of about 6 inches.

The method of recording the data will depend on the steadiness of the flux from HIFAR. Two methods can be used.

(i) The neutron pulses from the counter can be fed through a ratemeter on to a pen recorder and a continuous chart record obtained.

(ii) The counter can be moved a set angular interval when a certain number of counts have accumulated in a monitor placed in the beam from the monochromator. The number of counts received by the counter during this interval can be printed on tape.

The high penetrating power of neutrons makes neutron diffraction at high and low temperatures a simple matter, since reasonably thick furnaces and dewar flasks are transparent to neutrons.

The layout of apparatus at the pile face is shown in Figure I.

The whole apparatus must be adequately shielded to prevent unwanted neutrons entering the counter and to enable the operator to remain at the pile face for long periods without danger. On HIFAR, the radiation level in the working area will be 1 maximum permissible level.

Conclusion

The instruments described in this paper will be operating early in 1959. Initial work will be single crystal studies of the antiferromagnetic oxides NiO and CoO and powder studies of ceramic systems, but spectrometer time will be available for other problems.

Acknowledgements

The author wishes to thank Dr. G. E. Bacon and the staff of the Neutron Diffraction Group at Harwell for their help in planning the neutron diffraction facilities on HIFAR.

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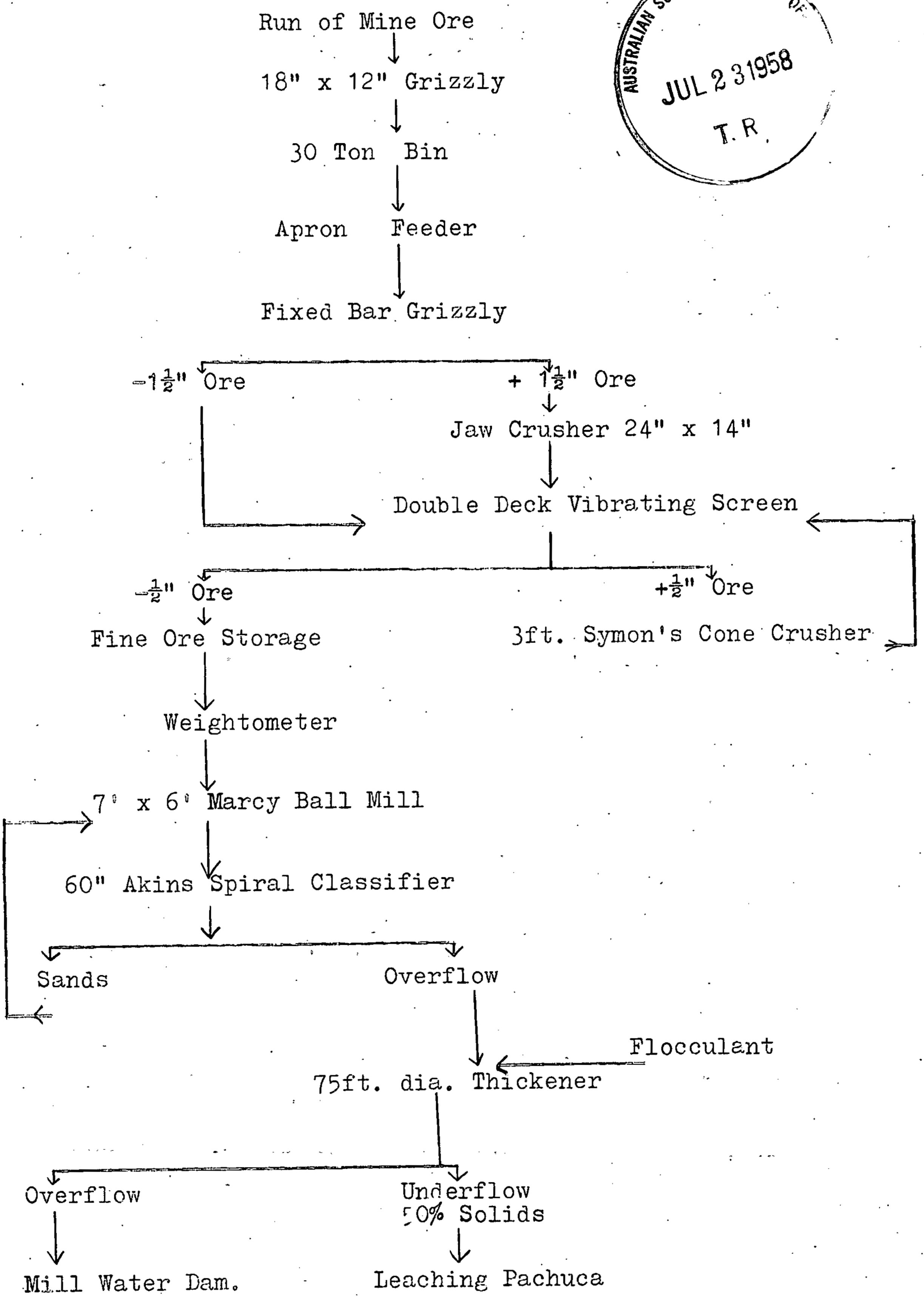

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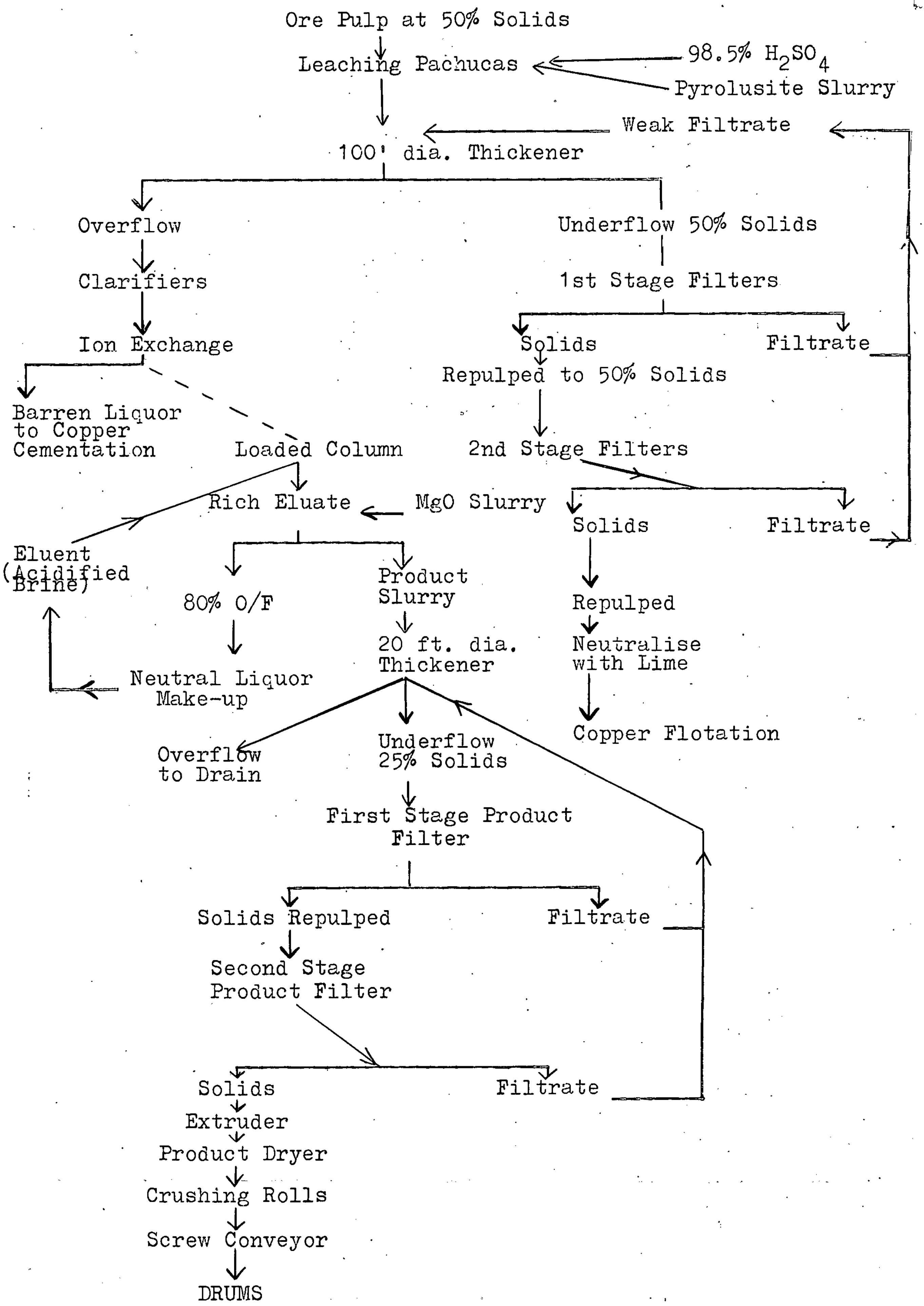

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Development of Nuclear Power in the United States

By L. H. Roddis, Jr.[*]

Introduction

Power reactor work in the United States extends back to the period of World War II. The major objective then was to construct and operate large-scale production facilities. Technical problems not contributing to this objective were largely ignored. Nevertheless, much was learned of great value to power reactor technology, particularly for water-cooled reactors. Most important of all perhaps was the introduction to nuclear work of many of the nation's most talented technical and scientific people. In July, 1945, there were more than 7,000 senior scientists and engineers employed at the installations operated by the Manhattan Engineer District, the wartime nuclear development agency.

Following the war there ensued a period of uncertainty as to the direction which atomic energy development would take. A large number of the scientists and engineers returned to their former employment.

Power reactor development work nevertheless proceeded. Efforts in the civilian power field, in naval ship propulsion, and in aircraft nuclear propulsion all began in 1946. None of the efforts begun that year came to fruition exactly as planned. The Atomic Energy Act of 1946 specifically charged the new Atomic Energy Commission with a responsibility in this field, and the Commission proceeded to organise for this purpose. In the fall of 1948, it established a Division of Reactor Development. Civilian power development was still at a very low level. A series of projects were initiated, which to a large extent reflected the interests and drives of particular individuals and groups.

The story of Rear-Admiral H. G. Rickover and the pressurized water reactor for the nuclear submarine **Nautilus** is too well known to need repeating. That effort began in a serious way in 1948.

Dr. Walter H. Zinn and his Chicago associates at the Argonne National Laboratory of the U.S. Atomic Energy Commission initiated work on a fast breeder reactor. This work led to construction at the National Reactor Testing Station of Experimental Breeder Reactor No 1, which in December, 1951, produced the world's first electricity from atomic energy, and which in 1953 provided the first demonstration of breeding.

Work at the Commission's Oak Ridge National Laboratory on a high flux reactor, led at first by Dr. Eugene Wigner, went through a variety of phases following its initial conception in 1947. It culminated in construction at the Idaho Testing Station of the Materials Testing Reactor, which commenced operation in 1952, and has been giving outstanding service ever since.

In 1952, the Homogeneous Reactor Experiment No. 1, which had been under construction at Oak Ridge National Laboratory, under the guidance of Dr. Alvin Weinberg, reached criticality.

From the point of view of realising at an early date the economic potentialities in the new technology, the early efforts in power reactor work, extensive though they were, suffered from not being part of a clearly articulated and co-ordinated program.

The Atomic Energy Commission recommended that this situation be remedied by formal establishment of a power reactor development program. In March, 1954, the U.S. Congress formally endorsed the Commission's outline of its objectives in the civilian power field for a period of about five years. This program centred on the technological development of five approaches selected by the Commission and its advisors as being the ones most likely to lead to economic nuclear power, and set forth the major projects involved in developing these five reactor types. The actual beginnings of the civilian power program occurred at this point in the U.S.

The five approaches and the respective projects were the following: Pressurized water—Shippingport Pressurized Water Reactor (PWR); boiling water — Experimental Boiling Water Reactor (EBWR); sodium-graphite — Sodium Reactor Experiment (SRE); fast breeder — Experimental Breeder Reactor No. 2 (EBR-2); and aqueous homogeneous—Homogeneous Reactor Experiment No. 2 (HRE-2).

All but the Shippingport plant were to be small-scale reactor experiments.

It is a source of very great satisfaction to us that by the end of 1957, less than four years after initiation of this so-called "five year program," four of these five reactors had gone critical.

The only one of the original reactors which remains to be completed is the EBR-2. This reactor was expected to be completed some time this year, but has been delayed by development difficulties, by decisions to place first priority on the shorter range Experimental Boiling Water Reactor, and by the need to obtain additional authorisation when the first cost estimates proved to be inadequate. Construction of this plant is under way, and it is now hoped it will be completed and in operation by 1960.

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[*] Division of Reactor Development, United States Atomic Energy Commission. Manuscript received April 17, 1958.

The year 1957 also witnessed the first criticality of the Organic Moderated Reactor Experiment. This was a variant not included in the original program, and emphasises the fact that in a young technology new ideas constantly emerge. We have sought to develop the best of these ideas as rapidly as possible.

I am now going to describe in some greater detail the reactors which form a part of the United States power reactor program. For convenience in reference, these are listed in an appendix to this paper.

Pressurized water reactors

Pressurized light water reactors have a strong lead in the power reactor program of the United States. More of them have been built, and more hours of successful operation have been logged for them than for any other type of reactor. This is due in part to the strong naval propulsion program, and in part to the ready availability of reasonably cheap uranium 235.

As you are aware, in pressurized water reactors the fuel elements containing fissionable material are surrounded by water under high pressure. The water serves both as a moderator and coolant to transfer heat. The water is heated by the fuel elements, and is pumped through a heat exchanger at about 500° Fahrenheit. In the heat exchanger the water gives up heat to generate steam, which operates a turbogenerator to produce electricity. The high pressure prevents water from boiling in the reactor and the primary system.

The pressurized water reactor at the Shippingport Atomic Power Station, the first full-scale nuclear power plant to operate in the United States, achieved criticality on December 2, 1957. By December 19 the station was producing electricity, and on December 23 it delivered on a test basis 60,000 kilowatts of electricity (net) to the Duquesne Light Company's system serving the Pittsburgh, Pennsylvania area. At this power level the Shippingport plant was producing more electricity than any other individual reactor in existence. When its second core is installed, the reactor is expected to produce 100,000 electrical kilowatts (gross).

The pressurized light water concept was selected for this first full-scale power reactor because considerable technology and experience with this concept had been developed in the design, construction, and operation of the Hanford production reactors and the Materials Testing Reactor, and in the naval program. The Shippingport plant employs a seed-blanket type core with something over one-half the heat energy coming from the natural UO_2 blanket.

Because the Shippingport plant is our first large-scale power reactor and because we desire to obtain scientific data, we did some things which may not be necessary in future plants. For example, the core is instrumented with special test equipment which will give us information and flexibility in designing future cores. I would like to underline the point that Shippingport will contribute much information useful in the design and operation of future nuclear power plants. A considerable additional investment in facilities and equipment has been put into this plant to obtain expected information.

Our operating experience thus far indicates that theoretical core calculations were accurate and that the core is performing according to specifications. Further, the blanket will last two seed core lifetimes. At present about 50 per cent. of the heat produced by the reactor is generated by the seed core, and about 50 per cent. by the blanket. As more full power hours are logged, a greater portion of thermal power will be produced by the blanket, with a significant portion of the total thermal power being derived from plutonium.

With the completion of acceptance tests, the Shippingport plant will develop information to assist in evaluating the design and performance of the core and to aid in the design of other cores and reactor plants. Tests will also be conducted to obtain data that will lead to simplification of pressurized water reactor auxiliary and control system operating procedures.

All technology developed by the Shippingport project is unclassified and has been rapidly and widely disseminated.*

Another operating nuclear power plant is the Army Package Power Reactor at Fort Belvoir, Virginia, which is a prototype of a small pressurized water reactor designed to produce 1,855 electrical kilowatts (net) for remote military installations, but also has potential for small central station applications. It achieved criticality on April 8, 1957. Since its dedication on April 29, the APPR has been supplying electrical power to the U.S. Army Engineer Research and Development Laboratories. A six-month acceptance test designed to establish the reliability of plant operation over an extended period, and to permit accumulation of operating data for designing additional plants of this type, was successfully completed. During this period the plant was operated at full power 21 days of each month, and at varying loads during the remainder of the month for the training of military personnel.

APPR performance thus far has been highly satisfactory. The reactor has demonstrated absolute stability, even over such extreme transients as complete loss of load, and has yielded much valuable operating information. By March 1 the plant had generated more than 7½ million kilowatt hours of electricity.

In addition to a large number of pressurized water nuclear power plants being built for the propulsion of naval vessels, both submarine and surface ships, two large size pressurized water nuclear power plants for central station use are under construction in the United States. These are the Consolidated Edison Plant of 275,000 electrical kilowatts being built at Indian Point, New York, and the Yankee Atomic Electric Plant of 134,000 electrical kilowatts being

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* (TID-LS-31, a list of unclassified reports on the Shippingport Pressurized Water Reactor, available from Technical Information Service Extension, Oak Ridge, Tennessee.)

built at Rowe, Massachusetts. In addition, a smaller 11,500 electrical kilowatt plant is under construction in Belgium by an American manufacturer, and several other plants generally similar to the Yankee Atomic Electric Plant have been announced for construction in Europe.

The first commercial merchant ship with nuclear power, the Nuclear Ship **Savannah,** will also employ a pressurized water plant. The **Savannah** is expected to be put under way in early 1960.

The technical feasibility and reliability of the pressurized water concept have been established beyond a doubt. The problem of making it economically competitive remains to be solved. The ultimate position of pressurized water reactors in the nuclear power industry will depend upon the success achieved in reducing the costs of power from these systems—success which can come only from operating experience.

Boiling water reactors

In the United States, the quality and completeness of boiling water technology is a close second to that of pressurized water technology, to which it is indeed most closely related. Intensive work on this concept is embodied in the Boiling Water Reactor Experiments (BORAX) conducted at the National Reactor Testing Station, Idaho, and in the Experimental Boiling Water Reactor at the Argonne National Laboratory, Lemont, Illinois. The inherent stability and safety of boiling water reactors were demonstrated by BORAX-1 operation during 1953 and 1954, prior to its deliberate destruction in a test to determine its reactions to extreme conditions.

Three additional BORAX experiments have been conducted to gain additional information from transient tests, to evaluate various new core combinations, and to investigate problems arising from steam radioactivity in the turbine generator.

BORAX-4, the current modification of this series of experiments, achieved criticality late in 1956. Its principal purpose is to test fuel elements made from mixed oxides of uranium and thorium. Early this year a series of tests on fuel element failures and other terminal experiments were conducted.

The Experimental Boiling Water Reactor at Argonne achieved criticality on December 6, 1956, and a month later achieved its rated power of 20,000 thermal kilowatts and 5,000 electrical kilowatts. Operation of the plant has been spectacularly successful. The operating level has since been boosted to 50,000 thermal kilowatts and later to 62,000 thermal kilowatts. This was done, without any change in the number of fuel elements or their arrangement in the reactor core, mainly by removing the control rods somewhat further than specified by the original design. J. M. Harrer, Project Manager of the EBWR reactor, will describe our operating history with this plant later.

A small privately-financed boiling water reactor built by General Electric as a prototype for the Commonwealth Edison Dresden Station is already in operation in California. Experimental operation of this unit has been very successful.

Indicative of the large effort in the development of the boiling water concept in the United States are four reactors scheduled for construction during the next few years. They are Commonwealth Edison Company's 180,000 electrical kilowatt Dresden Station, Rural Cooperative Power Association's 22,000 EKW plant, the Northern States Power Company's 66,000 EKW plant, and the Pacific Gas and Electric Company's 47,500 EKW plant.

Sodium-cooled reactors

Considerable work has been done on reactor systems using liquid sodium as a coolant.

Sodium remains a liquid at power plant temperatures and normal atmospheric pressures. Its melting point is 208° F.; it boils at 1,620° F. This range permits high temperatures for transfer of heat energy without the problem of boiling, even at atmospheric pressure. Sodium also is an excellent heat transfer medium.

The use of sodium is, however, not without certain disadvantages. It reacts strongly with oxygen and water and must therefore be isolated from them. Further, when exposed to neutrons, as in a reactor, it becomes a highly radioactive gamma-ray emitter with a half-life of about 15 hours, which creates a serious problem of radiation control.

Three general types of sodium-cooled reactors have been investigated in the United States. Reactors of the first type are operated on the basic thermal neutron spectrum and are generally referred to as sodium-graphite reactors, because graphite is the most common moderator. However, a sodium-cooled heavy water moderated reactor with thermal neutron spectrum is also incorporated in the Commission's program.

The second general type is operated on an intermediate neutron spectrum, which was at one time thought to possess special advantages from a neutron physics and control standpoint.

Fast-breeder reactors constitute the third group of sodium-cooled reactors. Fast-breeder reactors commonly employ sodium as coolant because it is an efficient coolant, and fits well with the requirements that the core of breeder reactors must be small in size and unmoderated. A reactor is said to "breed" when its operation produces a greater quantity of fissionable material than is consumed.

Sodium-thermal reactors

The sodium-graphite reactor was selected as one of the five original types for development and construction for civilian power purposes. The Sodium Reactor Experiment (SRE) achieved criticality in April, 1957. The reactor is designed to produce 20,000 kilowatts of heat, which will provide approximately 6,500 kilowatts of electricity.

Operation of the SRE was restricted during the first several months to the low power levels necessary to obtain basic reactor physics information. On July 12 the first power run was made, demonstrating the ability of the SRE to generate heat for conversion into electrical energy. During a subsequent power run a maximum output of 2 megawatts of electricity was reached, which is approximately one-third of the design power output. The power runs provided the occasion for other closely related tests, such as of shielding effectiveness, flow programming, and moderator cooling. Two substantial problems have become evident. First, thermal shock limitations have limited the operating power level. Secondly, the present metallic uranium fuel elements will not be satisfactory for long-term operations at the high temperatures required.

The plant was shut down in mid-January for several months for maintenance and modification. One of the major items being installed is an eddy current brake to slow down the coolant to a rate that matches the decay in power during a shutdown. This modification is necessary to reduce the thermal stresses and permit full power operation.

Following these modifications and subsequent low power tests, the SRE will be brought to full power, when we hope to find solutions for the problems which are still unsolved. Particular emphasis will be placed on achieving high integrated power to obtain information on the behavior of the various fuel elements being tested in the core. New types of fuel elements using uranium oxide and metallic alloys will be tested extensively.

The technology obtained in the design, construction, and operation of the SRE will be applicable to the 75,000-kilowatt nuclear power plant to be built under the AEC's power demonstration reactor program for the Consumers' Public Power District of Columbus, Nebraska.

A sodium-cooled heavy water moderated reactor plant is also under development in the United States by the firm of Nuclear Development Corporation of America. It is planned to install a Commission-owned small (10,000 electrical kilowatts) plant of this type on the system of the Chugach Electric Association of Anchorage, Alaska.

Sodium-intermediate reactors

The intermediate neutron energy range was at one time thought to allow a favorable combination of high specific power and high breeding ratio in the same reactor, because it had a greater available volume for coolant than the fast breeders, and because it was hoped that the neutron breeding ratio would be satisfactory. A serious effort was commenced in 1946 by the Commission at the Knolls Atomic Power Laboratory, Schenectady, New York, to develop this type of reactor, the intermediate power breeder. Unfortunately, as is now well known, further data on nuclear cross-sections showed that this concept would not be a satisfactory type of breeder reactor, and therefore in February, 1950, the research and development program was reshaped and re-directed toward the development and construction of a sodium intermediate spectrum submarine power plant. At the time, it was expected that this reactor would permit higher thermal efficiency and better features than the water-cooled plant. The greater maintenance difficulties were recognised at the time this development was undertaken. A land-based prototype of this power plant was completed in West Milton, New York, and began operation on March 20, 1955. In July the plant developed leaks in both superheaters and one steam generator. The superheaters were isolated, the generator repaired, and operations continued. In March, 1957, this plant was shut down as a result of the Navy's decision against using sodium-cooled reactor propulsion systems in additional naval vessels.

An essential duplicate of the land-based prototype was installed in the submarine **Seawolf**, which began sea trials on January 1, 1957. Leaks developed in the starboard steam generator during dock-side tests of the **Seawolf** in the summer of 1956. The plant was shut down to make necessary repairs. When operations were resumed and the superheaters isolated from the system, another leak developed and was also repaired. No difficulty was experienced with the nuclear power plant itself, and the safety of the plant was not impaired.

The **Seawolf** has been accepted for restricted service, and it has been recommended that her sodium plant ultimately be replaced with a pressurized water reactor. She has been operating satisfactorily in routine naval operations, and at the end of 1957 had steamed more than 29,000 miles—20,000 of them while submerged.

Fast breeder reactors

The very high heat release rates required in the small volume fast reactors dictate the use of a most efficient coolant. Sodium is a desirable and available material for this purpose.

Early work on fast reactors was initiated in 1945 at the Argonne National Laboratory, and it built the Experimental Breeder Reactor No. 1 (EBR-1) at the National Reactor Testing Station.

EBR-1 was designed primarily to study the physics and breeding characteristics of fast reactors, and secondly to study the technology of liquid metals as coolants. Construction of EBR-1 was approved in late 1947; the reactor went critical in August, 1951. In December of that year it produced the world's first electricity from atomic energy. Subsequently, it provided electricity for self-contained operation of the plant and associated laboratory facilities for some years.

Operation of EBR-1 on its first core from 1951 to 1954 demonstrated that breeding was feasible. Additional experiments were carried on with the second core until November 20, 1955, when the fuel rods melted as a result of an experiment concerned with measuring changes in reactivity as temperature of the fuel elements was increased. These experiments were undertaken to seek causes for power oscillations in the reactor. It was found that oscillations of power occurred when operating conditions were made severe; i.e., high power with reduced flow of coolant. Oscillations became more severe as power was increased or flow decreased. During the November 29 experiment, reactor power was made to rise with no flow of coolant, a calculated risk. EBR-1 was not shut down until powers exceeding those of normal operations were reached, and the overshoot created temperatures high enough to melt fuel rods in the core.

EBR-1 went critical with its third core late last year. This core will be used for detailed studies of the effect upon the stability of the reactor of bowing of fuel elements. In the previous core, heat caused fuel rods to bow. The new core is designed so that mechanical motions are rigidly controlled to prevent bowing.

Construction of the EBR-2 is now under way, with completion expected by early 1960, and full-power operation scheduled later that year. EBR-2, like EBR-1, will be unmoderated and liquid metal-cooled. It will have a thermal output of 62,500 kilowatts and an electrical capacity of 20,000 kilowatts.

EBR-2 will employ full-size components in its sodium system; for example, pumps and piping of 5,000 to 6,500 gallons per minute capacity. It is expected to yield much practical power operating information.

A major advance in the fast breeder concept is represented by the Enrico Fermi reactor being constructed by the Power Reactor Development Company near Monroe, Michigan. The reactor is being built under the Commission's power demonstration reactor program; completion is scheduled for late 1960. It will provide for an initial output of 90,000 electrical kilowatts net, and a possible later capacity of 140,000 kilowatts.

Homogeneous reactors

Fluid fuels in general offer a solution to the vexing problems of fuel element design, but they in turn have serious corrosion and maintenance problems of their own. And it was with this in mind that at a relatively early point in the development of nuclear reactors many investigators considered this one of the more promising approaches. Most work to date has been on aqueous types.

Aqueous homogeneous reactors

The Homogeneous Reactor Experiment No. 1 was subjected to extensive testing at Oak Ridge National Laboratory during a 24-month period between 1952 and 1954. During this time, it circulated liquid for about 4,500 hours and was critical for almost 2,000 hours. It was operated above 100 thermal kilowatts for 720 hours, and attained a maximum thermal power level of 1,600 kilowatts.

The reactor demonstrated an extremely high degree of inherent nuclear stability, a result of the large negative temperature coefficient of reactivity. It was therefore not necessary to use mechanical control rod mechanisms built into the experiment as an added safety device.

Operation of the HRE-1 also demonstrated that it was possible to construct leak-proof piping systems for fluid fuels, and to handle successfully the hydrogen, deuterium, and oxygen which result from the radiolytic decomposition of light and heavy water. Its operation also showed that the power output of this type of reactor was automatically responsive to the fluctuating demands of the electric load requirements of the turbogenerator.

Maintenance problems, severe in any reactor, but especially so in the extensive piping systems of the HRE-1, were overcome, frequently by the use of temporary shielding and special extention tools, without exposing personnel to more than allowable amounts of radioactivity.

Construction of the Homogeneous Reactor Experiment No. 2 (HRE-2) began in August, 1954, and was completed in the spring of 1956. It is a two-region, forced-circulation reactor operating on a heavy water solution of uranyl sulphate and, initially, with heavy water in the blanket. HRE-2 has a design thermal output of 5,000 kilowatts, and provisions for a small turbogenerator. The plant also includes chemical processing equipment for purifying irradiated sulphate fuel solutions.

During pre-operational testing in the summer of 1956, chloride ion contamination damaged the stainless-steel leak detection tubing and associated flanges. Repairs delayed nuclear operation of the reactor, but in the interim the reactor underwent extensive non-nuclear operation at design temperature, pressure, and uranium concentration with generally successful results.

Efforts are now under way to advance the concept from the experimental stage to fuller development. As a part of a joint effort of the AEC, the Pennsylvania Power and Light Company and the Westinghouse Corporation, investigations of technical and economic feasibility of a large-scale, single-region, aqueous slurry reactor for central station use have been in progress since 1955. The projected plant would have a capacity of 70,000 to 150,000 electric kilowatts. This plant would operate on the thorium U-233 fuel cycle. Thermal breeding may be possible in this unit.

Non-aqueous homogeneous reactors

In addition to the aqueous systems employing either light or heavy water, it is possible to have a number of other homogeneous type systems. At least four such systems have been investigated in the United States. These are:—

(i) The so-called liquid metal fuel reactor, which employs a solution or slurry of uranium or thorium in liquid bismuth.

(ii) The Los Alamos Power Reactor Experiment series which employs a uranium phosphate solution as coolant.

(iii) The fused salt systems based on solutions of various uranium salts in complex fluoride salts; and

(iv) The Los Alamos Molten Plutonium Reactor Experiment, which is based on a solution of plutonium in various metallic alloys.

All four of these systems offer the same basic advantages of the aqueous homogeneous systems — they possess freedom from fuel element fabrication problems, are not susceptible to radiation damage of the fuel, and possess inherent control advantages. As distinct from the aqueous systems, they are aimed at permitting higher temperature operation, and therefore more efficient thermal cycles, than possible with the aqueous systems. On the other hand, corrosion problems are much more serious than with the aqueous solutions and, in addition, start-up problems with frozen coolant are introduced.

The liquid metal fuel reactor concept has had development work under way for a number of years. The Liquid Metal Fuel Reactor Experiment (LMFRE) is currently the only project of this type. It is the outgrowth of extensive research and development studies by Brookhaven National Laboratory. These studies show that the liquid-metal fuel systems are feasible and have possibilities of achieving low-cost power. The Babcock and Wilcox Company has contracted to design, fabricate, assemble, and operate the LMFRE. Extensive development work in the areas of materials, components, and remote maintenance systems is being carried out. Evaluation of alternate designs is under way.

At the Los Alamos Scientific Laboratory reactor experiments have been undertaken involving aqueous homogeneous power reactors using phosphoric acid instead of sulphuric acid as the fuel solvent.

The first effort was Los Alamos Power Reactor Experiment No. 1 (LAPRE-1), designed for a thermal output of 2 megawatts. LAPRE-1 achieved criticality for short periods in October, 1956. Failure occurred in its gold cladding after relatively short periods of operation and the experiment was abandoned early in 1957.

A second experiment, LAPRE-2, now being constructed, is similar in design to LAPRE-1 except that it depends on thermal convection rather than on pumped circulation of the fluid.

At Los Alamos, development has also been proceeding on a small scale fast reactor which would use molten plutonium as fuel. Further work on this experiment, designated Los Alamos Molten Plutonium Reactor Experiment No. 1, is contingent on results of materials development work now under way.

A civilian power molten salt reactor program has been initiated at the Oak Ridge National Laboratory to use technology developed in the Aircraft Reactor Experiment, which operated in 1954. Molten salts as a reactor fuel could provide the benefits of a fluid fuel in an unpressurized reactor operating at temperatures high enough to generate 1,000° Fahrenheit steam, a desirable temperature for efficient production of power. The wide range of solubility of uranium, plutonium, and thorium fluorides in fluoride salts allows a variety of reactor types to be considered. The program in its present stage is directed toward the study and determination of rates of corrosion of various materials under contemplated operating temperatures and flow velocities, as well as design studies of power plants employing molten fluoride reactors.

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Gas-cooled reactors

Gas-cooled power reactors can be designed to generate steam in a secondary loop, or to use the primary gas coolant directly to operate a gas turbine. Temperatures of the order of 1,200° Fahrenheit are required to operate a gas turbine efficiently, and therefore this type of reactor requires special materials of construction. Somewhat lower temperatures are required for generating steam to operate a turbine, and thus the materials problems are less severe. The Calder Hall reactors in the United Kingdom are of this latter type of power reactor.

High-temperature approach

A substantial amount of high-temperature work utilising gas coolants has been done in the military Aircraft Nuclear Propulsion Program, which is contributing technology for civilian gas-cooled reactors. For example, the Heat Transfer Reactor Experiment No. 1 (HTRE-1), which went into operation at the National Reactor Testing Station in 1956 and was dismantled in 1957, employed this technology.

The Gas-Cooled Reactor Experiment (GCRE) being constructed as part of the Army Reactors Program at NRTS also involves high-temperature technology. Criticality is expected to be achieved in 1959. The GCRE is intended to develop engineering data for design and construction of gas-cooled military package power plants and small civilian central stations.

The Maritime Gas-Cooled Reactor (MGCR) would be a high-temperature, high-pressure gas-cooled reactor coupled with a closed-cycle gas turbine for ship propulsion and central station power. Investigation of the MGCR is a co-operative undertaking by the Commission and the Maritime Administration, Department of Commerce.

Low-temperature approach

Low-temperature, gas-cooled reactors are necessarily large and involve high capital costs. Although this application has received considerable study in the United States, the utility industry has shown little interest in it and there was no large-scale project involving this technology in the United States program until August, 1957, when Congress authorised a study leading toward a gas-cooled, graphite-moderated reactor, fuelled with natural uranium, to have an electrical capacity of 40,000 kilowatts. Construction of a reactor has not been authorised.

Organic-moderated reactors

Work on the Organic-Moderated Reactor Experiment (OMRE), a reactor not included in the original formulation of the Government's experimental program in 1954, developed as a modification of the pressurized water effort.

APPENDIX.—U.S. power reactors and power reactor experiments

TABLE 1 — PRESSURIZED WATER REACTORS.

Reactor and location	Electric Kilowatts (net)	Status	Startup
Full-Scale Civilian Reactors			
Shippingport Atomic Power Station, Shippingport, Pa.	60,000	Operated	1957
Consolidated Edison Co. of N.Y., Indian Point, N.Y.[1]	255,000	Being built	1960
Yankee Atomic Electric Co., Rowe, Massachusetts	134,000	Being built	1960
Nuclear Ship **Savannah**	14,900	Being built	1960
Carolinas-Virginia Nuclear Power Assoc., Parr Shoals, S.C.[2]	16,950	Under study	—
Large Scale Heavy Water Reactor	100,000	Under study	1962
Full-Scale, Prototype and Experimental Military Reactors			
Submarine USS **Nautilus** (SS(N)571)	—	Operated	1955
Submarine USS **Skate** (SS(N)578)	—	Operated	1957
Nautilus Land-Based Prototype (S1W) NRTS, Idaho	—	Operated	1953
16 Submarines (1 of which has 2 reactors)	—	Being built	—
Guided Missile Cruiser Long Beach (CG(N)160) — (2 reactors)	—	Being built	—
Destroyer Reactor Prototype (D1G) Schenectady, N.Y.	—	Authorisation requested	—
Large Ship Reactor Prototype (A1W) (2 reactors) NRTS, Idaho	—	Being built	1958
Submarine Advanced Reactor Prototype (S3G), West Milton, N.Y.	—	Being built	—
Small Submarine Reactor Prototype (S1C), Windsor, Conn.	—	Being built	—
Army Package Power Reactor No. 1 (APPR-1), Fort Belvoir, Va.	1,885	Operated	1957
Army Package Power Reactor No. 1a (APPR-1a), Alaska	1,689	Being built	1959
Aircraft Carrier (CVA(N)) — (8 reactors)	—	Being built	—

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[1] Includes 112,000 EKW conventional superheat.

[2] A pressurized heavy water reactor.

The reactor was authorised in the fall of 1955, and achieved criticality in September, 1957.

The OMRE is characterised by the use of an organic material, terphenyl, as moderator and coolant in place of water. Terphenyl has the ability to produce fairly high steam temperatures at modest reactor pressures, obviating the need for thick, expensive reactor pressure vessels employed in pressurized systems to prevent boiling. Terphenyl also has the further advantages that it does not cause corrosion problems, and does not become highly radioactive in the reactor.

The operation of OMRE will test these characteristics within the framework of actual reactor environment. If the tests prove favorable these factors may contribute to reduced capital costs. A major problem in the use of organic substances such as terphenyl, however, is their tendency to decompose under irradiation. Operation of OMRE will also provide the basis for measuring such decomposition, and the costs involved in replenishing the inventory required for adequate reactor operation.

During the month following first criticality of OMRE in September of last year, the temperature coefficient of reactivity under unpressurized conditions was determined, and found to be positive up to 430° Fahrenheit and negative above this point. Flux distribution measurements were also calculated.

Other measurements indicate that the void coefficient of reactivity is negative, and that the flow coefficient of reactivity is negligible.

During December, the reactor vessel head was installed and the reactor pressurized to 200 pounds per square inch. Temperature coefficients were recalculated, and were positive up to 440°

TABLE 2 — BOILING WATER REACTORS

Reactor and location	Electric Kilowatts (net)	Status	Startup
Civilian Reactor Experiments			
Boiling Reactor Experiment No. 1 (BORAX-1) NRTS, Idaho	None	Dismantled	1953
Boiling Reactor Experiment No. 4 (BORAX-4) NRTS, Idaho[1]	2,400	Operated	1956
Experimental Boiling Water Reactor (EBWR), Lemont, Illinois[2] ..	4,500	Operated	1956
Argonne Boiling Reactor (ARBOR) NRTS, Idaho	None	In design stage	1960
Full-Scale Civilian Reactors			
Vallecitos Boiling Water Reactor, Pleasanton, California	5,000	Operated	1957
Commonwealth Edison Dresden Station, Morris, Illinois	180,000	Being built	1960
Rural Cooperative Power Association, Elk River, Minn.[3]	22,000	Under negotiation	1960
Northern States Power Co.[4], Sioux Falls, S.D.	62,000	Contract let	1962
Pacific Gas and Electric Co.	47,500	Contract let	1962
Military Prototype and Experimental Reactors			
Argonne Low Power Reactor (ALPR) NRTS, Idaho	Very small	Being built	1958

[1] BORAX-2 and -3 were predecessor experiments in this series at the same location using some of the same equipment.

[2] Plant designed to produce 20 thermal megawatts but has successfully operated at powers up to 50 thermal megawatts with the original core loading.

[3] Includes about 7,000 EKW conventional superheat.

[4] Includes about 16,000 EKW conventional superheat.

Fahrenheit and negative above this point. Recent stability tests indicated that oscillations originally observed in reactor operation were due to the negative temperature coefficient of the coolant in the fuel channel, while the organic in the moderator region showed a positive coefficient. These oscillations, though not desirable, can be compensated, and the reactor

TABLE 3 — SODIUM COOLED REACTORS

Reactor and location	Electric Kilowatts (net)	Status	Startup
Civilian Reactor Experiments			
Sodium Reactor Experiment (SRE), Santa Susana, Calif.	6,000	Operated	1957
Experimental Breeder Reactor No. 1 (EBR-1) NRTS, Idaho	Very small	Operated	1951
Experimental Breeder Reactor No. 2. (EBR-2) NRTS, Idaho	16,500	Being built	1960
Full-Scale Civilian Reactors			
Power Reactor Development Co., Enrico, Fermi Plant, Monroe, Mich.	90,000	Being built	1960
Consumers' Public Power District, Hallam, Nebraska	73,000	Under development	1962
Chugach Electric Association, Anchorage, Alaska[1]	10,000	Design study	1962
Full-Scale and Prototype Military Reactors			
Submarine Intermediate Reactor Mark A (S1G), West Milton, N.Y.	—	Dismantled	1955
Submarine USS Seawolf (SS(N)575)	—	Operated	1956

[1] Heavy water moderated.

TABLE 4 — HOMOGENEOUS REACTORS

Reactor and location	Electric Kilowatts (net)	Status	Startup
Civilian Reactor Experiments			
Homogeneous Reactor Experiment No. 1 (HRE-1), Oak Ridge, Tenn.	Negligible	Dismantled	1952
Homogeneous Reactor Experiment No. 2 (HRE-2), Oak Ridge, Tenn.	Negligible	Operated	1952
Los Alamos Power Reactor Experiment No. 1 (LAPRE-1), Los Alamos, N.M.	None	Dismantled	1956
Los Alamos Power Reactor Experiment No. 2 (LAPRE-2), Los Alamos, N.M.	None	Being built	1958
Los Alamos Molten Plutonium Reactor Experiment No. 1 (LAMPRE-1), Los Alamos, N.M.	None	Under development	1960
Liquid Metal Fuel Reactor Experiment (LMFRE)	None	Under design	—
Full-Scale Civilian Power Reactors			
Wolverine Electric Cooperative, Hersey, Michigan	9,560	Cost study	—
Pennsylvania Power & Light Co.	70-150,000	Under development	1963
Military Experiment			
Aircraft Reactor Experiment (ARE), Oak Ridge, Tennessee	None	Dismantled	1954

operated at a steady power level with either manual or automatic rod programming.

The reactor is now being operated at a steady n-13 megawatt level to gather data on the organic decomposition rate.

Further development of this concept is planned as a joint effort between the AEC and the City of Piqua, Ohio, with the construction of a power demonstration nuclear station with an electrical capacity of 12,500 kilowatts.

Plutonium recycle program

Work is proceeding in an effort to demonstrate the feasibility of recycling plutonium as fuel in thermal heterogeneous reactor systems. If successful, a system recycling plutonium generated in its own operation will be able to produce three to four times as much power from a given quantity of natural uranium, or even of a somewhat depleted uranium, as would be possible without recycling. By this method, thermal reactors with auxiliary chemical separations and fuel fabrication facilities could be designed to operate continuously without dependence on a virgin supply of enriched uranium. A reactor system of this type might be useful also for nations with raw uranium resources, but without gaseous diffusion plants.

Construction of an experimental Plutonium Recycle Test Reactor at the Commission's Hanford Works, and of a facility to develop plutonium-bearing fuel elements for the plutonium recycle program, was initiated early this year.

Further data on plutonium as fuel will be obtained from the Materials Testing Reactor by using a full charge of plutonium-aluminium alloy fuel elements currently being fabricated at Hanford.

Final remarks

The large deposits of fossil fuels—coal, oil. and natural gas—which abound in the United States fortunately have resulted in a very low power cost over most of this country. For this reason, the requirements to be met for producing economic nuclear power in the U.S. are substantially more stringent than those in most other countries. Largely because of this situation, the United States has a program in which simultaneous efforts are being made in many directions aimed at developing a nuclear power system which can be made economic in the near future in the U.S.

TABLE 5.—OTHER ELECTRIC REACTORS

Reactor and location	Type	Kilowatts (net)	Status	Startup
Civilian Reactor Experiments				
Organic Moderated Reactor Experiment (OMRE), NRTS, Idaho	Organic moderated	None	Operated	1957
Full-Scale Civilian Power Reactors				
City of Piqua, Ohio	Organic moderated	11,400	Under negotiation	1961
East Central and Florida West Coast Nuclear Groups	Gas cooled	46,000	Under study	1963
Gas Cooled Power Reactor NRTS, Idaho	Gas cooled	55,000	Under study	1962
Maritime Gas Cooled Reactor (MGCR)	Gas cooled	15,000	Under development	——
Military Prototypes and Experiments				
Heat Transfer Reactor Experiment No. 1 (HTRE-1) NRTS, Idaho	Gas cooled	None	Dismantled	1954
Gas Cooled Reactor Experiment (GCRE). NRTS. Idaho	Gas cooled	None	Being built	1959

Atomic Power in Australia— Some Economic Considerations

Commonwealth Bank Discussion Group[*]

The possible applications of nuclear power are considered as an alternative to "coal" power, on which is based 80 per cent. of Australia's electricity generation. In some localities, such as Mount Isa or South Australia, special circumstances may favour its adoption in a relatively small way within the next decade or two. The general adoption of nuclear power in Australia before it reaches comparative cost status with "coal" power would not seem warranted, particularly if extensive redundancy in the coal industry were involved. The technical possibilities more than justify present research and training programs, but the claim of the power industry to the additional resources necessary for general conversion to a nuclear basis may not be very great. The balance of payments aspects and the possible security aspects of extensive national reliance on overseas fuel supply sources would need to be considered.

Introduction

To date, the impact on Australia of atomic energy advances overseas has been manifest mainly in:

- (i) The search for, discovery, and subsequent exploitation of uranium deposits and deposits of certain other minerals associated with atomic development;
- (ii) The impetus that overseas scientific progress has given to the training of Australian scientists and technicians in the schools and universities, in Government organisations such as the Atomic Energy Commission, and in industry generally;
- (iii) The adoption of overseas innovations in the use of radio-isotopes in medicine, industry, and agriculture.

These developments are important economically, and there is little doubt their importance will increase. As yet, the possibilities of adapting atomic power to the propulsion of ships, trains, aeroplanes, and motor vehicles in commercial usage have not, so far as one can gather, greatly affected industrial planning in Australia. On the other hand, however, considerable attention has been directed towards the prospects of using atomic processes for electricity generation in Australia, perhaps in the very near future, and this is the aspect with which this paper is concerned.

First, the power generation industry in Australia will be briefly considered.

Power generation in Australia

Power generation is undoubtedly one of the most important service industries in Australia, and is likely to become even more important in the future. Whereas the Australian gross national product in real terms has risen at an average rate of the order of three per cent. per annum during the past 10 years, the annual output of electrical energy in Australia has expanded at something like 10 per cent. per annum.

In Australia, responsibility for the public power supply is mainly a Government function. The States have electrical networks which are more or less independent of each other, although there is substantial inter-connection within State boundaries. In most States, regulatory authority over the State network is vested in a Government commission and, in some States, these commissions also act in varying degrees as constructing, generating, distributing, and retailing bodies for electric power. In the Commonwealth territories (Australian Capital Territory, Northern Territory, and Territory of Papua and New Guinea) electricity supply in the more important centres is provided by the Commonwealth Government, which either arranges the necessary generation, or purchases bulk power from a State authority. The Snowy Mountains Hydro-Electric Authority, a Commonwealth power producing authority, is also empowered to sell in bulk to State authorities power generated in its stations.

The great bulk of Australia's power generation plant is thermal (coal-fired) equipment located around the coal fields of the east coast and south-west corner. However, fairly extensive use is also made of hydro-electric power (c.f. Tasmania and the Snowy Mountains Hydro-Electric Authority) and there is some trade in coal between States for use in power stations (e.g., N.S.W. ships coal to South Australia). Internal combustion plant is also used quite widely in some commercial installations, and in comparatively small communal plants in the lesser towns.

Table 1 shows an analysis of 1955/56 electrical capacity and output in Australia, according to types of generating plant used:

TABLE 1.—ELECTRICITY IN AUSTRALIA, 1955/56.

ELECTRICITY SUPPLY UNDERTAKINGS ONLY

Type of Plant	Installed Capacity kW million	Installed Capacity % of total	Electricity Generated kWh million	Electricity Generated % of total
Steam	2.97	78	12,211	80
Hydro (a)	0.65	17	2,712	18
Internal Combustion	0.18	5	374	2
Total	3.80	100	15,299	100

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[*] Manuscript received March 27, 1958.

(a) Includes Snowy Mountains Hydro-Electric Authority. 186 million kWh from 60,000 kW installed.

Steam plant is clearly the predominant type, fuelled mainly by coal, which, in 1955/56, accounted for more than 96 per cent. of the total weight of fuel used in steam generating plant. At the same time it should be recognised that, as a use for coal, electricity generation is of considerable importance. On 1954/55 figures, approximately 30 per cent. of black coal and 60 per cent. of brown coal produced in Australia was used as fuel for steam generating plant.

Within this general setting, the prospects of using nuclear power for electricity generation can be considered.

Use of nuclear power in Australia

Obviously one of the fundamental aspects of the problem is that of comparative costs. In this field, estimates have been made for a number of different reactor systems, but the present choice appears to be mainly between the British gas-cooled natural uranium reactor (Calder Hall type), and the American water-cooled enriched uranium reactor. This paper is based mainly on the operating estimates and forecasts of Calder Hall type stations. However, in view of the uncertainties to which this data is subject, and of the possibility that other systems adapted to commercial usage will prove superior to the Calder Hall type, it seems necessary to make at least one reservation before proceeding to use the figures. This reservation consists of the assumption that the costs of nuclear power in Australia will prove eventually to be fairly close to present estimates and that the means of using nuclear power will not depart greatly from current expectations. If, for example, through the use of fast breeder reactors or the harnessing of fusion processes, nuclear power were to become available in much greater abundance and/or at much lower costs than at present envisaged in the projections of Calder Hall costs, the analysis would undoubtedly need substantial revision.

In terms of current expectation however, it appears that the Calder Hall type power station might cost about £S.130 per kW to build, and about 0.66d. per kWh to run (Jukes, 1957). Comparable costs in Australia are difficult to estimate, but minimum figures obtained from a simple conversion of the sterling costs are £A.160 per kW to build and 0.8d. per kWh to operate.

By way of comparison, the capital cost of a large modern coal-fired station has been estimated at about £A.80 per kW, and the base load running cost in reasonable proximity to good coal supplies is about 0.6d. per kWh (Department of National Development, 1955), i.e., about half the capital cost and threequarters of the running cost of a nuclear station. In localities further from good coal supplies, running costs would be correspondingly higher. On the other hand, although the capital cost of a hydro-electric station would probably be, on the average, somewhat higher than that of a coal-fired steam station of similar size (of the order of £A100 per kW), its running costs would be significantly lower (0.3d.-0.4d. Australian) per kWh or perhaps even less.

However, further development of hydro-electric resources in Australia appears to be limited, and mostly encompassed by present planning. Internal combustion plant, as mentioned above, is mainly used in small installations away from the main industrial centres, and although running costs are comparatively high, plants of the Calder Hall type could scarcely be regarded as possible substitutes, mainly on the score of economic size.

Looking to the future, it appears that technical prospects for the reduction of nuclear power costs over the next decade or two are bright. Allowing for some further progress in reducing conventional power costs, it had been estimated that nuclear power might reach competitive cost status with Australian coal-fired steam stations by about 1975.* Even then the capital cost of the nuclear station would probably still be higher than that of the coal-fired station. However, it would seem that a decision would need to be taken by power authorities on the possible use of nuclear stations in place of coal-fired stations for electricity generation from about 1975. In terms of Calder Hall type stations, to which the present analysis is largely restricted, this would involve a choice between natural uranium and coal as the principal fuel. Upon such considerations, therefore, will hinge the main economic significance of a changeover.

The change to nuclear power

In terms of the above analysis the economic implications of a decision to switch from coal to uranium-fired power stations should take account of three distinct possibilities, which may be generalised in the following terms:

(i) An independent decision to change over before 1975 taken by one or more remote towns or industrial projects currently using high-cost conventional power—*"isolated change-over"*;

(ii) An independent decision to change-over before 1975 taken by a single electricity supply authority facing comparatively high conventional fuel costs in expanding capacity—*"limited change-over"*;

(iii) A general decision taken by the main electricity supply authorities to change over about 1975—*"general change-over."*

In any of these circumstances, it is presumed that the decision to change-over would mean that existing plant would not be scrapped but that a growing proportion of new and replacement base-load plant would be of the nuclear type.

Some of the economic implications in these three situations will be examined.

Isolated changeover

There are some heavily industrialised areas of Australia which are not well supplied with fuel for power generation, and where the costs of

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* e.g., statement by Minister for National Development on October 19, 1957:—"It is thought that by 1975 nuclear power costs in Australia may be competitive with conventional power costs."

transporting the necessary fuel (or transmitting the necessary electricity) make power costs comparatively high. An example is the mining town of Mount Isa, in Queensland (Crivelli, 1957). There, power generation costs are approximately 2d. per kWh because of the high transportation costs of coal, yet expected future power requirements may justify the installation of a comparatively large base load station (at least 60MW) within a few years.

Because it has been estimated that a natural uranium reactor of this size could probably be operated at rates significantly less than 2d. per kWh, it appears that Mount Isa could satisfy the twin conditions of size and cost.

From the economic viewpoint, such a saving of power cost could be of great benefit to the Mount Isa project. Even though the capital costs of nuclear plant would be extremely high (estimated at about £A.15 million for a 60 MW station—probably at least three times that of a comparable coal-fired station) the amounts involved would not seem to be beyond the resources of the project.

Such a nuclear station would probably be a greater charge on the balance of payments than a conventional station, because of a higher foreign exchange component of capital costs and because fuel must be imported (discussed below). These aspects would need to be considered in relation to other priorities, although the actual amounts involved may be relatively insignificant from the national viewpoint. A change from coal to uranium at Mount Isa would have little effect on the Australian coal industry. Present coal requirements of Mount Isa are only 70,000 tons per year.

Another matter of importance would be the arrangements to be made regarding the supply of fabricated fuel elements and the reprocessing of the discharged fuel. There is, in Australia, a ready, although at present limited, supply of uranium ore, but it would need considerable processing before it could be used in a power reactor. There does not appear to be any technical barrier to Australia's setting up plant to process fuel for natural uranium reactors, and to reprocess the radioactive residue. However, construction of such plant to service isolated and comparatively small installations could scarcely be undertaken on anything like economic lines (Jukes 1957). Furthermore, like the reactor itself, such plant would probably have a comparatively high foreign exchange cost component.

For such installations, therefore, it would seem necessary to arrange fuel purchases overseas, and probably also have spent fuel reprocessed overseas. As mentioned above, this would be a new charge on the balance of payments, but for Mount Isa alone the amounts involved would scarcely be significant from the national viewpoint. An arrangement to import fuel could, of course, work extremely well from the practical viewpoint. But the possible hazards involved in individual firms relying extensively for vital power supplies on overseas sources of supply of a material likely for many years to be of considerable international political significance would need to be carefully considered by such firms.

Limited changeover

Extending the analysis of the isolated changeover, it is apparent that the whole State of South Australia, for example, is somewhat less favourably placed than most of the other States in regard to conventional fuel for electricity generation. It has no hydro-electric power, and its limited coal resources are currently supplemented by imports from New South Wales. As a result, its power costs are comparatively high—say, about 0.9d. per kWh for base load power generated with imported coal (Department of National Development, 1955). It could, therefore, be an early prospective user of nuclear power for base load requirements, especially because these requirements are rapidly rising towards the point at which they could justify the installation of a nuclear base load station of economic size. It seems that Tasmania, which could outrun its hydro resources within the next two or three decades, might also find itself similarly placed.

At the same time, however, it should be recognised that nuclear power is by no means the only alternative power source offering. For South Australia, importation of conventional fuel either from Victoria or New South Wales on more advantageous terms than at present may be possible, and inter-connection with the Victorian network, wind power, and other technical possibilities have apparently been considered.

If South Australia should decide within the next decade or two to use nuclear power as an alternative to generating with imported coal supplies, the main economic effects would clearly be more important than the effects of Mount Isa deciding to change over. Balance of payments aspects as mentioned above would need to be considered, and the amounts involved would be more significant from the national viewpoint. The impact on the New South Wales coal industry would also be somewhat greater, but not necessarily critical, and much the same considerations as mentioned for Mount Isa would apply regarding the fabrication of fuel elements and reprocessing— at least until nuclear power costs became so cheap that general implementation was decided upon.

General changeover

By far the most important possibility to consider is a general decision by Australian electricity supply authorities to change to nuclear power. It has been suggested that comparative cost considerations are unlikely to favour a general changeover in Australia, even for base-load power, until about 1975—whether or not some isolated or partial changes are in fact made before that date does not seem very relevant, except to the extent that they would provide operating experience and data to enable more accurate assessments of comparative costs under Australian conditions to be made.

Assuming, however, that nuclear power costs are to become competitive under Australian conditions about 1975, it seems that a number of other aspects of the problem would need to be considered before a general changeover decision could be made:

(i) It appears that Australia's resources of coal are more than sufficient to support electricity generation requirements well beyond the year 1975—in fact, well beyond the year 2000 on the basis of foreseeable requirements (Department of National Development, 1955). At the same time, although prospects are promising, on present evidence it seems that we must retain some reservations about the adequacy of local uranium resources to sustain future power requirements much beyond the limitations of our coal resources. (Australian A.E.C. 1957.)

(ii) In the national interest, it would seem advisable to consider the optimum usage of coal and uranium together. Australian uranium might prove to be too valuable (e.g., as an export) to use as a fuel for power generation whilst we had adequate coal supplies for this purpose. Alternatively, other uses for coal (e.g., as a source of industrial chemicals) might be developed which could make coal too valuable to burn for electricity generation whilst we had uranium which could serve the same purpose at something like comparable costs. Such considerations would be particularly relevant to the circumstances of Australia where power is almost exclusively a Government service supplied "at cost," rather than on a commercial profit-seeking basis. Further, the possible social implications of an extensive redundancy in the coal industry cannot be overlooked.

(iii) The actual base-load generation costs of electric power are not so important an element in the Australian cost structure that it would be imperative to take advantage of any comparatively small margin of cost advantage as soon as it became available. Distribution and other costs in the power industry in Australia are an important element of power costs. In N.S.W., for example, technically possible base-load generation costs using conventional fuels are apparently in the region of 0.6d. per kWh, the actual cost to commercial and industrial consumers ranges from about 2d. to 4d. per kWh.*

The place of power costs in the cost structure generally is also a relevant consideration. In the U.S.A. it has been estimated that in 1952 total energy cost in all manufacturing industries only amounted to 4.4 per cent. of the total value added (U.S. National Planning Association), and it seems likely that a relationship of this order probably holds good for Australian conditions, too. Distribution costs in Australia could be comparatively greater than in U.S.A., but allowance for comparative degrees of mechanisation in the two countries might suggest higher energy usage in U.S. manufacturing processes.

In these terms it is suggested that, taking an overall view of costs, the national priority which should be accorded to taking advantage of marginal reductions in technically possible base-load generation costs may not be as great as popular opinion might suggest.

(iv) On the score of priorities in the use of capital resources, it may also be that the claim for the conversion of the electric power industry to nuclear power may not be very high. At present rates of expansion, and in terms of current money values, it appears that by 1975 Australia's capital requirements for new and replacement power generation and distribution equipment might be running at the rate of about £375 million per year—† assuming a continued rate of expansion of capacity of 8 per cent. per annum.

On the other hand, it is doubtful whether a rate of increase in the gross national product of more than about 4 per cent. per annum in real terms could be reasonably expected. This would mean that approximately 3.3 per cent. of our gross national product in 1975 might be required for capital expenditure on new and replacement power generation and distribution plant, compared with a figure of about 1.5 per cent. in 1955/56. In themselves these percentages are pretty small, but in relation to national priorities they could be very significant.

If, as seems likely, capital availability in 1975 is still as important a factor as it is today, it may be difficult enough to meet demands of this order in terms of conventional capital costs, much less undertake a power program in which nuclear plant made additional demands. These demands would be either by way of higher direct capital costs or capital costs for service installations for fuel processing, over and above possible reductions in capital requirements for coal mining and transportation. In absolute amount such additional capital costs might be relatively small (say, for example, £50 million); whether they would be critical must remain a matter for conjecture.

The effect of capital investment programs of these dimensions on Australia's balance of payments position can only be guessed at. However, at least in the initial stages of the change-over, it seems reasonable to expect that the foreign exchange cost component for technical know-how and specialised plant would be considerable.

Assuming that a general changeover to nuclear power were decided upon, there would still remain the question as to whether Australia should provide its own fuel processing and re-

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* Sydney County Council average rates for 1956.

† Estimate based on statistics for 1955-56, published by Electricity Supply Association of Australia.

processing installations or rely on overseas sources for such services.

On purely economic considerations it does not necessarily follow that, because Australia has uranium ore, it should aim at a completely self-sufficient industrial set-up taking in the mining of uranium, processing and fabrication into fuel elements, use in power generation, and the recovery of radioactive elements from reactor residues.

At least in the initial stages of a general change-over to uranium power, considerations involving the economic size of fuel processing installations would probably not favour their construction locally. Furthermore, without some degree of uniformity in the types of reactor system used by the various power generation authorities (to ensure reasonable through-put in relation to capital cost) it might be many years before such installations could be justified economically.

The capital cost of a fuel processing plant to service, on an economic basis, ten 300 MW natural uranium stations would apparently be the equivalent of about £A20 million (Goldring, 1957; Jukes, 1957), and costs of this order might be regarded as a relatively small price to pay for the benefits of a fully integrated uranium industry. However, comparative fuel processing costs in Australia and overseas would need to be considered. Furthermore, the possible security aspects of national reliance on overseas sources of supply for such a vital service as power generation should not be overlooked, although local storage of reserve fuel stocks as we have done with petroleum might be expected to cover most eventualities.

Conclusion

This analysis but skims the surface of a problem on which there is considerable scope for further work to be done. The tentative conclusion which arises, however, is that, although the technical possibilities more than justify present Australian research and training programs in this field, on economic considerations alone, the present case for the early adoption of nuclear power in Australia is not particularly strong. In some localities, special circumstances may favour its adoption in a relatively small way within the next decade or two, but general adoption of nuclear power in Australia before it reaches comparative cost status with "coal" power, would not seem warranted. Even then the national economic advantage may lie in holding off further, particularly if the comparative cost advantage over conventional generation processes were only marginal. However, it would be unrealistic not to recognise that scientific advancements may eventually override many of the current reservations, and that non-economic considerations arising from such aspects as national security, national pride, and the practical limitations of international industrial specialisation could outweigh purely economic considerations.

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Possible Developments in Nuclear Fuel Cycles in Australia

By J. P. Baxter and C. N. Watson-Munro[*]

The need for nationally integrated nuclear fuel cycles is stressed and, based on a number of speculative assumptions, some calculations are made of the order of requirements of fissile and fertile material.

Introduction

A wide range of reactor systems are under investigation in the world today; in terms of the kinds of nuclear fuel they use, these reactor systems may be divided into two broad classes:

(a) Those using natural, or near natural, uranium and producing plutonium with a conversion factor of order of 0.75—0.8. Some power reactors of this type are available now.

(b) Those reactors using highly enriched fuel [U233, U235, Pu239] and breeding from fertile materials [thorium or U238] new fuel with possible conversion factors ranging from close to unity to about 1.5. Feasibility studies have shown that some of these systems could be economically attractive if the research problems can be solved. It is unlikely that such reactors would be available to produce power commercially in Australia before 1970.

Reactors of type (a) in their present state of development are not economically competitive with coal power in the main industrial areas of Australia, located near substantial coal fields. Reactors of type (b) may well be economically competitive, after their technical and economic possibilities have been evaluated by experiment.

It therefore seems likely that in the last quarter of this century Australia may install a substantial amount of type (b) nuclear power capacity. This will require a considerable capital investment in fuel and fertile material, together with extraction, fabrication and chemical processing plant. Unless there is a nationally integrated fuel plan, costly confusion could result.

Possible methods of providing fuel are shown in Figure 1. A high capital cost isotopes separation plant consuming electricity does not seem to be particularly applicable to Australia; nor does the purchase of fissile material from abroad in the quantities likely to be required look feasible.

It seems that the best way for Australia to meet her capital requirements from 1970 onwards of fuel for type (b) nuclear reactors will be to install adequate natural uranium capacity of type (a).

It is obviously necessary to provide a national balance between the production and use of fissile fuel; it is equally necessary to ensure that the best use be made of Australia's raw materials.

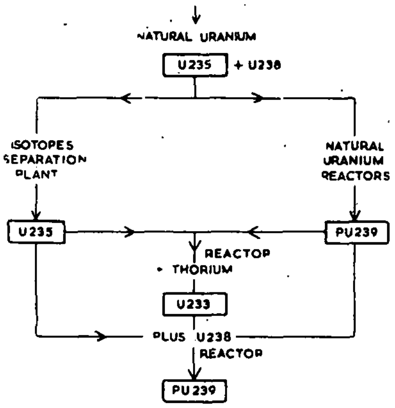

FIGURE 1.

Requirements of electric power for Australia

To study the possible kinds of fuel cycles that might be applicable to Australia, it is necessary to make some assumptions of the likely developments of electric power, and the fraction of these that may be supplied from nuclear fission sources. There are a number of possible approaches to these estimates. They are as follows:—

(i) In a private communication (dated 26/6/57) the Commonwealth Department of National Development estimates the growth by assuming an increase of 9% for the next decade, 8% for the following decade, then 7%, 6% and finally 5% for the decades thereafter. The installed capacities calculated from these assumptions are shown as Curve I of Figure 2.

(ii) In a paper presented at the first Geneva Conference, the U.N. Department of Economic and Social Affairs (1955) analysed the world energy demands on the basis of data supplied by member states. It is estimated that there will be an increase of stationary power (92% electricity) by a factor of 5 between 1952 and 1975 and a further 3.8 in the following 25 years. Applied to Australia, these estimates are shown as Curve II of Figure 2.

(iii) The Commonwealth Department of National Development (1955) gives some estimates for N.S.W. based on the current

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[*] Australian Atomic Energy Commission. Manuscript received March 4, 1958.

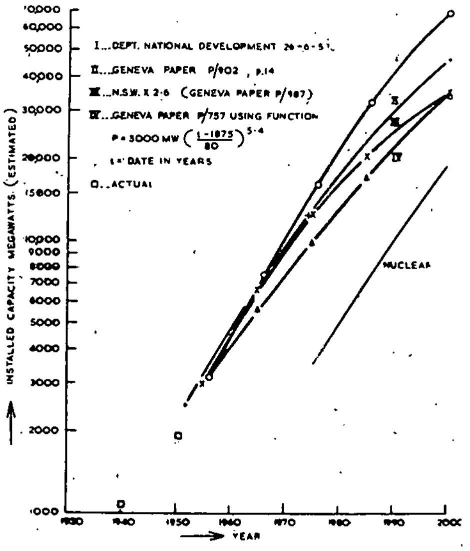

FIGURE 2.

annual increase of 9%, falling to about 2½% by the end of the century. If we assume a constant ratio of 2.6 to 1 for Australian power requirements relative to those of N.S.W., we have the results shown as Curve III.

(iv) Robinson and Daniel (1955) and Daniel (1955) adopt an historical and statistical approach. Production of electricity over the years 1900 to 1950 has been analysed, and found to fit in closely with a curve:—

$$E = k \left[\frac{(t - 1875)}{77} \right]^{y}$$

where k = demand in 1952.

In the case of world demand the value of exponent y was found to be 4.3 (Robinson and Daniel, 1955) and in U.K. demand 4.92 (Daniel, 1955).

The Australian Department of National Development (1955) quotes a figure of 4091 x 10^6 kwh for 1938/39 and 12397 x 10^6 kwh for 1953/54.

These figures give an exponent of 5.4.

Curve IV shows the function:

$$E \text{ (Australia)} = 3000 \text{ MW} \left[\frac{t - 1875}{80} \right]^{5.4}$$

plotted with respect to base year 1955.

In Figure 3 the exponent 5.4 is examined in the light of historical evidence kindly supplied by the Department of National Development; the fit justifies surprisingly well the use of the exponent 5.4.

It will be noted that both III and IV predict by the year 2,000 a total installed capacity of 35,000 MW; II and I predict 47,000 and 70,000 MW respectively. We shall be conservative and

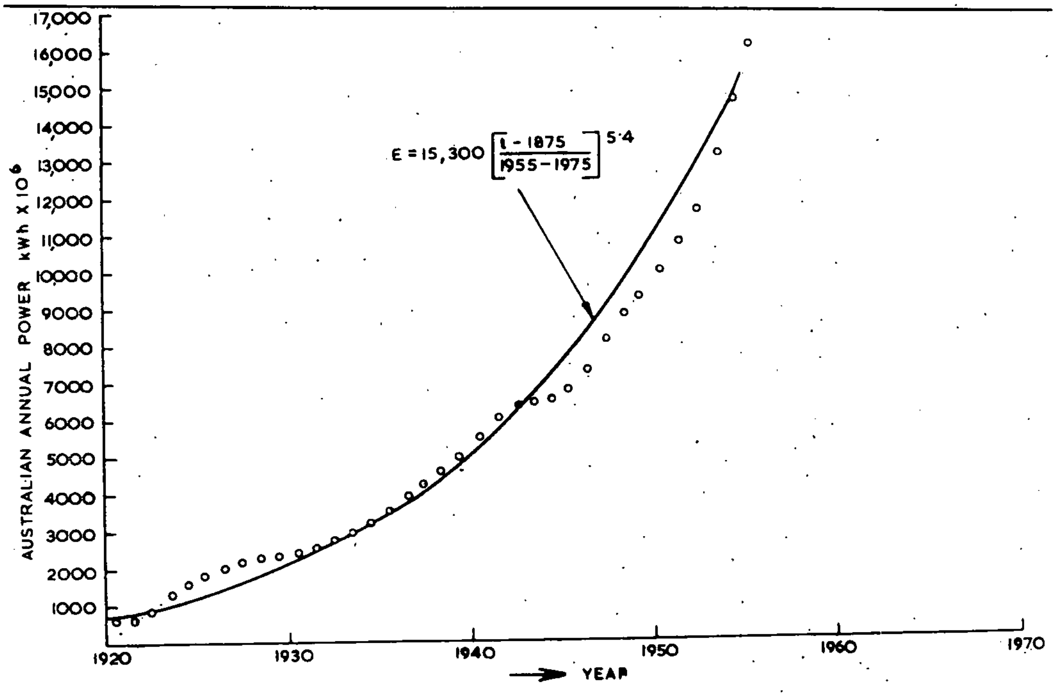

FIGURE 3.

select the lowest figure of 35,000 MW, and assume that 20,000 MW of this is supplied from nuclear sources. This will provide for some conventional coal and hydro capacity to be operated, mainly on peak loads.

If we assume a doubling time of 10 years for the nuclear capacity we have the estimates given in Table 1.

We have assumed for the purposes of these calculations that there is no significant contribution from thermonuclear power.

Breeder reactors

Let us define the conversion factor C as the number of new fissionable nuclei produced for each nucleus consumed (in both fission and non fission reactions) i.e.

$$C = \eta - 1 - L$$

where L = neutrons lost by leakage and parisitic capture

and let G = breeding gain = c — 1

and c — 1 = η —2 — L

If p = specific power reactor in watts per kilogram, and if we assume that each kilogram of material fissioned per day produces heat energy at the rate of 10^9 watts, then—

kilograms fissioned/day/kgm = p x 10^{-9}

kilograms consumed per day per kilogram = p (1 + a) x 10^{-9}, where a = $\sigma c/\delta f$

kilograms bred per day = p (1 + a) G x 10^{-9} and, if we assume a load factor of 80%, the amount bred per year per kilogram

$$= .80 \times 365\ p\ (1 + a)\ G \times 10^{-9}$$
$$= 292\ p\ (1 + a)\ G \times 10^{-9}$$

TABLE 1.

Predicted Electrical Requirements in Australia:

III: Requirement according to Curve III of Fig. 2.
IV: Requirement according to Curve IV of Fig. 2.

Year	Total Electric MW		Total Nuclear MW	% Nuclear	
	III	IV		III	IV
1975	15,000	10,000	3,500 MW	23%	35%
1980	17,000	13,500	5,000	29%	37%
1985	21,000	17,500	7,000	33%	40%
1990	26,000	23,000	10,000	39%	43%
1995	31,000	29,000	14,000	45%	48%
2000	35,000	35,000	20,000	57%	57%

To illustrate we select three possible breeder reactor systems of the assumed characteristics listed in Table 2.

Table 3 presents the calculations of fissile material required; a five-year installation cycle is assumed, and no allowance is made for processing losses.

This would imply that Case A would require 10,500 kilograms to commence and would finish up with a credit at the turn of the century of 69,700 kilograms. Case B would require an initial investment of 3,500 kilograms and would finish up at the turn of the century with a credit of 12,100 kilograms. Case C would require an initial investment of 3,500 kilograms and would demand for its operation 46,100 kilograms over the period.

TABLE 2.

Characteristics of Possible Reactor Systems

Type	Case A	Case B	Case C
	Fast	Thermal	Thermal
C	1.4	1.1	0.9
G	+0.4	+0.1	—0.1
Rating heat "p"	1 MW(H)/kgm	3 MW(H)/kgm	3 MW(H)/kgm
Rating Electrical	⅓ MW(E)/kgm	1 MW(E)/kgm	1 MW(E)/kgm
Fuel Cycle	Pu-U238	U233-Th	U233-Th
a	0.15	0.098	0.098
Capital Investment Fuel	3 kgm/MW(E)	1 kgm/MW(E)	1 kgm/MW(E)
Load Factor	80%	80%	80%
Fuel Bred per year:			
per kgm.	+0.134 kgm	+0.0952 kgm	—0.0962 kgm
per MW(E)	0.402 kgm	+0.0962 kgm	—0.0962 kgm

In the calculations a considerable simplification has been made in order to illustrate the orders of magnitude of fissile materials required. The changes in conversion factor which will result from the accumulation of fission products, the accumulation of heavy element poisons, and the production of high atomic weight fertile and fissionable materials have been neglected. These will cause considerable changes in a and in conversion factor, but would have to be specifically calculated for any reactor system. The conversion factor will also be affected by the burn-up obtained before chemical processing, whether the fuel is batch or continuously irradiated, the degree of flux flattening in the reactor, and the reactor flux.

Some cases for natural uranium fuel heavy water reactors have been calculated by Lewis (1957), and for slightly enriched sodium graphite reactors by Benedict and Pigford (1957).

For a U233-Th cycle thermal reactor the expression for neutron balance will be:—

Neutrons produced from U233 fission = $\eta_{23}\, f p\, N_{23}\, \sigma_{23}\, \phi$

Neutrons produced from U235 fission = $+\eta_{25}\, f p\, N_{25}\, \sigma_{25}\, \phi$

≡ Neutrons absorbed

By U233 and U235 = $N_{23}\, \sigma_{23}\, \phi + N_{25}\, \sigma_{25}\, \phi$

By Th 232 = $+ N_{02}\, \sigma_{02}\, \phi$

By U234, U236, etc. = $+\Sigma\ (N_h\, \sigma_h\, \phi)$

By fission products = $+\Sigma\ [N_F\, \sigma_F\, \phi]$

By control rods = $+ N_C\, \sigma_e\, \phi$

By structural materials, coolants, etc. = $+N_S\, \sigma_S\, \phi$

Thermal leakage = $+ D B^2 \phi$

TABLE 3: FISSILE MATERIAL REQUIREMENTS

Year	Assumed Nuclear Breeder Capacity	Fuel Balance	kg fissile material		
			Case A	Case B	Case C
1975	3500 MW(E) Initial Installation	Initial Fuel Investment.	—10,500	— 3,500	— 3,500
		Fuel Bred	+ 7,000	+ 1,700	— 1,700
		Cumulative Credit	+ 7,000	+ 1,700	— 1,700
1980	5000 MW(E) (+1,500 MW(E) Additional)	New Fuel	— 4,500	— 1,500	— 1,500
		Fuel Bred	+10,000	+ 2,400	— 2,400
		Cycle Credit	+ 5,500	+ 900	— 3,900
		Cumulative Credit	12,500	+ 2,600	— 5,600
1985	7000 MW(E) (+2,000 MW(E) Additional)	New Fuel	— 6,000	— 2,000	— 2,000
		Fuel Bred	+14,000	+ 3,400	— 3,400
		Cycle Credit	+ 8,000	+ 1,400	— 5,400
		Cumulative Credit	20,500	+ 4,000	—11,000
1990	10,000 MW(E) (+3,000 MW(E) Additional)	New Fuel	— 9,000	— 3,000	— 3,000
		Fuel Bred	+20,000	+ 4,800	— 4,800
		Cycle Credit	+11,000	+ 1,800	— 7,800
		Cumulative Credit	+31,500	+ 5,800	—18,800
1995	14,000 MW(E) (+4,000 MW(E) Additional)	New Fuel	—12,000	— 4,000	— 4,000
		Fuel Bred	+28,000	+ 6,700	— 6,700
		Cycle Credit	+16,000	+ 2,700	—11,700
		Cumulative Credit	+47,500	+ 8,500	—30,500
2000	20,000 MW(E) (+6,000 MW(E) Additional)	New Fuel	—18,000	— 6,000	— 6,000
		Fuel Bred	+40,000	+ 9,600	— 9,600
		Cycle Credit	+22,000	+ 3,600	—15,600
		Cumulative Credit	69,700	12,100	—46,100

where the subscripts 02, 23, 25, h, F, C and S refer to Th232, U233, U235, non fissile heavy elements, fission products, control rods (needed if reactivity is changing) and structural materials respectively. The flux ϕ is assumed to be constant. N refers to the number of atoms present per unit volume and σ to the absorption cross section.

During the life of the reactor system the reactor parameters f, p, DB^2, the absorption by fertile materials and, after running for a few days, the absorption by the principal poisons Xe135 and Sm149 will not vary greatly, but the other factors will.

Walker (1956) tables some figures on the effective cross section of fission products with irradiation time. However, while fission products and non-uranium heavy element products can be removed by chemical processing, there will be progressive production of higher uranium isotopes.

Table 4 is quoted from Benedict and Pigford (1957) for a sodium reactor containing initially 1.9% of U233 in thorium.

However, in this paper the speculative figures of nuclear power needs, fuel power ratings, and conversion factors are such that the variation of conversion factors during the fuel cycle is relatively unimportant.

Provision of capital investment of fissile material

If we assume 1975 as the date when breeder systems are installed in Australia, and inspect Table 3 it will be seen that between 3.5 and 10.5 tonnes of fissionable material will be required for capital investment. It is likely that a combination of Cases, A, B, and C will be used, and it might therefore be reasonable to assume that about five tonnes of fissile material will be required. This may be provided by:—

(a) purchase from abroad

(b) manufacture in an isotopes separation plant from natural uranium

(c) manufacture in natural uranium reactors.

As discussed in Section 1 of this paper, the latter course seems the best for Australia to follow.

It is unlikely that any natural uranium reactors will be in operation in Australia before 1965. At 25% thermal efficiency, 80% load factor, and 0.75 conversion factor each natural uranium reactor will manufacture per annum per MW(E):—

4 x 0.75 x 365 x 0.8 gm of fissile material i.e. 0.876 kgm per MW (E) per annum.

To produce five tonnes, 5700 MW (E) years will be necessary. This could be met with approximately 1,000 MW(E) of natural uranium capacity installed between 1965 and 1975. About 2,500 tonnes of uranium would be required.

This material produced will be plutonium, and as such quite suitable for use in the fast breeder type of reactor discussed in Case A. The thermal

TABLE 4.

Atomic Ratios of Heavy Elements (sodium reactor):

Isotope	σ_a	α	Initial	20,000 MWD/ton Th.	Equilibrium
Th232	4.1	—	209.1	209.1	209
U233	342	0.098	4.0	4.922	4.8
U234	208	—		0.722	0.35
U235	378	0.184		0.397	0.69
U236	6.6	—		3.537	3.45

σ_a and α are for 400°C, initial conversion factor, 1.06.

breeders discussed in Cases B and C will give higher conversion factors if operated on the U233-Th cycle. The U233 required may be produced—

(a) in natural uranium reactors by providing some excess k, either in the original design or by recycling of plutonium. A small quantity can also be produced from the excess k during the early part of the natural uranium cycle of conversion of U235 to plutonium.

(b) by starting off (with lower conversion factor) the breeder reactors with plutonium or uranium 235 from diffusion plant.

Fertile material needs

The fertile material requirements would be greatly dependent on the type of reactor, and the atomic ratio of fertile material to fuel. This can vary with different forms of reactor from about 30 to 1 to 200 to 1.

The amount of fertile material burnt will be negligible in comparison with the capital requirements.

If we return to Table 3 and assume an atomic ratio of 1:100, then Case A will require 6,000 tons of uranium 238, a good fraction of which could be met from the depleted uranium resulting from previous natural uranium reactor operations. Cases B and C will require 2,000 tons of thorium.

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The Canadian Nuclear Power Program

By D. G. Hurst*

Canadian industry has developed in regions where cheap hydroelectric power is available. However, within a few years all the hydrcelectric potential of certain areas, notably Southern Ontario, will be utilised. When that happens, increased power requirements will have to be supplied by thermal generating stations. Fossil fuel for these will probably be imported but a very attractive alternative is to use indigenous uranium in nuclear plants provided that the power can be produced cheaply enough (5 to 6 mills kWh). A program now under way to develop a suitable nuclear reactor using a heavy water moderator and natural uranium fuel gives promise of meeting the cost target.

A superficial look at Canada's potential and developed energy resources (Table 1) suggests that she is one of the nations least in need of nuclear power. The error in this conclusion becomes apparent only through detailed studies of the separate geographical regions.

The most highly industrialised region—the Great Lakes and St. Lawrence watershed in the provinces of Ontario, and Quebec—is almost devoid of fossil fuels. Its industries were attracted and encouraged by cheap hydroelectric power and good transportation. The Maritime provinces on the east coast have some water power and considerable coal, but mines are beset with difficulties, arising in part from submarine location or other characteristics of the seams, and, in part, from competition with sea-borne foreign fuel. The central region, the prairies, has abundant coal and oil and some water power, but has not yet developed industrially to match its resources, the discovery of oil being very recent. British Columbia, the west coast province, has large reserves of coal and water power, but the mountainous nature of the countryside hinders transportation of the coal, and the need for international cooperation and the possible destruction of important salmon fisheries retard the further utilisation of water power in the more settled sections. The vast Arctic and sub-Arctic regions are unsuitable for normal settlement, but are rich in minerals.

It is possible to set aside from immediate consideration the textbook atomic plants which are to be located in remote areas to provide small amounts of electric power. The intermittent character of the load, the cost of maintaining the skilled staff, the uncertain degree of reliability, and the high capital cost per kilowatt make such plants still uneconomic in Canada.

The same factors apply with less force to intermediate plants (20 to 50 MW electrical) in a power system of moderate size, e.g., in the Maritimes. The estimated cost of power is on the economic borderline, and serious consideration has been given to these. However, there is no real power shortage, and until the cost has been shown to be competitive and the continuity of operation proved to be satisfactory, a plant like this is unlikely to attract serious backing.

(It should be pointed out that the energy resources are under provincial jurisdiction, and that atomic energy development has been largely a federal matter. Both publicly and privately owned utilities are engaged in the supply of electric power. It is federal policy to support research and development, but not to enter the field of commercial nuclear power production.)

Some important industries, e.g., paper making and ore refining, use large quantities of heat in their processes. This offers a field for reactors without the complication of the electricity generating plant. Several companies have been looking into this possibility, without a definite commitment as yet.

It would seem from the above that a pressing need for nuclear power does not exist generally throughout the country. For this we must turn to the power-wealthy province of Ontario. The Hydro-Electric Power Commission generates and distributes most of the province's electric power through one of the world's major electrical grid systems. The demand has been increasing steadily with an average annual increase of 6.56% from 1922 to 1955 when, to satisfy the demand of 4.1 million kW, there were available 4.5 million kW made up of 3.2 hydroelectric, 0.64 fuel electric and 0.68 purchased from outside the province. The total potential power from water is about 5.4 million kW. By 1960, the St. Lawrence Seaway Development will be complete, and essentially all the water resources within an economic distance of the main customer demand will be developed, and full use made of devices such as pumped storage. After 1960, increased demand can be supplied by thermal plants only, and it is at this stage that nuclear power will become of prime importance.

Because of its size, the Ontario system satisfies two criteria for early successful use of nuclear power. It requires power increments to be large, i.e., several hundred megawatts, and it is not seriously affected if a plant of this capacity drops off the line. However, power has been cheap, and even the advent of thermal stations will not greatly affect the price, because coal can be obtained for $8 to $9 a ton. Nuclear power will therefore have to compete economically in a cheap-power area. An otherwise nearly even balance might be tipped in favour of nuclear power by the existence in Ontario of very large uranium deposits, and the need to import coal from the U.S.A. (It is unlikely that

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fuel from other parts of Canada will undersell the imported coal.) If Ontario's increased requirements for power are supplied exclusively from coal, this item in the annual American-Canadian trade balance amounts to an estimated $270 million by 1980.

Thus the most promising role for the first nuclear plants in Canada is to supply power in blocks of 200 to 500 MW into the Ontario grid system, starting about 1965. To do this, the stations must produce power at a cost less than 0.6 cents per kWh, a target cost being 0.5 cents (5 mills) per kWh. This cost will be based on public utility financing with low interest and tax rates. Current planning does not envisage nuclear power replacing the coal completely, but supplying perhaps one-half to three-quarters as much power as coal.

The Canadian program began to take practical form about 1950, when it was demonstrated with the original NRX reactor fuel charge that uranium metal can produce at least 3,000 MW days/tonne without mechanical failure, and when water-cooled loops were successfully operated in NRX at temperatures and pressures of interest in steam plants. As the promise of economic power grew, the power-producing and equipment-manufacturing industries were brought into the picture. Engineers were sent from several companies to form a Nuclear Power Group at the Chalk River Laboratories, where they could work closely with Atomic Energy of Canada Ltd. staff in a feasibility study. After a detailed analysis, they proposed a demonstration reactor with 20 MW electrical output. The conceptual design of this reactor, NPD-1, is a natural uranium oxide fuelled, heavy water moderated, pressurised reactor. It was not expected to be economical for Ontario, but to be of importance for testing the interaction of the reactor with the turbine and electrical grid, for developing components, and as a basis for cost estimation on a scale which could be extrapolated to full-size plants.

A joint project to construct and operate this reactor was undertaken by Atomic Energy of Canada Limited, the Hydro-Electric Commission of Ontario, and the Canadian General Electric Company.

The Nuclear Power Group then turned their attention to a full-size (200 MW electrical) power plant. The pressure vessel for NPD-1 was taxing the technology of fabrication almost to the limit, and the increase in size of a 200 MW reactor would be difficult, if not impossible. It was decided to use pressure tubes to contain the heavywater coolant, the moderator to be at atmospheric pressure. This removes a limit on reactor size and has led to a very promising design.

NPD-1 was no longer a prototype. It might still have been a possible reactor for small systems, except that as design and construction progressed the estimated costs rose to an uneconomic level. For these reasons design and construction of NPD-1 were halted, and a new design with pressure tubes was worked out. This is now being actively developed as NPD-II. It will use much of the equipment planned for NPD-1, and with small changes the excavation on site will be suitable. There has been some delay and no lowering of cost due to the shift, but the gain from bringing the demonstration reactor into line is worthwhile. Although for mechanical reasons pressure tubes are preferable to a pressure vessel, they could not be considered when NPD-1 was being planned. At that time pressure tubes would have been thought of as stainless steel, and their neutron absorption, by destroying reactivity otherwise available to prolong irradiation of fuel, would have raised fuel costs to a level incompatible with the requirements.

TABLE 1—CANADA'S ENERGY RESOURCES
(Figures as of 1955)

ESTIMATED RESERVES:	
Coal (readily mineable)	30,000 million tons.
Oil	11,000 million tons.*
Natural gas	11,000 million tons.*
Hydroelectric	37 million kW total potential.
Annual consumption 100 million tons.*	
SUPPLY OF ELECTRICITY:	
Installed hydroelectric capacity	13 million kW.
Installed thermal capacity	2 million kW.
Comparative average selling prices:	
Norway	0.4 cents per kWh.
Canada	0.8 cents per kWh.
Switzerland	1.4 cents per kWh.
U.K.	1.5 cents per kWh.
U.S.A.	1.5 cents per kWh.
Per capita production:	
Norway	6500 kWh.
Canada	5200 kWh.
U.S.A.	3800 kWh.

* These are tons of coal equivalent.

The reserves do not include the Athabaskan tar sands (66,000 million tons) or lignite and sub-bituminous coal.

When the large reactor study was begun, technical advances gave two different pressure tube designs having economic promise. The development of zirconium technology had reached a stage where production of suitable zircalloy pressure tubing could be expected with confidence (a hope which has since been fulfilled, notably by the pressure tube in the NRX central thimble). Also, the concept of insulating the pressure tubes from the hot coolant, e.g., by wrappings of thin foil of aluminium-nickel alloy, permitted the designers to consider aluminium or magnesium pressure tubes.

Power can be produced for 5 mills/kWh only with great care in design to ensure long irradiation and high load factor. Infrequent shutdowns will keep the load factor high, but if long intervals occur between fuel changes, there will be flux distortions and reactivity changes which waste neutrons and result in reduced fuel burnup. Consequently, the large reactor is to be refuelled in an almost continuous manner. The fuelling machine will be capable of opening a pressure tube and discharging or charging fuel slugs without letting down the pressure. It is expected that 7,000 to 10,000 MW days will be obtained from each tonne of natural uranium without reprocessing. NPD-II will likewise be fuelled at power to give operational experience with these machines. The plan is to move the slugs without letting down the pressure. It is from one end to the other, but the direction of motion will alternate from tube to tube. This bi-directional fuelling will give all regions along the reactor similar average properties.

TABLE 2—NPD-II

This reactor is being built at the Des Joachims hydroelectric generating station and will feed into the Ontario grid. The estimated cost is $30 million and the completion date 1961.

PRELIMINARY ENGINEERING DATA

Power:	Total 80 MW Electrical 20 MW.
Core construction:	132 horizontal pressure tubes of Zircalloy II, 3.25 inches I.D., passing through a calandria which contains the heavy water moderator.
Calandria:	17ft. diameter, 15ft. long.
Coolant:	Heavy water at 1,030 psi, temperature in 485°F., out 530°F.
Steam:	Generated from ordinary water at 400 psig (saturated).
Fuel:	UO_2; 0.95 inch diameter, sheathed in Zircalloy II and assembled into 7 element bundles. Max. surface heat rate 225,000 BTU/ft.^{2}/hr.
Fuelling:	Bi-directional, at power.
Flux:	Average across hottest rod 5.4 x 10^{13} n/sq. cm./sec.

TABLE 3—LARGE POWER REACTOR

A new division of Atomic Energy of Canada Limited has been set up in Toronto, closely associated with the engineering branches of the Hydro-Electric Power Commission of Ontario, to complete the design and development of a reactor having an electrical output of 200 MW. The following figures refer to the outline specifications and conceptual design, which are the starting point for the next phase of the work. The many reactor calculations, designs, and cost estimates which were done in arriving at this approximately optimised design required about 14 man-years.

DESIGN DATA

Power:	Total 794 MW. Net Electrical 200 MW.
Core construction:	Similar type to NPD-II, 252 pressure tubes.
Calandria:	20ft. diameter, 18ft. long.
Coolant:	Heavy water at 1,000 psi, temperature in 430°F., out 525°F.
Steam Conditions:	419°F., 290 psi.
Fuel:	UO_2, expected burnup 8,100 MWd/tonne of uranium.
Fuelling:	Bi-directional under power.
Flux:	Average across hottest rod 1.7 x 10^{14} n/sq. cm./sec.
Cost:	$60,000,000, not including development costs.

The completion of the 20 MW NPD reactor and the design and development of the 200 MW reactor will be the main part of the nuclear power program for the next four or five years. Both these reactors are natural uranium, heavy water cooled and moderated, pressure tube reactors. Some details are given in Tables 2 and 3.

The past importance of heavy water and natural uranium in the Canadian reactor program has given Canada a prominent place in their technology, but it is not the sole reason underlying their choice. Heavy water is to be used on the basis of neutron economy despite the high price. It is not possible in a paper like this to do justice to the details of the cost analysis. The fundamental methods will be found in the references. (Duret et al., 1957; Lewis, 1954, 1956; Canadian Study, 1958.)

As an example, the estimated carrying charge for heavy water coolant in the large reactor is 0.15 mill/kWh and a similar cost would be added by a loss of one pound/hr. Neither of these occurs with light water, but its neutron absorption would add much more to the fuel cost than 0.3 mill/kWh. Similar considerations rule out graphite as a moderator.

A case might be made for slight enrichment because it increases the attainable burnup and therefore, in some circumstances, reduces the overall energy cost. The reduction is slight for a large increase in irradiation (e.g., 0.2 mills/kWh with irradiation increased from 7,000

MWd/tonne to 14,000 MWd/tonne. A loss in burnup or operating time due to failure during the extra irradiation when the fuel is more fragile would soon nullify the advantage.

Although heavy water is undisputed as moderator for the presently planned power reactors, gases and organic fluids are possible alternatives to heavy water as coolants. Several studies are under way to assess their value.

Fuel changing during operation, which seems to be necessary with uranium fuel, is associated with complicated expensive machinery and involves a risk that minor malfunctioning will shut down the reactor. Reactors requiring no interference with a fuel charge during its life would offer advantages in simplified fuelling equipment, and increased reliability of operation. They might also serve more remote districts, refuelling being done at long intervals by a team of specialists brought in for the occasion. Some proposals along this line have been made recently (Lewis, 1957), using the fact that over at least the integrated flux range from four to 10 neutrons/kilobarn, initially pure thorium reaches the condition in which neutron yield is very nearly equal to neutron absorption. To maintain criticality in a finite reactor would require that the leakage neutrons be supplied by enrichment. It is shown in the reference that by appropriate arrangement and self shielding, the reactivity of a mixture of thorium and uranium 233 or uranium 235 can be kept nearly constant up to 1.5 fissions per initial fissile atom nine years at a flux of about 10^{13} n/sq. cm./sec.). This leads to a fuel cost less than 2 mills/kWh in a reactor sealed up for about 10 years. Such considerations will not interfere with the present plans, but are preliminaries for the future program.

However, the immediate problem is to develop a reactor type which can be successful in the Ontario system. Once developed, this would be suitable for other regions. The developmental work may also apply to medium power reactors of assistance in smaller systems throughout the country, or even to reactors which will compete with other thermal plants in any region.

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Liquid Metal Fuel Reactors

By K. F. Alder*

The advantages and disadvantages of a liquid metal fuel reactor system are outlined in relation to other possible power reactors. Liquid metals which are possible fuel carriers and coolants are discussed, and in particular a comparison is drawn between a solution-type fuel using bismuth and a dispersion-type using sodium. The reasons for the choice of a sodium dispersion fuel for the A.A.E.C. research project are presented and the problems posed by such a reactor system are summarised.

Introduction

Present trends of development in nuclear power reactors are towards higher operating temperatures to improve thermal efficiency, and towards more irradiation-resistant fuels to achieve increased reactor reliability and greater fuel utilisation.

"High temperature" operation requires definition. Considering existing large-scale power reactors, it may be said that the upper temperature limits are about 300°C for pressurised water units and 450°C for gas-cooled systems. Temperatures of 500°C and up can be regarded as high in nuclear reactors.

Operation at temperatures higher than 500°C should be possible with two types of coolant—gases and liquid metals. The Australian Atomic Energy Commission's research program has as its basis the study of such high temperature systems applied to thermal reactors. Advanced types of gas-cooled reactors are discussed by Dalton (1958); this paper considers the preferred alternatives among the possible liquid metal systems.

Liquid metal coolants

The necessity for liquid coolants to operate at high temperatures has arisen in many instances before the advent of nuclear power. Classical examples are the General Electric Company's mercury boiler for increased efficiency of power generation, dating from 1922 (Hackett, 1942), and the use of liquid sodium to cool aircraft exhaust valves since 1928 (Heron, 1928).

Ideally, a liquid coolant for a thermal reactor should have low capture cross-section for thermal neutrons, a low melting point, high boiling point, and a low vapour pressure in the operating temperature range. It must be compatible with constructional and container materials which it contacts in the reactor core and external circuit, so that negligible corrosion or mass transfer of these materials will occur. It is desirable for the coolant to have good radiation stability and a low level of induced radioactivity after neutron irradiation to avoid the necessity for heavy shielding of circuit components external to the reactor core, and to minimise health hazards should leaks develop. Axiomatically, the coolant must have good heat transfer characteristics, i.e., high specific heat and thermal conductivity, coupled with low viscosity.

A number of liquids are available for high temperature operation, and include organic compounds, fused inorganic salts and liquid metals. Organic liquids such as "Dowtherm" (diphenyl-diphenyl oxide eutectic) have acceptable heat transfer properties, but only a small useful temperature range and indifferent radiation stability. Fused salts introduce problems of high neutron capture irradiation instability, and corrosion of container material.

The heat transfer coefficients of liquid metals are high, thus allowing high heat ratings per unit of reactor core volume or, alternatively, per unit of fissile material invested. Liquid metal systems need not be pressurised, giving considerable advantages in engineering design. Liquid metals are stable under heat and irradiation to the extent that, being elements or mixtures thereof, they do not decompose, polymerise or otherwise suffer in a reactor environment.

All liquid metals present some problems of compatibility and containment when considered for reactor service, and this factor, plus the effect of the coolant on the neutron economy of the reactor, constitute two of the main criteria in deciding the metal to be used.

Unfortunately, there is no one liquid metal which combines all the desirable attributes for a reactor coolant. Consequently, some compromise must be reached in making a selection, and other aspects of the reactor design so affect this selection that a number of research projects on nuclear reactors utilising different liquid metal coolants are in progress in the Western world (Williams, 1954; Siegel, 1955; Hoffman, 1956; Abraham 1957).

Liquid metal-cooled and liquid metal-fuelled systems

Liquid metal coolants offer the opportunity of operating reactors with conventional type fuel elements at high temperatures and under conditions of high fuel rating. The additional step of mixing the fuel intimately with the coolant increases the possibilities of high ratings, and also removes the problem of the irradiation behaviour of fuel elements.

The desire to develop reactors having high reliability and good fuel utilisation has focused attention on irradiation damage in fuels since

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* Australian Atomic Energy Commission. Research Establishment. Manuscript received March 26, 1958.

the successful operation of the earliest reactors. Under reactor irradiation, unalloyed uranium may distort by surface wrinkling, growth and swelling, causing failure of the fuel element of which it is a constituent (Ball, 1955). Heat treatment and alloying have been developed to minimise such damage, and present trends in ceramic fuels indicate that even greater resistance to irradiation damage is forthcoming. However, all solid fuel reactors are liable to the disadvantages of fuel element failure, and any system which overcomes these problems has potential advantages in reliability and high fuel burn-up.

One method of forming a "high integrity" fuel is to disperse fissible material in a matrix which is known to be stable under irradiation. This allows high heat output per unit of fuel, because the matrix may be used to extend the heat transfer surface of each fuel particle. The fuel for the Australian reactor HIFAR is of this type, being an enriched uranium compound dispersed in aluminium. Power reactors of high fuel rating may be built, using such fuels and suitable coolants, e.g., the U.S. Navy submarine "Sea Wolf" uses such a fuel in stainless steel, cooled by sodium (AECL CRR-590). However, so much parasitically neutrol-absorbing material has been added to this type of fuel that a reactor using it is unlikely to be economical for civil purposes. An alternative is to disperse fuel in part of the reactor moderator. This achieves a high fuel rating by dispersing the heat source, but the irradiation stability of such dispersions in moderators suitable for high temperature operation (graphite, beryllium, beryllium oxide) has yet to be studied. The Australian high temperature gas-cooled reactor project involves studies of this type of fuel (Dalton, 1958).

In general, high temperature operation aggravates the problems of fuel element damage, and to utilise the advantages of liquid metal cooling it would be highly desirable to eliminate them altogether. A possible method is to use fuel intimately mixed with the coolant so that mechanical irradiation damage is either absent, as in a fuel solution, or of no consequence, as in a fuel slurry.

A variation of the latter approach is to replace a conventional fuel element by a "can" filled with liquid metal fuel solution or slurry. The first alternative, a canned liquid metal solution, is not attractive because of the very dilute nature of the available solution-type fuels. The "canned" slurry fuel element has been discussed widely, but has not been investigated seriously for thermal reactors because of unfavourable neutrol economy. However, should circulating fuel systems prove technologically not feasible, the concept of a canned slurry fuel, liquid metal-cooled, should receive further attention. It may be particularly suitable for a fast reactor.

Circulating combined fuel-coolant systems of several forms have been proposed and in some cases investigated. The aqueous homogeneous reactor is such a system, in which the fuel-coolant is also the moderator (AECD.3646, 1955). This reactor suffers from two disadvantages—the danger of a highly-pressurised and extremely radioactive circuit, and the corrosive nature of the liquid. Solutions of uranium salts in fused salt mixtures have been proposed also, but neutron capture in the salts and the compatibility of the liquid with constructional materials pose severe problems.

Circulating fuel systems have potential advantages in elimination of the charge-discharge problems of solid fuel reactors, and in the possibility of continuous de-poisoning and processing of the fuel. Such advantages would be lost in liquid metal systems employing canned fuel, with the exception that venting of the cans might allow continuous removal of xenon from the core.

The choice of liquid metal for a circulating fuel system is much more limited than for a liquid metal-cooled reactor, because an extra criterion—solubility of the fuel—must be imposed.

Possible liquid fuels

Four types of liquid metal fuels may be postulated. These are:—

(i) Low melting point alloys rich in uranium (or plutonium);

(ii) solutions of uranium or plutonium in acceptable coolants;

(iii) slurries or dispersions of fissile material (metal or compounds) in equilibrium with solutions, i.e., added in quantities exceeding the saturation solubility; and

(iv) slurries or dispersions of fissile material (metal or compounds) with negligible solubility in the coolant.

No low melting point alloys with high uranium or plutonium contents and reasonably low thermal neutron capture cross-section are known. Consequently, the other attractive liquid metal fuels are those based on low melting point metals with good heat transfer and nuclear properties. Liquid uranium and plutonium alloys may find application in the future as fast reactor fuels, where the range of possible alloying elements and container materials can be extended considerably because of the decrease in their neutron capture cross-sections with rising neutron energy.

Metals which are possible thermal reactor coolants, and which may be considered for suitability as fuel carriers, are listed in Table I. Water and some unsuitable liquid metals are included for comparison purposes; the undesirable properties which cause their rejection are underlined.

Only three metals, lithium, lead and bismuth, have appreciable solubilities for uranium, and of these bismuth is outstanding in both this property and low neutron capture. It may be possible to increase the solubility of uranium in lithium and in lead by additions of further alloying elements, and work is proceeding on these lines in Great Britain. The

TABLE 1*

Metal	Melting point °C	Boiling point °C	Density gm/cc at 500°C	Thermal conductivity cal sec^{-1} cm^{-1} °C^{-1} at 500°C	Viscosity Centipoise at 500°C	Heat capacity cal gm^{-1} °C^{-1} at 500°C	Thermal neutron capture Cross-section (barns)	Solubility for Uranium	Remarks
Bismuth	271	1477	9.75	0.037	1.84	0.0365	0.015	1000 ppm (300°C) 2.7% (650°C)†	Attacks ferrous alloys
Gallium	29.9	1983	5.8	0.08 (30°C)	0.81	0.082 (to 200°C)	2.2	Very low	Expensive and attacks all known containers.
Lead	327.4	1737	10.39	0.037	1.85	0.037	0.2 (lead 208. 0.00045)	Less than 1000 ppm (600°C)‡	Cheap, readily available, but attacks most containers
Lithium	179	1317	0.48	0.09 (230°C)	0.455 280°C)	1.0	65 (lithium 7. 0.033)	Approx. 1500 ppm (500°C)	Relatively scarce and expensive Attacks most containers
Magnesium	651	1103	1.6	–	–	0.317 (650°C)	0.3	500 ppm (650°C)	
Mercury	-38.9	357	12.88 (300°C)	0.03 (200°C)	1.01 (200°C)	0.032 (300°C)	430	–	
Potassium	63.7	760	0.72	0.090	0.17	0.183	2.5	Negligible	
Rubidium	39.0	688	1.475 (at M. pt.)	0.07 (at M. Pt.)	0.32 (220°C)	0.09 (at M. Pt.)	0.56	–	Scarce and expensive.
Sodium	97.8	883	0.83	0.160	0.239	0.302	0.45	Negligible§	Cheap, readily available. Relatively easy to contain
(Water)	0	100	1.0 (0°C)	0.001	0.259 (100°C)	1.0 (0°C)	0.6	–	

* U.S.A.E.C. Liquid Metals Handbook, 2nd Edition

† Teitel, R. J 1952

‡ Teitel, R. J. Gurinsky, D. H., and Bryner, S. J. 1954.

§ Mogard, H. 1955

solubility of plutonium will not be considered further, because supplies of this element are not likely to be available in Australia in the near future. Also, plutonium fuelled systems have lower possibilities in conversion factor, which is likely to affect their economy adversely.

The thermal neutron capture cross-section of lithium is so unfavourable that this metal would have to exhibit outstanding advantages in other properties to make it attractive for thermal reactors. Similar reasoning applies to lead, its cross-section being much lower than that of lithium, but still high compared with bismuth. In both cases these elements occur in nature as mixtures of isotopes with widely differing nuclear properties, and an interesting possibility is the use of the low capture cross-section isotopes lithium 7 and lead 208 as reactor coolants. Present isotope separation processes are not likely to produce lead 208 economically, but the greater ease of isotope separation with light elements may mean that lithium 7 is a feasible coolant in the future.

Both lead and lithium dissolve many other metals, and consequently pose compatibility problems with container materials, but these difficulties appear not insuperable (Hoffman, 1955, 1956). However, bismuth also exhibits slight solubility for many other metals, and it should be emphasised that the magnitude of the corrosion problems in the use of this metal was not apparent from a study of its properties, but emerged as a result of a considerable research program (Weeks, 1955).

Liquid fuels of higher uranium content may be made by adding amounts of fissile material exceeding the saturation solubility in bismuth and lead. In both cases uranium forms an intermetallic compound with the coolant (UBi_2, UPb_3) and the result is a slurry of the relevant compound in equilibrium with liquid metal solution. The solubility of uranium in the liquid phase increases rapidly with rise of temperature in both systems, and this causes difficulty when the slurry flows in a circuit of varying temperatures, as would occur in a reactor and heat exchanger combination. Small particles dissolve preferentially in the hot region because of their greater surface to volume ratio, and precipitation in the cold region tends to occur on the remaining coarse particles. The result is particle growth to such an extent that deposition and eventual blocking ("plugging") of the circuit occur. Any system containing particles in equilibrium with a solution is prone to this effect, which is a case of "temperature gradient mass transfer." For example, the system thorium-bismuth has been investigated for possible use as a neutron absorbing blanket for thermal reactors, but the thorium content required is such that a slurry of $ThBi_2$ in bismuth is necessary. The alloy systems thorium-bismuth and uranium-bismuth are very similar, and particle growth of the $ThBi_2$ is proving a serious problem in such a blanket design (Barton, 1957).

The final alternative for a liquid metal fuel is a slurry in which the dispersed fuel has negligible solubility in the liquid, so that temperature gradient mass transfer and consequent particle growth is also negligible. Such slurries could be based on any of the preferred liquid metal coolants, and could contain insoluble uranium compounds such as UO_2, UC or UBe_{13}, or uranium metal where this is insoluble. Again bismuth appears most attractive because of its favourable cross-section, but most of the problems associated with this metal are with container materials, and if these can be solved, it appears more logical to use bismuth as a solution type fuel rather than in a slurry. It remains, then, to consider whether a slurry formed with any other liquid metal has sufficent advantages to compete with a bismuth solution.

Sodium and potassium have negligible solubility for uranium, and hence could be used as slurry vehicles for the metal or its compounds. Potassium has poor nuclear properties, and is more difficult to contain than sodium, and its only advantage appears to be a lower melting point. Alloys of potassium and sodium (NaK) have been used (Trocki, 1955) as reactor coolants, the potassium being added merely to lower the melting point of the coolant.

The thermal neutron capture cross-section of sodium is high compared with that of bismuth, but reference to Table I indicates that the heat transfer properties of sodium are much better than those of bismuth. This means that less sodium coolant is required for a given heat output, i.e., the poor nuclear properties of sodium are partly offset by its good thermal properties. In an insoluble slurry type fuel, the fissile material concentration is not dependent on a metallurgical property of the coolant. A liquid metal fuel could be made, based on sodium and containing uranium, in which the neutron losses to the coolant would be no greater than in a bismuth solution fuel, and in which a similar fuel rating could be achieved.

Comparison between bismuth and sodium as fuel carriers

Metallurgical aspects

A considerable amount of metallurgical research has been devoted to the problem of finding suitable container materials for a bismuth-uranium fuel solution. Workers at Brookhaven National Laboratory (Weeks, 1955) have shown that the most promising and readily available constructional materials for bismuth circuits are steels containing 2.5 — 5 per cent. chromium and some molybdenum, but low in nickel and carbon. However, the extent of corrosion and mass transfer in these materials has led to investigation of corrosion-inhibiting additives in the bismuth. Additions of magnesium as a deoxidant and zirconium as an inhibitor have been found beneficial, and the mechanism of inhibition has been proved to be the formation of a film of zirconium nitride on the surface of the steel, the nitrogen being an inherent impurity in all steels. Horsley, working at Harwell, has shown that deliberate addition of further nitrogen to the steel prolongs the inhibition of corrosion (Horsley, 1957). However, the effects of high flux irradiation on the inhibition mechanism, and on the long-term corrosion resistance of a reactor circuit, are still unknown. Research at Ames Laboratory (Fisher, 1956) has shown that bismuth can be circulated for long periods at high temperatures (1000°C) in loops of pure tantalum, but this metal is expensive, difficult to fabricate, and has an undesirably high thermal neutron capture cross-section (22 barns). Generally, it appears that the problem of circulating bismuth for long periods under reactor conditions in materials which have desirable engineering, nuclear and economic characteristics, has not yet been solved.

On the other hand, sodium and NaK have been circulated in loops and in reactor circuits for long periods at high temperatures with little difficulty. Several reactors have been built and are operating (Siegel, 1955, AECL CRR-590) using sodium as coolant and stainless steel of the 18/8/1 type as constructional material. It is recognised that oxygen as an impurity has a profound effect on the corrosion of many metals by sodium (Liquid Metals Handbook, 1955), but adequate measures for control of oxygen content in sodium as a coolant have been developed. Thus, on the basis of compatibility with containers, sodium has distinct advantages over bismuth.

Engineering Aspects

An important factor in a liquid metal fuel reactor design is the total investment of fissile material in the reactor circuit. Because fuel must pass through heat exchangers, pumps, processing vessels, and other components external to the reactor core, the total amount of fuel in a system based on bismuth may be three to five times that in a reactor core (Dalton, 1957). As the fuel rating of the reactor in megawatts per kilogram of fissile material must be considered on the basis of total fuel investment, the calculated rating of the core must be divided by the ratio of total hold-up to core investment.

In the case of sodium, a fuel slurry having neutron losses corresponding to those in a bismuth solution must have a higher uranium content. In the reactor core this is satisfactory for heat removal because of the high specific heat of sodium, but in the heat exchangers this same property makes necessary a greater heat transfer surface per unit of fuel than is required for the bismuth solution. As a result, the minimum possible volume of heat exchanger for a given power output contains more fuel for a sodium slurry than for a bismuth solution. Advances in heat exchanger design may reduce the actual value of this extra fuel hold-up, but preliminary calculations (Berglin, 1957; Dalton, 1957) indicate that the ratio of total investment to core investment for a sodium slurry system may be between 10 and 15. Thus, the overall fuel rating in a sodium system is probably inferior to that in a bismuth system.

A circulating liquid metal fuel solution would be homogeneous liquid initially, although fission product release would be expected to produce some solids (e.g., halides) and gases (e.g., xenon, krypton). Continuous fuel processing outside the reactor core would keep these to low equilibrium values. On the other hand, a sodium slurry presents new problems in the circulation of the fuel itself, because there is little experience of maintaining dispersion in light liquid systems containing fine dense particles. Possible fuel additions are discussed later, but the density range of these is between 4.4 gm/c.c. and 19 gm/c.c., whereas the density of sodium at 500°C is 0.8 gm/c.c. The problems of maintaining dispersion in such liquid-solid systems are being studied by Cairns (1957), using tungsten and other powders in water as analogues for uranium and other fuel additives in sodium. Initial results indicate that uniform dispersion can be maintained in circulating systems at reasonably low velocities of about 6.5 ft./sec. for tungsten particles smaller than 10 microns diameter in water.

This analogue work is to continue, because it is necessary to investigate the behaviour of such a dispersion under all conditions likely to be encountered in a reactor circuit. The behaviour of a reactor, fuelled by uranium particles dispersed in sodium has been studied theoretically by Dalton and Thompson (1957), who concluded that short-term variations of fuel concentration by more than 2 per cent. in the circulating fuel/coolant would have serious consequences in the form of power and temperature transients. Thus, the conditions necessary to ensure uniformity in the fuel feed must be established.

Work with aqueous suspensions may not give adequate information on the sodium based systems, because the wetting characteristics are different, and ionic phenomena do not occur in sodium. Thus, similar studies of particle behaviour in flowing sodium must be undertaken. Small loops to do this, using electromagnetic pumps, are being built as part of the metallurgical research program at Lucas Heights, and a larger loop has already been constructed by the chemical engineering section.

Sodium has several minor advantages over bismuth in the engineering of reactor circuits. The lower melting point of sodium results in less difficult "heat tracing" of circuits to ensure that all regions are molten at start-up. Heat tracing for bismuth circuits must be carefully designed because the metal expands on solidification and may fracture pipework if allowed to freeze indiscriminately. Another factor in favour of sodium is its high electrical conductivity, enabling it to be pumped by electromagnetic means far more readily than can bismuth. Finally, the sheer weight of large volumes of bismuth constitutes a disadvantage in necessitating heavy pipework and supports, whereas sodium is lighter than water.

Chemical aspects

There is no clear distinction between chemistry and metallurgy when considering phenomena in liquid metals. However, certain problems of both the liquid fuels may be termed chemical.

The bismuth fuel solution under study in the U.S.A. (Williams, 1954; Weeks, 1955) contains 600 ppm of uranium. Under reactor conditions, and operating on either U233 or U235, other non-fissile uranium isotopes would be produced and would build up in the fuel, thus limiting the fissile content if the saturation solubility is not to be exceeded. This problem does not occur in a slurry fuel, because the allowable concentration does not depend on solubility.

Continuous chemical processing of bismuth-based fuel is being studied, using molten salt contacting techniques, which are promising (Dwyer, 1955). No simple system of continuous processing for sodium slurries has been devised to date, and it may be necessary to separate fissile material from coolant before processing. A relatively simple means of separation exists in the hydroclone (Berglin, 1957), and work on this principle is proceeding in the A.A.E.C. research laboratories.

A further point favouring bismuth fuels from the processing viewpoint is the possibility of a simple operation to replenish the fuel with uranium produced in a neutron-capturing breeder blanket surrounding the reactor core. If a thorium-bismuth slurry (containing $ThBi_2$) is used as blanket, U233 is produced in the $ThBi_2$ particles. Raising the temperature of the alloy to the point at which all thorium and uranium is in solution, and cooling to reconstitute the slurry, results in significant concentration of uranium in the liquid phase, which could be further concentrated and added to the core (Dwyer, 1955). The particle growth problems of a thorium-bismuth alloy have been described above, but there is considerable incentive to solve them.

Finally, the effects of neutron irradiation on the coolant itself lead to chemical problems in the case of bismuth. The product of neutron capture in bismuth is polonium, a highly active alpha emitter, which would constitute a severe health hazard in bismuth circuits. Neutron irradiation of sodium produces the isotope Na24, a short-lived gamma emitter, which decays to magnesium 24; this is no problem and may even be beneficial is some reactor circuits as magnesium is a powerful deoxidant. In the sodium slurry liquid metal fuel other elements present are likely to have higher affinity for oxygen.

Summary—the Australian L.M.F.R. project

Of the two most promising liquid metal fuels, the bismuth solution has been chosen by workers in both the U.S.A. and England on the grounds of low thermal neutron capture cross-section, ease of handling in comparison with a slurry, and the promise of simple high temperature chemical processing. Unfortunately, the problem of corrosion of structural materials by liquid bismuth has proved much more difficult than was anticipated and, indeed, may prove insurmountable.

Liquid sodium has some disadvantages when compared with bismuth, in that a fuel slurry is more difficult to circulate than a solution, and the higher neutron absoption leads to larger fuel investments in the reactor system as a whole. However, the fact that sodium has extremely low solubility for most container materials gives promise that the compatibility problems of a sodium-based system will be less difficult to solve. The sodium slurry system for thermal reactors is not being studied elsewhere, and for these reasons it was selected as a suitable project for the Research Establishment of the Australian Atomic Energy Commission.

There are many possible combinations of moderator and fuel additions for use with a sodium coolant, and it is not possible to study all of these with the limited staff and facilities available. Thus, some elimination was necessary, although it is realised that in the process some promising combinations may have been deleted. The choice of materials for detailed study is outlined briefly in the following sections.

The choice of moderator

For high temperature service, possible reactor moderators are graphite, beryllium and beryllium oxide.

The smaller critical sizes of reactors using beryllium and its oxide as moderators give these materials advantages over graphite in permitting lower fuel inventory and power output. Further, there is a reasonable possibility of making the fuel/coolant compatible with beryllium or its oxide, whereas graphite and sodium are incompatible. In the only sodium-cooled graphite moderated reactor built to date, it was necessary to can the graphite in zirconium (Siegel, 1955), because the available graphites were all penetrated and disintegrated by liquid sodium.

Beryllium exhibits a nuclear property which enhances its value as a moderator, but which may have serious mechanical consequences. Two nuclear reactions occur in beryllium under fast neutron irradiation; these are the n, α and n, 2n reactions, both of which produce helium as one end product. Calculations based on the available data shows that a "neutron enhancement" of several per cent. is possible by the n, 2n reaction in a beryllium moderated system ("Beryllium," ASM, 1956), and this increases the possibility of breeding on the Th232-U233 cycle in a uranium-sodium-beryllium reactor. On the other hand, the effect of the helium atoms on the beryllium is not known. Calculations by the author, and later by Hickman (1958), have indicated that at high temperatures the beryllium may swell as a result of helium bubble formation. Experimental irradiations of beryllium at high temperatures in HIFAR are being undertaken, and will provide both fundamental and technological data on this problem.

Beryllium oxide has several disadvantages compared with metallic beryllium; the neutron enhancement by n, 2n reaction is likely to be less, the material is more difficult to fabricate into shapes of low porosity and the thermal shock resistance is inferior.

The mechanical effects of helium production in beryllium oxide are difficult to predict. On one hand, the relatively open lattice structure of the oxide may accommodate helium atoms more readily than the almost close-packed metallic lattice; and, on the other, the oxide must be considered as a brittle material, so that sufficient internal strain set up by the presence of many foreign atoms would cause cracking and possible crumbling.

Beryllium metal has been chosen as a moderator material for initial investigations on the feasibility of a sodium-based system, because it has the best combination of nuclear properties, it is relatively easy to fabricate, has good resistance to thermal shock, and it is negligibly soluble in sodium. The disadvantages of the metal, compared with its oxide, are mainly the temperature limitation imposed by the low creep strength of beryllium at high temperatures, and corrosion problems caused by oxygen in the sodium. Beryllium oxide corrosion in sodium is being studied in France (Grison, 1956), and the results of this work are of continuing interest to the Australian group.

Possible fuel additions to sodium

The fissile material may be uranium metal or any compound capable of being prepared as a fine powder which is wetted by sodium, and which does not contain elements of unacceptably high thermal neutron capture cross-section. Because fission of uranium atoms in a compound will ultimately release the other combined atoms, these must not be of a substance which will cause corrosion of the moderator or circuit materials. Three uranium compounds have acceptable nuclear properties, are readily prepared and readily wetted by liquid sodium. These are UO_2 UC and UBe_{13}.

The particle size of the powder is determined by two factors:

- (i) To enable fission products to escape from the particles, with the attendant possibility of continuous de-poisoning of the reactor, particles less than 10 microns in diameter are desirable (Hickman, 1958).
- (ii) To mantain uniform dispersion, it is desirable to have particles with minimum settling velocity, i.e., as small as possible.

Consideration of settling rates of metallic uranium in sodium led to the choice of particles of less than 10 microns for initial studies (Kelly, 1958).

Uranium metal may be prepared in the form of particles smaller than 10 microns by repeated hydriding and dehydriding of massive uranium (Kelly, 1958). Uranium powder is wetted by liquid sodium at approximately 400°C, and does not de-wet on cooling (Taylor, 1955). Uranium is not corroded in liquid sodium provided that the oxygen content of the liquid metal is kept sufficiently low (Mogard, 1955). A disadvantage of elemental uranium as a fuel is its density, 18.3 gm/c.c., compared with that of sodium at 500°C (0.83).

Uranium oxide UO_2 has the advantage of being readily prepared as a fine powder. It is wetted by sodium at about 380°C (Taylor, 1955); a dispersion of UO_2 in sodium is being studied by Abraham (1957) at Argonne National Laboratory as a possible fast reactor fuel. A possible disadvantage of UO_2 is the liberation of oxygen to the system by fission of uranium. The most serious compatibility problems of sodium are those involving oxygen as an impurity, and this is particularly true of circuits in which metals with a high affinity for oxygen are present, e.g., beryllium. The influence of oxygen released from UO_2 on the corrosion of beryllium in sodium is to be investigated by irradiation tests in HIFAR.

Uranium carbide may have a similar disadvantage; if carbon is released by fission, it will undergo mass transfer in sodium to any metal forming a stable carbide. This is an example of "concentration gradient mass transfer," in which even minute solubility of a substance in a liquid metal will cause appreciable transfer from a "source" to a "sink" in a circuit, even in the absence of a temperature gradient. No work is being done on uranium carbide because it does not possess any outstanding advantages over uranium metal or UBe_{13}.

The most interesting fuel addition is UBe_{13}, the inter-metallic compound formed between uranium and beryllium. It has the advantage of low density (4.4 gm/c.c.) compared with uranium metal, and this reduces settling out problems of the dispersion. The compound is relatively easy to prepare by heating finely-divided mixed powders of uranium and beryllium to 1,100°C. Further advantages of this fuel addition emerge when considering metallurgical aspects of the sodium-uranium-beryllium system.

Metallurgical problems of the sodium-based fuel/coolant

Mention has been made of the influence of oxygen impurity on corrosion and mass transfer of metals in flowing sodium, and the effect is greatest for metals of high affinity for oxygen. In systems using sodium as a coolant only, several methods have been developed to maintain a low oxygen concentration in the liquid metal.

One such system is "cold trapping," consisting of passing the sodium stream, or a portion of it, through a region at temperatures just above the melting point. Because the solubility of oxygen in sodium falls with decreasing temperature, precipitation of sodium oxide occurs in the cold trap, and filtration or settling in a region of low velocity can retain this oxide. By this means oxygen content can be minimised to less than 100 parts per million. This system is not applicable to circuits containing metals with oxides more stable than sodium oxide.

A second system is to "hot trap" the sodium (Siegel, 1955) by passing it through a region containing material with a higher affinity for oxygen than any other material in the circuit. Results to date indicate that it is necessary to run hot traps higher in temperature than the material to be protected, particularly for reactive materials such as beryllium or zirconium. Attempts have been made to protect beryllium in sodium circuits by hot-trapping with thorium (Bett, 1958). Theoretically this should be effective if both metals are at the same temperature, because thorium oxide is more stable than beryllium oxide. However, the results indicate that considerable oxidation of beryllium occurs despite the hot trap, indicating that an important factor is the rate of oxidation. The kinetics of oxidation of reactive metals in sodium will be studied by chemists and metallurgists as part of the Australian project.

In the case of a liquid metal fuel containing fine particles, neither of these systems can be adopted, because both depend for removal of oxygen on a low-velocity reaction vessel, which would allow settling of the fuel. A third alternative is the use of a soluble "getter" in the sodium, of which calcium is the most promising. Preliminary work indicates that corrosion of beryllium metal in sodium is reduced considerably by the presence of calcium (Bett, 1958). However, calcium and nickel interact, and the effect of calcium additions on nickel-bearing stainless steels is being studied. Initial results indicate that the quantity of calcium required for "gettering" has negligible effects on 18/8/1 stainless steel (Bett, 1958).

The use of UBe_{13} as fuel addition may provide another method of oxygen control. Fission of uranium atoms in the compound particles will release beryllium atoms, and it is hoped that the higher chemical activity of this beryllium compared with the moderator material will result in continuous "gettering" of oxygen without appreciable corrosion of the moderator. This hypothesis will be evaluated by irradiation experiments in HIFAR.

In addition to the problems of oxygen impurity, fission products released within the sodium may cause corrosion and mass transfer of moderator and circuit materials. The system is so complex that experimental work, including irradiation tests, will provide the only real guide. It may be seen in general that a considerable amount of metallurgical research is required before the fuel-coolant, moderator, and constructional materials for a reactor can be defined.

Summary

The reasons for the choice of a sodium-based liquid metal fuel system as a high temperature reactor study in Australia may be summarised as follows:—

(i) A liquid metal fuel system has potential advantages in high fuel rating, high burn-up, and high temperature operation, the latter leading to high thermal efficiency.

(ii) Only two liquid metal fuels are attractive; these are a solution of uranium in liquid bismuth, and a dispersion of uranium or a compound in liquid sodium. Bismuth solution systems are being studied in detail in at least two other countries, whereas sodium fuels are not receiving much attention.

(iii) Sodium presents less problems of compatibility with container materials than does bismuth. It is in this field that the bismuth system has encountered its most critical technological barriers.

Not all of the many possible combinations of fuel, moderator, and container materials have been discussed, but the aim has been to indicate the main reasoning behind the current research programme, and the nature of the problems involved.

Sufficient design calculations have been done to indicate that the sodium-based system is a promising one if the technological problems can be solved. Design of a liquid metal fuel reactor experiment must await satisfactory completion of compatibility and fluid flow studies, which involve considerable metallurgical, chemical, and engineering effort.

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The Philosophy and Practice of Radioactive Waste Disposal

By R. B. Temple*

This general review is largely concerned with the problems of disposing of the large amounts of highly-radioactive waste that will result from a large-scale nuclear power program employing fission reactors. The dangers of dilution-dispersal are outlined, and particular attention is drawn to the possibility of biological reconcentration of the dispersed radioactivity. The conclusion is reached that it is imperative to find some method of permanently storing the activity in a non-leachable solid form, if the fullest use is to be made of nuclear power production. At present the "temporary expedient" of tank storage in liquid form is the only practicable method, but by 1965 other methods will have to be found.

Introduction

Radioactively contaminated waste is potentially harmful, and must be disposed of in such a way that it cannot form a hazard to mankind. In practice we can recognise two schools of thought on the disposal of radioactivity—the "concentrate and store" school and the "dilute and disperse" school. At the present time each method is used to a greater or lesser extent, which is largely determined by expediency. There is as yet no international control of the disposal of radioactive waste, although this is becoming a matter of increasing importance and urgency as the number of atomic energy sites multiplies. Of the three types of waste involved, gas, liquid and solid, the second is of the greatest immediate importance, but in a large power program it will also be necessary to consider the escape of radioactive gases of long half-life to the atmosphere, in particular krypton 85. The toxicities of radioactive materials are many orders of magnitude higher than those of non-radioactive substances, and extraordinary care has to be taken to avoid contaminating air, water, and food supplies.

Although for a given absorption of energy the effect on the human body is the same whether the source is internal or external, from the point of view of waste disposal the ingestion hazard is the more important. It must, of course, be realised that there is slight natural radioactivity in the air we breathe and everything we eat and drink, deriving from minute traces of the natural radioactive elements. This background may vary from one place to another, and is in addition to the irradiation by cosmic ray particles to which we are continually exposed. The problem is to avoid increasing this background to a dangerous level, although unfortunately it is not easy to decide what level of activity constitutes a danger.

The followers of the "dilute and disperse" school derive their philosophy from the fact that we are all continually exposed to radiation throughout our lives, and they argue that small increases in the background will produce no measurable ill-effects in the exposed organism. Based on clinical observations dating from the discovery of radium, and on experimental data acquired in the last 10 to 15 years, upper limits have been laid down for exposure to external radiation and for radioactivity in air and drinking water. These are the so-called maximum permissible levels or concentrations, usually designated m.p.l. or m.p.c. These are believed, on the basis of present knowledge, to be unlikely to produce detectable ill-effects in human beings continuously exposed to them. These maximum permissible concentrations are exceedingly minute by the ordinary standards of chemistry. Some of the more important ones are given in Table 1.

This philosophy would justify the disposal of radioactive liquid into a domestic water supply if this were done in such a way that the ingested concentrations did not exceed the maximum permissible ones for a large population. Furthermore, this might be achieved solely by dilution, as in the case of the activity induced in the Columbia River by the Hanford

TABLE 1.—DRINKING WATER LEVELS FOR CONTINUOUS EXPOSURE FOR LARGE POPULATIONS (I.C.R.P., 1955)

Nuclide	Drinking water level for continuous exposure for large populations (microcuries/ml)	ppm.
Pu239	1.5×10^{-8} microcurie/ml	2.4×10^{-11}
Sr90	8×10^{-9}	6×10^{-11}
Ra226	4×10^{-10}	4×10^{-16}
Rn222	2×10^{-8}	1.3×10^{-15}
I131	3×10^{-7}	2×10^{-13}
Cs137	1.5×10^{-5}	2×10^{-7}
	Air level for continuous exposure of large populations	
I131	3×10^{-11}	
Rn222	10^{-9}	
A41	5×10^{-9}	
Xe135	2×10^{-8}	
Kr85	4×10^{-8}	

Compare these values with the following estimated Ra content of tap water supplies. (Lowden & Solon, 1956).

Location	Activity Present (microcuries/ml)
Deep wells, near Chicago, serving 100,000 people	$6\text{-}16 \times 10^{-9}$
Joliet, Ill. (pop. 50,000)	6×10^{-9}
Frankfurt a. M.	0.3×10^{-9}
Average for 42 U.S. cities (range from 0.0-0.17 x 10^{-9} microcurie/ml)	0.042×10^{-9}
Thames, below AERE Harwell discharge point	0.01×10^{-9}

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*Australian Atomic Energy Commission Research Establishment. Manuscript received March 14, 1958.

plutonium piles, or by a combination of chemical treatment and subsequent dilution as at Harwell, where the Thames provides the dilution and also part of the drinking water for London and other towns.

On the other hand, the "concentrate and store" school believes that any increase in activity of the background is potentially dangerous, and that when the effects are integrated over a large population, they may materially add to the burden of suffering and disease to be borne by the community. The tolerance levels laid down by the I.C.R.P. have been gradually lowered over a period of years, and there now appears to be general agreement among radiobiologists that there is no threshold level of irradiation below which the exposed organism would be free from damage. The difficulty is to calculate the effects of chronic exposure to low levels of irradiation and to relate the hazard to other dangers of life which are normally accepted. Because the release of radioactivity is an irreversible process, and the rates of decay of the biologically significant nuclides very slow ($T_{\frac{1}{2}}$ Cs137 = 30 years. Sr90 = 28 years and Pu239 = 24×10^3 years), it is obvious that the emphasis should be on caution until further data have been collected. Furthermore, the calculations on which the so-called safe levels of intake are based can be upset by unexpected concentration steps which may occur in animal or vegetable matter forming part of a human food cycle. Certain organisms have extremely large concentration factors for some elements present in fission products, and this is particularly probable where the element is not normally present in sufficient quantities for it to be in equilibrium between the organism and its environment. Under such circumstances biological concentration factors up to 850,000 have been reported by Krumholtz (1954) (in this case for P32). Because of such possibilities, and because many more data are required on the human uptake of radioactive elements and on the long term effects produced on the body by chronic exposure to low concentrations, the "maximum permissible levels" should be regarded merely as *upper limits* that are *thought to be tolerable,* and until further information is available, actual discharge concentrations should be kept as far below these levels as possible. In practice, however, there seems to be a growing tendency for those responsible for waste disposal from atomic energy sites—engineers, and not biologists or geneticists—to look on the maximum permissible levels as routine discharge levels, to be approached as closely as possible at all times so as to reduce the amount of storage or chemical treatment that is needed. It must be emphasised here that the maximum permissible concentrations refer to intake only, not to discharge levels. In relating the two, concentration factors have to be taken into account. Rather too great a reliance is perhaps placed on these maximum permissible concentrations, without the gaps in the knowledge on which these figures are based being realised.

Almost complete containment of all radioactive wastes from atomic energy sites could be achieved if desired. (The qualification is necessary because a limiting figure for the amount discharged is set by the volume discharged and the limit of detection for the radio isotopes involved.) However, removal of all radioactivity down to such limits would be expensive and under present circumstances would probably make the cost of nuclear power unattractive. It is, therefore, up to a community to balance its need for nuclear power against the risk involved in raising the radioactive content of the environment. At the present stage of atomic energy development, the total amounts of activity that have been released from nuclear energy establishments are small, and the possibility of harmful effects probably negligible. (This is not true of H-bomb debris; because of the quantities of radioactive nuclides released and the wide dissemination, it is certain that their effect will *not* be negligible.) As the use of radioisotopes becomes more widespread and atomic energy sites multiply, however, the practice of dilution and dispersal may have to be modified considerably to prevent exposing large sections of the population to levels much above the background.

Present practice

At present most major waste-producing sites effect a compromise. Very high level wastes (of the order of 100 curies/litre) are stored. Intermediate level wastes are chemically treated to yield a low-level liquid and a sludge containing most of the activity, which also has to be stored. The effluent is disposed of (with other low-level waste) by dilution and release to inland water, or to the sea. The usual chemical treatments involve flocculation with calcium phosphate or aluminium hydroxide, at a pH of about 11. Such a process may be used in conjunction with columns of vermiculite, or even wood sawdust, which will remove most of the remaining activity from solutions. Small quantities of short-lived isotopes, such as may be used therapeutically in hospitals or as tracers in small research laboratories, can be disposed of to domestic sewers, but the increase of this practice will have to be watched to pevent build up in pipes, sewers and sewerage treatment plants.

At present, the United Kingdom is one of the few countries practising dispersal on any considerable scale. At Windscale, for example, 6,000 curies of mixed beta-gamma emitters (mainly 9-month-old fission products, the bulk of which will be Sr90, Nb95, Cs137, Ce144 and Pm147), are discharged to the Irish Sea each month, while at the new Dounreay processing site it is hoped to discharge 1,000 curies per day of beta-gamma emitters. As an example of the somewhat unusual phenomena which may arise during waste

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[*] The "maximum permissible concentrations" are laid down for occupationally exposed persons under medical supervision. For small non-occupationally exposed populations, the levels must not exceed 1/10th of the m.p.c., while for large populations the tacit agreement is that the levels must not exceed 1/100th of the m.p.c.

disposal, the discharge levels at Windscale are thought to be limited by the uptake of ruthenium by seaweed growing nearby, which is eaten in parts of Wales (Farmer, 1956), and at Dounreay it will be limited by the accumulation of plutonium in the flesh of the considerable number of lobsters caught for market in this area.

Sea disposal

At one time it was thought that very large amounts of activity could safely be injected into the sea, because this already contains considerable natural radioactivity. (Table 2.)

Ignoring the practical difficulties of effecting perfect mixing in any such disposal operation, it has, however, been shown by several authors (Rodger, 1954; Renn, 1955; Claus, 1955) that ocean disposal is out of the question as a remedy for the problems of waste disposal created by even a moderate-sized program of nuclear power production, because the quantities of radioactive material involved are enormous compared to anything previously experienced.

For example, it has been estimated that if the world production of electricity by fission reactors increased to the point where it was consuming 1,000 tons of fissile material per year, the equilibrium amount of artificial activity arising from Sr90 alone would be about 7×10^{10}c. requiring between 5 and 10 per cent. of all the oceans for dilution to drinking water tolerance (Rodger & Fineman, 1956). As Glueckauf has pointed out, this amount of fission product waste is probably an outside estimate of the amount with which we shall have to cope in the foreseeable future (Glueckauf, 1955a). However, the situation is still alarming, even for a much more modest program such as that for the Central Electricity Authority in the U.K. At the present rate of expansion this would call for the consumption of about 20 tons of fissile material per year by about 1958. After 50 years at this rate of consumption, the long lived fission products would have built up to the following approximate levels.

TABLE 2.—NATURAL RADIOACTIVITY IN THE OCEANS (REVELLE, 1955)

Nuclide	Total activity in oceans (curies)	Approximate specific activity	
		d/ml/sec	microcuries/ml
K40	4.6×10^{11}	1.2×10^{-2}	3.2×10^{-7}
U238	3.8×10^{9}	1×10^{-4}	2.7×10^{-9}
Th232	8×10^{6}	2×10^{-7}	5×10^{-12}
Ra226	1.1×10^{9}	3×10^{-5}	8×10^{-10}
H3	1.2×10^{7}	2.5×10^{-5}	7×10^{-10}

Even for this program, the completion of which is perfectly feasible on the basis of present technology, the dispersal of the Sr90 alone would require 50 million cubic miles of sea water for reduction to drinking water tolerance for large populations. In any case, it is by no means certain that drinking water levels can be applied to sea water, because of the complicated biological chains involved and the probability of caesium and strontium concentration by plankton and by predatory animals, by mechanisms of which we are largely ignorant. The release of large quantities of Kr85 to the atmosphere would also be undesirable because of the possible formation of local high concentrations. It is, therefore, obvious that dilution alone is incapable of solving the waste problem, and that some method of safe permanent storage must be found. Such storage arrangements must be virtually everlasting, because up to 1,000 years would be required for some wastes to decay to harmless levels. It has been suggested by Glueckauf that the problem would be alleviated if the long-lived biologically significant isotopes Cs137 and Sr90 were removed for separate storage, because the remaining nuclides would have decayed enough for disposal by dilution after about 15 years' storage (Glueckauf, 1955a). At this stage it might then be possible to extract the most valuable disintegration products, such as the stable Xe, Ru, Rh, Pd and the Tc, and offset them against the cost of effluent treatment (Glueckauf, 1955b). However, this scheme of treatment ignores the fact that if the activity from Sr90 is reduced by a factor between 100-1,000, further reduction is of little help, because the controlling factor is then the concentration of the long lived alpha-emitters Np237 ($T\frac{1}{2} = 2.2 \times 10^{6}$ years), Am241 ($T\frac{1}{2} = 470$ years) and Cm242 ($T\frac{1}{2} = 163$ days). The concentrations of these may be expected to rise as higher burn-up is achieved in reactor fuels or reactors using plutonium as fuel are brought into operation (McKay, 1956). Furthermore, even if the most hazardous isotopes could be effectively isolated in solid form from the short-lived nuclides, their fission heat might cause fusion and partial vaporisation unless continual cooling were applied.

TABLE 3.

Amounts of fission products produced after 10 years by the continuous burn-up of 20 tons of fissile material per year (curies)	Sea water volume to dilute to drinking water tolerance level for large populations. (Cubic miles).
Nuclide	
Zr95 2.9×10^{9}	1.7×10^{4}
Ce144 2.4×10^{9}	1.4×10^{4}
Ru106 2.3×10^{8}	55
Pm147 1.2×10^{9}	30
Sr90 2×10^{9}	6×10^{7}
Cs137 1.8×10^{9}	3×10^{4}
Tc99 4.6×10^{5}	0.5
pu 239 6.5×10^{4}	10^{3}
Kr85 1.6×10^{7}	6.3×10^{4} cu. miles of air to dilute to breathing tolerance for exposure of large populations.

Estimated total volume of oceans—5×10^{8} cu. miles.

Ground disposal

As an alternative to dispersal, various schemes of "fully" or "partially" controlled containment have been put forward. Partially controlled containment is the name given in the United States to schemes in which wastes are discharged into the ground. At present this practice is being followed regularly only in the United States, although some ground disposal has taken place at Chalk River. Ground disposal was first used at Hanford to dispose of low level wastes containing up to 50 microcuries/ml. of total beta-gamma radiation, and by 1955 several hundred thousands curies had been disposed of in rock-filled pits.

At Chalk River, contaminated water from the NRX disaster was disposed of to a sandy area adjoining the Ottawa River, as an emergency measure. Subsequently, some 2,000 curies of Sr90 and 100 gm. of plutonium have been deposited there, largely from the fuel element trench water. At one time, liquid waste containing plutonium was pumped into a small lagoon in the sand, but it was discovered that the fauna in the area were exhibiting growing levels of acquired radioactivity in their bodies. The practice has now been stopped. At Oak Ridge National Laboratory ground disposal has been tried for "intermediate level" wastes, while low-level laboratory wastes totalling 150 curies/yr diluted in a flow of 700,000 gallons per day have been run for some years into White Oak Lake, an artificial lake formed on the Clinch River, which acts as a settling pond for the ORNL wastes. The results of this operation have not been encouraging for future ground disposals of this type. This has been shown by an ecological study made over three years on White Oak Lake, during which 25,000 specimens of the flora and fauna were analysed and the results compared with others for neighbouring lakes which had not received radioactivity (Krumholtz, 1954). The average level of activity of the water was estimated to be 5×10^{-5} microcuries/ml. and it was computed that some of the fish had received continuous doses of radiation of 50 rads/yr. The growth of all species of fish was found to be slower than those in nearby lakes and the average life span of at least seven species about 25 per cent. lower; two species had become extinct. While there was no positive evidence that this was due to irradiation the circumstantial connection was strong. Every fish examined had selectively accumulated radioactivity, with Cs137 primarily in the soft tissues and Sr90 in bones and skin, in amounts up to 20-30,000 times that in the water. Concentrations of P32 up to 850,000 times over that present in the water were shown by certain algae. Krumholtz (loc. cit.) remarked that the concentration factors for some other elements present only in traces may be even higher. Migratory water fowl appeared to have accumulated an average of 5 micro-curies, mostly P32, in the edible parts of the body over the three years of the survey. Most serious of all, one muskrat had acquired a total body concentration of 100 microcuries of Sr90—about 1 microcurie/gm. of bone. This animal had an advanced bone cancer with secondaries in kidneys and lungs. One species of tree had absorbed enough radio-ruthenium to cause irradiation damage to the leaves.

In spite of these results, which have been obtained with relatively small quantities and may serve as a warning of the difficulties and dangers of disposing of major amounts of active waste, various other schemes for the earth disposal of high level liquids have been suggested in the U.S. Among them are injection into deep wells, or storage in cavities in salt mines and in the old rock domes of discarded oil wells. None of these can, however, really be entertained as a responsible method of solving the problem since control over possible movement of the wastes is lacking. Pending some more permanent method of fully controlled storage, the bulk of high level waste is best held in suitably protected tanks.

Tank storage

For a nuclear power program producing 20 tons of fission products per year, conventional solvent extraction processing with nitric acid as salting-out agent would produce about 200,000 cubic metres of high-level waste annually (Glueckauf, 1955a). After preliminary removal of the bulk of the nitric acid, it is possible to concentrate such a waste to about 1/400th of its original volume before deposition of solid begins. This would mean that an annual provision of about 500 cubic metres of very high-level tank storage capacity would have to be provided. This is a perfectly reasonable amount and the total cost of this operation has been estimated (Glueckauf, 1955a) at around £750,000. Though this is a large amount, it adds very little to the cost of the electricity generated.

The design of tanks suitable for holding such wastes is an extremely difficult and onerous task. One of the difficulties, apart from ensuring complete freedom from leaks and an extremely long life, is that the fission heat liberated is about 125 BTU/gallon/hour for a typical six-month-old fission product waste. This rate of heat evolution is enough to cause bumping and boiling of the contents unless continuous cooling is provided, and trapping devices are necessary to prevent the loss of active spray or vapour. Cooling might have to be applied for periods of 50 years or more. Such tank storage is potentially vulnerable to accidents (for example, earth movements) and to chemical corrosion, and the true life of the tanks under these conditions is unknown. However, it will almost certainly be less than the length of time necessary for the stored activity to decay to harmless levels.

This can, therefore, only be regarded as a temporary expedient, and a search is going on in the U.S.A., Canada and the U.K. for a truly permanent method of storage (Hatch, 1953, 1955; Amphlett, 1956; Evans, 1956; McVay, 1957). The proposal which seems to hold the greatest prospect at the moment is to evaporate the wastes to solid form, and—most promising of all—to reduce them to a solid containing the activity in non-leachable form (Durham & Watson, 1956; Durham, 1957).

Fixation in solid form

Although installation and operating costs are high, owing to the necessity for shielding and for obtaining decontamination factors (*) of the order of 10^7, partial evaporation has been used for a considerable time to reduce the demand for high level tank storage. Worthwhile further reduction in stored volume could be achieved by conversion of the waste to dry solids,

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* Decontamination factor = Activity present before treatment ÷ Activity present after treatment.

although the engineering difficulties of doing so would be very great. The present-day high level wastes that arise in a process using nitric acid as salting-out agent could be reduced to about 1/700 of their original volume if converted to nitrates, and this could probably be improved to 1/1000 by conversion to oxides. With such a concentration factor, the total yearly demand for storage for 20 tons fission products would be about 200 cubic metres of dry solids. In a typical waste of the type being discussed, the bulk of the solid present would be iron oxide from corrosion products. According to Amphlett (loc. cit.), spinel formation takes place during the conversion to oxide, with fixation of the bulk of the activity against subsequent leaching with water. As a further protection, conversion to a glass has been suggested by Chalk River workers (Durham, Durham & Watson, loc. cit.). The waste would be mixed with suitable proportions of base and flux and fired at temperatures between 1000° and 1500°C. to produce an aluminosilicate glass. Chalk River is examining the use of the mineral nepheline syenite for this purpose, while Amphlett, at A.E.R.E., Harwell, has obtained very satisfactory resistant glasses by using the local greensand soil with fluxes such as sodium nitrate, sodium carbonate or borax. Although some encouraging results have been obtained in laboratory tests, a great deal of work remains to be done to determine the optimum conditions for the formation of the glass and to measure the leaching rates of the various isotopes when "fixed." In particular, the long-term stability of the glasses to high levels of radiation has still to be determined.

In spite of formidable engineering difficulties, plans for pilot plant-scale tests are going forward at several places in North America. Such a plant must be capable of being run remotely for long periods without maintenance at extremely high levels of alpha, beta, and gamma activity. The most ambitious approach to the problem is that being made at the Reactor Testing Station at Arco, where a combination of spray drying and fluidised-bed roasting is being set up for the conversion to oxides. The intention of the Arco workers is to leach the resultant oxides to remove caesium and strontium in almost pure form for use as radiation sources (Jonke, 1956). However, the plant might be used as a first step in the glass-forming technique.

The density of the resulting glass is such that its volume equals the volume of the original concentrated waste. Our hypothetical output of 20 tons of fission products would thus mean that we should have to store 200 cubic metres of solidified waste per year. Some kind of cooling would certainly be needed to prevent self-melting in the first five years (Coppinger and Tomlinson, 1956). Amphlett has suggested that the glass blocks could be stored in water-filled tanks and the leach liquor processed as low level waste before release.

Unfortunately, the storage situation is much less promising for some more recent types of alloyed fuel elements or for solvent-extraction processes where, for example, aluminium nitrate is used as salting-out agent. Such a process may yield a waste 2.5M in aluminium nitrate, with a fission product concentration of about 1 gm/litre. Conversion of a waste of this kind to oxide provides a meagre 6-10-fold volume reduction. The resulting demand for storage capacity would be serious. It is therefore perhaps not too much to suggest that future methods of fuel processing will have to be chosen with one eye on the waste disposal problems involved, which is hardly the case at present.

Urgency of the problem

In spite of the number of possible methods of disposal that have been discussed, and the thousands of words that have been written about the subject for the past three or four years, the only practical way of handling high-level wastes at present is the method introduced as a temporary measure under wartime pressure, namely tank storage. Brightsen (1957), President of the Nuclear Science and Engineering Corporation, Pittsburgh, has recently observed that at present there are no other solutions to the problem in sight that could be brought into use in a short time. He also points out that the basic problem is one of biological hazard, not of economics, although this fact tends to be lost sight of frequently in the discussions of the subject. Research should therefore be directed towards the goal of lessening the biological danger, with cost a secondary consideration. Greatly increased efforts should also be made to assess the biological hazards of of the nuclides involved.

Already, many hundreds of tons of concentrated fission product solutions are in store at various points in the world, representing a very great potential danger to neighbouring populations in the event of accidental release. The U.K. program alone is expected to yield almost two tons of solid fission products by 1965. In spite of the manifest difficulties, it is therefore essential that some practicable method of safe storage should be speedily forthcoming if the development of fission power is not to be seriously hampered.

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Canadian Research Reactors

By D. G. Hurst*

Of the four Canadian research reactors in operation, NRX is chosen for detailed discussion because NRU and PTR have been commissioned only recently, and ZEEP is specialised in application to lattice measurements. The discussion of NRX covers its use for pure research in physics and chemistry, for isotope production, for loop tests, and for reactor research, including a study of uranium rod failures and their detection.

Introduction

The Canadian atomic energy project was founded during the war to pursue the study of heavy-water moderated reactors, in particular to design and build the NRX reactor. This reactor has been the mainstay of the project, and although it went into operation nearly 11 years ago it is still one of the world's principal research reactors. In 1952 a power surge severely damaged the core, destroying several uranium rods and releasing intensely radioactive fission products in the building. Despite the handicap of this radioactivity, the reactor was restored to operation in about 14 months.

A low power reactor, ZEEP, was completed in 1945 and operated until NRX was ready. After a few years of idleness it was re-commissioned to measure lattice parameters for a new research reactor and for power reactors.

The new reactor, known as NRU, is a 200 MW heavy-water moderated and cooled natural uranium reactor which went critical in October, 1957, and is now being brought up to power. It has high flux (3×10^{14} neutrons/sq. cm./sec.) over a large volume and extensive equipment for experimental work.

Within a few days of NRU's criticality a low power swimming pool reactor, PTR, was also brought to criticality. This reactor is intended for reactivity measurements by the "swing" or pile oscillator method and will permit an extension of measurements of the effect of irradiation on the neutron economy of fuel assemblies. Several arrangements of core and reflector can be set up including a central region of water or graphite to provide a choice of neutron spectra and of "importance" functions.

All of these reactors are at the Chalk River Laboratory which is a research establishment operated by Atomic Energy of Canada Limited, a Crown company. The first non-government reactor will be at McMaster University, Hamilton, Ontario, where a pool-type reactor is being installed. Operation will be at 1 MW with provision for operation at 5 MW if desired in future. It will provide facilities for neutron and radio-chemical research at the University and irradiation services for the heavily industrialised area of southern Ontario.

It is somewhat too soon to discuss NRU or PTR in detail, and most of this paper will be taken up by a discussion of NRX.

Description of NRX

General

NRX is a heavy-water-moderated, light-water-cooled high flux reactor. The core is contained in a cylindrical aluminium vessel, 8.75 ft. in diameter and 10.5 ft. high, surrounded on the curved sides by a graphite reflector. The present operating power is 40 megawatts and at this power the maximum flux is 6.8×10^{13} neutrons/sq. cm./sec. The fuel assemblies consist of vertical uranium metal rods, 1.36 in. in diameter and 10 ft. long, encased in aluminium and cooled by river water. The working limit of power from such rods is 330 kilowatts. The behaviour of uranium, although not ideal and as yet incompletely understood, has been better than was expected for 10 megawatt operation. Calculations of stresses at the full power of 40 megawatts give values which would be unacceptably high were there not the simple experimental fact of reasonable performance.

Beams of neutrons are taken out horizontally through holes in the graphite and the surrounding concrete shield. Access may be had to the maximum flux by using one of the vertical channels normally filled by a fuel rod. A central thimble 5.5 in. diameter permits a large assembly to be irradiated. None of the vertical holes has been used for neutron beams, in spite of a factor of three in available flux, because of interference from rod handling equipment. During the early stages the reactor was used for physical experiments, for production of radioactive isotopes and for studying the reactor characteristics. Since then loops for testing fuel and other materials inside the reactor in separate coolant circuits have become of prime importance.

Operation

The shift in emphasis from neutron beam and activation experiments to loops has required frequent changes in operating procedure. At first a weak link between the operating timetable and the research programme sufficed, but nowadays careful planning of each scheduled shutdown starts weeks in advance, and the changing of fuel rods, which at one time was the primary purpose of shutdowns, occupies only a small fraction of the downtime.

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A typical shutdown includes inspection and removal of uranium rods, examination of some of the fuel undergoing test in loops, and the changing of loops, i.e., cleaning, flushing, cutting out of old sections, welding in the new sections, filling, venting and testing.

The growth of the atomic energy program in Canada and the U.S.A. has placed heavy demands on NRX loop facilities. At present the space available for external equipment, the congestion at the reactor headers, and the reactivity balance, limit the number of loops to five in fuel rod positions and one in the central thimble.

Control

The control system of NRX has been through many changes. The first core had four control rods and eighteen shut-off rods. It was realised early that one control rod gave ample adjustment between settings of heavy-water level. Consequently the three others were removed, making space for isotope production. Later the number of shut-off rods was reduced.

Analysis of the NRX control system as a basis for design of new reactor systems has suggested improvements which are incorporated in the new reactors and which have influenced the NRX system.

Following the 1952 accident, the control system was thoroughly revised, and another revision is in process. The coincidence system developed for NRU may be roughly described as requiring signals in two out of three channels before it shuts down the reactor. A similar system is being applied to NRX.

Advantages of this coincidence system are reduction in spurious shutdowns resulting from failure or false signals in one channel and the ability to test a channel without interfering with reactor operation. The single control rod will shortly be removed and the power will be controlled entirely by automatic adjustment of the heavy-water level. The rate of discharge of the heavy water to the storage tanks has been increased and fast acting valves have been installed. Six shut-off rods (of a new design) are provided, enough to compensate for loss of the cooling water even if some fail to drop. The reactor is shut down by both shut-off rods and automatic lowering of the heavy water.

Use

The experimental facilities include two thermal columns roughly 6 ft. square and fifteen horizontal beam holes—three 12 in. diameter and twelve 4 in. diameter. The neutron flux in the thermal columns is predominantly Maxwellian, the cadmium ratio being about 1 in 20,000. This feature has been less important for many of the experiments than the availability of large regions flooded with slow neutrons. Holes in the thermal columns have also been used for neutron beams. The horizontal experimental holes extend through the shield and graphite reflector to the aluminium calandria wall. Each has a gate with which the radiation may be very greatly reduced.

At the outside face of the shield the spacing between holes is not large enough to prevent interference between adjacent experiments. On this account neutron spectrometers have been moved back from the reactor. The targets are then over 20 feet from the neutron source and the collimators are evacuated to reduce the attenuation of the neutron beam by air. Distance per se is not important as long as the desired collimation restricts the field of view, as seen from the target, to the inner end of the hole. Intensity at the target falls off when the angle of collimation includes more than the inner end of the hole.

The results of many neutron physics experiments have been published and need not be detailed here. These experiments include the measurement of neutron-capture gamma rays with a precision magnetic pair spectrometer, and recently also with a double crystal diffraction spectrometer; the study of the decay of the neutron; the determination of structure of matter by slow neutron scattering (angular distribution giving information about atomic locations and, together with energy distribution, about atomic oscillation and diffusion) and the measurement of cross sections.

Some of the experimental holes have been supplied with autoclaves for the study of corrosion in irradiated water at high temperature and high pressure, and with hollow uranium cylinders to convert the slow flux to fast flux for studies of irradiation damage.

One of the positions originally intended for isotope production has been converted to a beam hole. The lack of a built-in gate has been overcome by including a gate in the collimator.

The reactor power is automatically controlled in terms of neutron flux at a control ion chamber. However, each experimenter who needs an accurate record of the intensity must monitor the neutron flux at his positon, because variations in control rod position and heavy-water level alter the relation between the flux at the experiment and at the control point. Where energy is resolved, as in the neutron spectrometers, the resolved beam must be monitored on account of spectral intensity shifts. For example, the fraction of the total which appears in the diffracted beam changes as the heavy-water temperature rises following reactor start-up.

From the early days of NRX a wide variety of isotopes have been produced. At first the production was in the facilities on the west side of the reactor and in an annulus in the reflector. Later, positions inside the core were made available for isotopes. Samples in these positions are in higher flux, but can be changed only during reactor shutdowns.

The removal of the control rod will leave the top of the reactor bare and will permit the rod-removal flask to be placed in position during operation so that isotope rods may be charged or discharged. The major item of production is Cobalt 60, which is produced on a commercial basis in quantities limited by the space and reactivity available.

For chemical research, samples can be irradiated in a pneumatic carrier and delivered directly from the reactor to the chemistry building. There are hollow uranium cylinders in lattice positions for fast neutron irradiation of samples.

For biological research the radioactive isotopes rather than the reactor itself have been of interest, because the radiations inside the reactor are too mixed to permit ready interpretations of radiation effects.

Engineering research is becoming the most important use for the reactor, largely through the need to study the behaviour of fuel assemblies and coolants for future reactors. After some preliminary experiments in the central thimble with fuel elements under normal NRX conditions, there has been a gradually increasing programme for testing fuel elements under high pressure and temperature.

In the central thimble pressures of 2200 lbs./sq. inch and temperatures up to 560°F. are now used regularly. In standard lattice positions loops pressurised to 2200 lbs./sq. inch are in service. The auxiliary equipment of these loops takes up a great deal of space. Most of this equipment has to be shielded and some of the major changes around the reactor have to do with the provision of shielded space. The unexpectedly small evolution of radiolytic gas from the heavy water meant that the recombination system could be much smaller than that originally planned and the unused space was suitable for loop equipment.

There is a pit below the floor in front of the south thermal column. This measures 28 ft. by 20 ft. by 10 ft. deep and was intended to hold water for neutron diffusion experiments. It is now crowded with equipment from the loops. The helium gasholder which had been installed in a well below the floor of the main pile room has been removed to an area outside the building thereby making another shielded room available.

During 1952, a concrete structure was built in the reactor hall. This contains shielded space about 15 feet above the reactor floor and immediately adjacent to the top of the reactor. In other instances concrete block shielding has been built around loop equipment.

Until recently there was no alternative to steel, usually stainless steel, as the material for loop pressure vessels. The neutron absorption by the steel depresses the flux and imposes a large reactivity load which requires compensation. Enriched rods may be added to restore the reactivity. By enriching the fuel under test the desired power output may be attained but the thermal conditions in the reduced flux may not match the operating conditions planned in the future reactor. For over twelve months a zircalloy pressure vessel has been in the central thimble. This vessel is 4.52 inches O.D., 3.19 inches I.D., and 24 feet long and was designed for a pressure of 2500 psi at 650°F. The successful production of this pressure vessel points to progressive replacement of stainless steel by zircalloy in such applications.

The increased use of the reactor is reflected in the requirements of staff. The number of supervisors (graduate engineers) has risen from about 15 in 1948 to 22 and the number of operators from about 30 to 44. The increase is to take care of loop operation, handling and shipping irradiated fuel, and special irradiations not classed as loops, such as a UO_2 rod and a slurry experiment.

Fuel rod behaviour and monitoring

High flux and reactivity are valuable in a research reactor and there is an economic balance to be struck between the number of fuel rod failures, the maximum working flux and the thickness of absorbers like the fuel rod sheaths and cooling water. The power of NRX has been increased progressively from the intended 10 MW to the present 40 MW and the amount of aluminium and water in the fuel rod structure has been reduced. These changes have been guided by the rod failure rate and were associated with an investigation into the relation of uranium fabrication with its behaviour under irradiation.

The investigation was started as a result of the successful irradiation of some of the first fuel charge to 3000 MWD/tonne, an order of magnitude greater than had been predicted. As a consequence of this work much information has been collected on rod failures.

During the past four years about 50 rods have failed to the extent of releasing activity into the coolant. In most cases the mechanical effects are small, often the only obvious result is a split in the aluminum sheath. About half the failures occurred within an hour of the reactor being started after a shutdown. Many rods with a new style of sheath having a thickness of 1 mm instead of the previous 2 mm have been failing after irradiations of 500 to 1500 MWD/T when in high flux positions. Examination shows cracks in the metal under the split in the sheath. The increased failure rate with thin sheaths may be due to the additive effects of smaller restraint on the uranium and decreased ductility of the thinner sheath.

Another type of failure not necessarily accompanied by activity release is a transverse break. This seems to occur in rods which are in positions where the flux is at least 0.75 of the maximum.

Failures are detected either by a change of coolant pressure at the rod exit (each rod is connected to a pressure gauge and also to pressure switches in the safety circuit), or by activity in the cooling water.

Several methods are employed to detect this activity. A gamma-ray sensitive ion chamber in the effluent pipe with a delay of about 120 seconds from the reactor serves as a burst rod monitor and trips the reactor on a large increase in the signal. A neutron-sensitive counter at the same location monitors the neutron emission from the water. After 120 seconds the neutron emitting N^{17} has decayed and the detection of neutrons from the coolant is a specific indication of delayed neutron emitting fission products and therefore of exposed fissile material. The development of practical monitors based on this principle has recently been described by Millar (1957).

From the point of view of fuel rod monitoring NRU differs from NRX in the two important respects that the coolant is heavy water and that it is recirculated. High energy gamma rays produce photo-neutrons in the heavy water, tending to mask the delayed neutron signals, and due to the recirculation long lived activities build up in the coolant. By proper design the masking effect of the photo-neutrons can be overcome.

Because of the build-up due to recirculation it is desirable to monitor short-lived products. This points to delayed neutrons rather than overall gamma rays. However, the gaseous fission products have, in the main, short lives and gaseous fission products as well as delayed neutron monitors are being used in NRU.

Prototypes of both have been operating on the effluent from NRX. Although they are not considered part of the NRX instrumentation they have proved useful in the study of rod failures. Neither has shown a decisive superiority over the other but they are both considered superior to simple gamma ray detection. The delayed neutron monitors are effective only when the reactor is operating. To the extent that gaseous fission products have short lives the same is true of the gaseous fission product monitor, but there is some sensitivity to long-lived fission products.

The activity signals from a burst rod are diverse and have not yet been correlated with the mechanical effects. In about 30% of the cases a small, often transient, signal appears at least 10 minutes before the ultimate failure is indicated by a reactor "scram" due to either excessive activity or pressure change. There are of course rod failures due to mechanical distortion without sheath rupture and these are shown by pressure alone.

Future

The completion of NRU has not made NRX redundant. It will reduce the demand for full operation of NRX and will permit greater freedom in its programme. As to development of NRX we have a guide in CIR, the reactor, similar to NRX, which is being built in India under the Colombo Plan. The core vessel of CIR will have six vertical holes 4 inches in diameter. They will be available for loops in addition to the central thimble. It is not unlikely that the calandria of NRX will be replaced at some future time. This could come about either through accidental damage to the present calandria or through a deliberate choice; in fact there is a standby calandria which can be modified to incorporate new ideas.

Almost certainly, special positions in addition to the central thimble will be provided in this calandria. One of the difficulties with hot loops is the heating of the calandria tubes and resultant stress due to expansion. The insulation necessary to isolate the hot pipe from the calandria tube takes up considerable space and reduces the volume available for the fuel element. Some means of cooling loop positions may be installed in the new calandria.

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Many details and characteristics of NRX are given in Progress in Nuclear Energy Reactors, Vol. 1, Chapter 1. This also contains a list of references.

The NRU reactor is described by F. W. Gilbert in the August, 1957, issue of The Engineering Journal.

The First Year of DIDO Operation

By F. W. Fenning*

Introduction

Research reactor experience at the Atomic Energy Research Establishment, Harwell, England, until 1957 was almost entirely with gas-cooled, graphite-moderated reactors. Some of the Harwell staff had had experience of heavy water reactors at Chalk River in Canada but, to most, the operation of DIDO at high flux in March 1957 was a quite new experience.

The situation in Australia would appear to be very similar, in that some Australian Atomic Energy Commission staff have worked in Chalk River and at Harwell, and HIFAR is now operating. The hope is, therefore, that a review of Harwell experience during the first year of high flux operation of DIDO will be of value.

The DIDO reactor

The DIDO reactor has been described elsewhere by the author (1956), so only a brief description is given here. It is a slow neutron reactor using a U235—aluminium plate-type fuel element, which is cooled by heavy water which also acts as a moderator. Ten megawatts of heat are transferred from the heavy water to an ordinary water secondary circuit, which in turn disposes of its heat to atmosphere through cooling towers.

The main reactor tank is of aluminium about 6ft. in diameter, surrounded by a 2ft. thick graphite reflector. This is contained in a lead and steel walled tank followed by a barytes concrete shield. The overall dimension of the reactor at working floor level is 24ft. diameter and 14½ft. high. To limit the spread of fission products should an accident occur, the reactor and control room are enclosed in a gas-tight building, 70ft. in diameter, 70ft. high.

The main purposes of the reactor were:

(i) The irradiation of small fissile specimens to high burn-up under different control-conditions to enable better fuels to be developed. These require thermal neutrons.

(ii) The irradiation of non-fissile materials of use in power reactors. These include moderators (C, Be, BeO), canning materials (Be) and structural materials (Fe). Here the interest is in radiation damage effects, primarily those due to fast neutrons.

(iii) The production of high specific activity radioisotopes (mainly Co60). These require thermal neutrons.

(iv) One or two "loops" in which a fuel is exposed to thermal neutrons while a coolant is circulated over it under conditions very similar to those in a power reactor.

(v) Radiation chemical experiments where, for example, it is required to know how a coolant reacts with a moderator in the presence of fast neutrons and γ-rays.

(vi) Neutron beam experiments.

The importance of different features of DIDO's behaviour in relation to each of these categories will be indicated below.

Neutron fates in DIDO

The predictions of performance of DIDO, as of any nuclear reactor, depended very much on knowing the fates of neutrons born in the reactor under different conditions of loading. The usual approach to this problem is to apply diffusion theory to calculate the relative thermal neutron fluxes in the reactor core components (in DIDO, these are heavy water, aluminium and U235). The theoretical predictions were in fact, confirmed by experimental observations. The predicted value of k_∞ for this lattice can therefore be taken as known to the accuracy of its components,

i.e. $k_\infty = \eta\, p\, f\, \epsilon$, where

η for U235 can be assumed known to ± 1 per cent.,

p for 46 per cent. U235 has been calculated to be greater than 0.97 and Hicks (1957) suggests it is very nearly unity,

f, using experimental flux distributions and the known lattice dimensions and uranium content of aluminium, must be known to ±1 per cent.,

ϵ, with so little U238 present, will be 1.00.

Consequently $k_\infty = 2.07 \times 0.99 \times 0.91 \times 1.00 = 1.88$, with a maximum uncertainty of about 3 per cent.

The second part of the problem of predicting neutron fates can be tackled in a variety of ways. It is the problem of predicting how many neutrons will leak from a core comprising a given number of fuel elements. The simplest approach is to use a two-group theory as described by Hill (1949). With a small system like DIDO, with a heavy water reflector on all sides, this technique would be expected to overestimate leakage, because Hill's technique neglects the annular reflector in the corners. On the other hand, the slowing down distribution of neutrons in heavy water cannot be represented by a single characteristic length, and the assumption that it can might be expected to lead to an underestimate of neutron leakage in the fast group. However, Spinney (private communication) has used a four-group method, deriving his group constants by a three-group fit to the experimental slowing down distribution in heavy water, to show that the error is small.

The most satisfactory theoretical method for this part of the problem is therefore considered to be that set out by Hassett (1956) for use with

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the Ferranti Mark I computor. However, the value of K_∞ deduced by combining the experimental results in DIMPLE with calculations using Hassett's program was 1.609. By varying the number of fuel elements with top reflector height to make a series of critical assemblies Hicks (1957a) has shown that the Hassett type calculation yields the same value K_∞ over a wide range of core sizes.

At least half of the discrepancy in K_∞ remains unexplained.

Neutron fates have been dealt with at some length because any experimental result obtained needs correlation with some theoretical method so that accuracy of prediction can improve. First of the basic requirements are the neutron flux distributions, required to predict product yield in an experimental hole, or the heat output from a fuel specimen.

Neutron flux distributions

Experimentally, flux measurements can be made in DIDO alongside each fuel element and in experimental holes. Some additional measurements have been made in the DIMPLE assemblies, where access is easier. However, the latter are somewhat idealised because DIDO itself has a fixed number of fuel elements and suffers perturbations by experiments and control arms. While the former will be constant between shutdowns, the latter will cause major changes in the course of a run. Figure 1 shows examples of operational flux distributions.

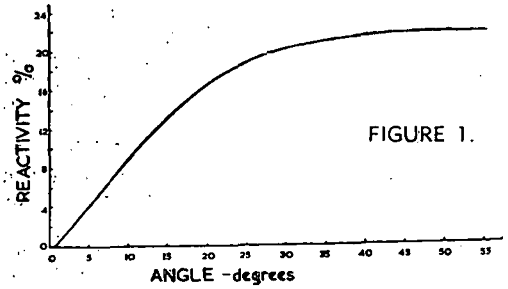

FIGURE 1.

The flux perturbations caused by experimental equipment can be of much more concern in the early stages of reactor utilisation. Again, it is usual to base flux depression calculations on diffusion theory. The complex geometry of most experiments and multiplicity of regions make the accuracy of such calculations doubtful. Because of this a number of experimental flux measurements in simulated irradiation rigs were made in preparation for experiments in DIDO. Hicks (1957b) has developed an electrical analogy to assist in flux depression calculations and has checked the method by comparing with experimental results for some typical assemblies.

A typical example of a complex rig proposed for DIDO is a part of a high-pressure water-cooled loop containing uranium fuel in a 1.7in. diameter steel tube of 0.1in. wall thickness. It is instructive to examine the contribution to the overall flux depression of the various components of the loop and this has been done by Smith and Constantine (1958).

The thermal flux in the reflector at the position chosen for the loop when DIDO is operating with a maximum flux in the fuel of 10^{14}n/sq. cm./sec. is 5.7 x 10^{13}. When the 6in. diameter thimble of 0.25in. wall-thickness is inserted the peak flux in the thimble is 4.1 x 10^{13}. This depression is partly an effect of absorption in the thimble walls, partly an effect of neutrons streaming up the thimble. When the pressure tube with the coolant is inserted the flux becomes 3 x 10^{13}. The fuel itself introduces a depression to 0.7 x 10^{13}. Without the coolant water the flux would rise to 1.9 x 10^{13}.

The problem of the effect of such a rig on the flux in neighbouring holes is one that is being tackled but is not yet solved.

Absolute measurement of flux

The design of the DIDO reactor envisages two limiting operating conditions. One is the overall limit of 10MW heat removal. The other is the onset of surface boiling in the fuel elements. This latter condition is difficult to check because of practical difficulties in fuel plate temperature measurements. It is therefore guarded against by relying on heat transfer calculation, coolant flow measurement and temperature rise. Knowledge of the peak absolute neutron flux in the fuel is desirable in establishing the required operating condition. The position of the peak is located by scanning the beta-activity produced in nickel wires which have been exposed in the flux measuring tubes attached to the fuel elements. The absolute calibration of this peak flux close to 10^{14}n/sq. cm./sec. is difficult, so the method which has been employed on DIDO is outlined. The difficulty stems from the high activities acquired by the usual standardising detectors in this flux.

A pellet of sodium carbonate of known weight is exposed with some silica rods of standard form in a flux of about 10^{12}n/sq. cm./sec. The sodium carbonate is then dissolved in water and the absolute activity of an aliquot is measured in a standardised equipment. From the known sodium activation cross-section it is then possible to deduce the flux in which the silica rods were exposed. The activity of these is measured in a quartz-fibre electroscope of standard pattern and so similar rods exposed at 10^{14}n/sq. cm./sec. can be compared with them. The factor of 10^2 in flux is bridged by electroscope range. Silica is used for the regular calibrated detector because it is readily available in pure form, and silicon has a low cross-section and a convenient half-life. Alternative methods are being explored, the direct use of cobalt with subsequent coincidence counting being an attractive one.

Reactivity effects

To operate a reactor safely it is essential to have an up-to-date record of reactivity changes in that reactor. Not the least of the problems of achieving this is the setting up of a scale of reactivity against which to make measurements.

A method of calculation of reactivity changes which gives results in reasonable agreement with the empirical scale is essential.

This problem, too, was tackled in DIMPLE by Hicks (1957a) and his results have since been extensively applied by the physicists working with DIDO. Ultimately, the safety of the reactor depends on the rate of rise of power resulting from a reactivity change. It is reasonable therefore, to base the scale of reactivity on the usual "in hour formula" relating excess reactivity and rise-time. Actual experiments using rise-time measurements are in practice limited in scope and the range of measurement has to be extended by uniform poisoning of the DIDO core. By combination or rise-time measurement over a limited range and uniform core-poisoning the DIDO control-arms have been calibrated to give a more useful reactivity scale (Figure 2).

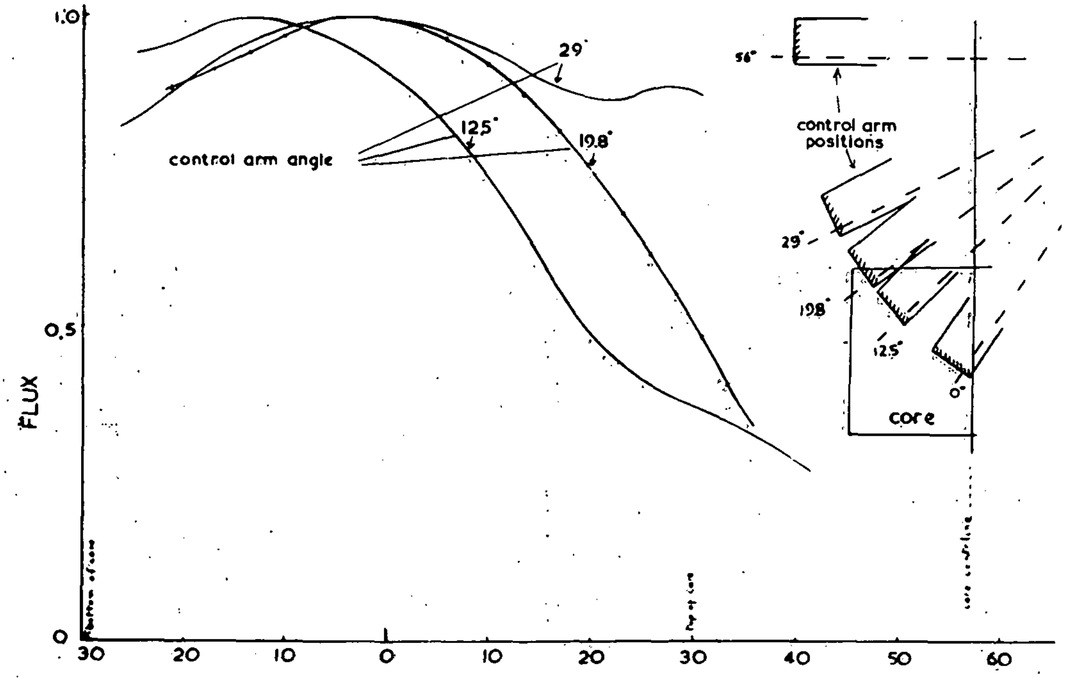

FIGURE 2 DISTANCE FROM CORE CENTRE – cms

With this calibration, measurements have been made of reactivity changes during normal operation. These come under the headings of:

(i) Temperature effects

(ii) Xe135 growth

(iii) Long-term fission product poison growth

(iv) U235 burn-up effect.

For conditions other than full power operation the reactivity changes of importance are:

(v) Xe135 build-up after shut-down

(vi) The effect of voids in fuel elements created by a transient causing boiling and some heavy water expulsion.

Temperature effects

The change in reactivity when the whole of the heavy water in the reactor tank is raised from 7°C to 50°C has been observed to be —.036 per cent./°C and —.038 per cent./°C for two different conditions of the reactor-core. The important difference between the two cases is the position of control arms (12.5° and 25° respectively). Calculations have been made by Wade (1958) of the reactivity change expected when the heavy water density and neutron temperature are changed appropriate to this temperature change. To simulate the two experimental states of the reactor spherical cores of two sizes were assumed, the critical condition being satisfied by assuming different amounts of uniform poisoning in the two cases. Satisfactory agreement is not obtained, and so both the experiments and calculations are being re-examined.

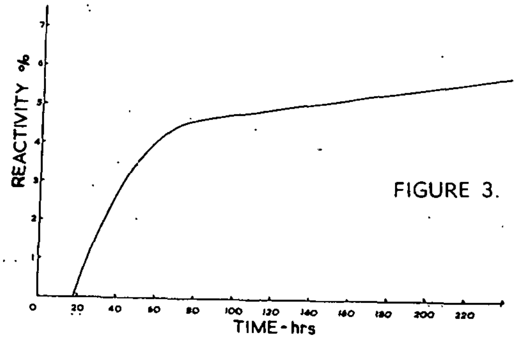

FIGURE 3.

Xenon 135 growth

Figure 3 shows the reactivity change which can be attributed to the growth of Xe135. Relating these observations to theoretical expectation is difficult, and changes in neutron distribution during the run must be taken into account.

Long-term changes

When DIDO was started with a fresh fuel charge and very little experimental loading, the fall in reactivity over 20 days at full flux was observed. This must be explained in terms of build-up of poisons and burn-up of U235, but again the change in flux distribution in the course of the run makes interpretation complicated initially. Reasonable agreement with calculation was obtained over the last stages, when the control arms produced only small perturbations. Figure 3 gives the observed results.

Void coefficient

This was measured in the DIMPLE mock-up experiments and is reported by Hicks (1957a) as a k-loss of 7 per cent. for complete loss of coolant from a 25-element core, 1.7 per cent. for loss of the upper half of the coolant from a 15-element core, and 0.9 per cent. for loss of the upper quarter of a 13-element core. Interpretation of these results is not yet compelte.

Tritium build-up

Whenever a leak occurs or equipment in DIDO is changed, heavy water escapes into the sealed building. Because tritium is built up steadily in the heavy water and presents a serious breathing hazard, a check must be kept on its growth. Accurate checks have not been made, but the tritium level has increased as expected, and yield is not inconsistent with previously published cross-sections. The largest uncertainty is in the detail of the flux distribution in the reactor. The rate of build-up is roughly 10^{-8} curies/cc/MW hour.

Heating effects

The rise in flux from 10^{12} in BEPO to 10^{14} in DIDO has brought with it a number of problems hitherto of no significance. One of these is the heating effect of neutrons, gamma-rays and beta-rays on non-fissile materials used in experimental equipment. In BEPO, these were of the order of 10 milliwatts per gram of material, and this amount was easily dissipated in the pile without intentional cooling. In DIDO, the heating can be 1 watt per gram of material, and this generated inside a series of concentric tubes, such as arises in all rigs in DIDO, leads to temperatures as high as 500°C. Most rigs are therefore cooled by an experimental cooling circuit using ordinary water, and this leads to complication and sometimes loss of neutrons.

Experimental observations have been made by Anderson (1957) of the heating in various materials in DIDO. These observations were carried out while the reactor was at 1 MW. The table gives the figures scaled to maximum flux operation.

TABLE 1: HEAT DEPOSITION IN SOME MATERIALS IN THE DIDO REACTOR.*

	At horizontal central plane of hole number—		
Material	2V8	4V5	4VGR1
Graphite	0.6	0.08	0.012
Polythene	0.8	0.13	0.02
Zirconium	0.6	0.08	0.01
Aluminium	0.6	0.11	0.014

* Units are watts per gram of material with Dido operating with a peak flux in the uranium of 10^{14}n/sq. cm./sec.

Operational problems

Hitherto, the physics information gained from operation of DIDO has been emphasised. There have, of course, been many other lessons learned during the first year of operation. Some of these have led to modifications in reactor design, some to the development of special operating techniques. In general, these do not fall under particular headings, and so they will be mentioned in sequence.

One of the earliest difficulties apparent was a fluctuation in power of about 5 per cent. peak to peak when the control arms were in a particular part of their path. This was caused by flutter of the arms when directly in the path of coolant leaving the fuel elements. It fell by a factor ten in magnitude when the control arms were withdrawn beyond the reach of the main coolant flow. By strengthening the arms and reducing clearances, the difficulty has been minimised. As experimental equipment builds up in the reactor, control arms spend less and less of their time in the range of flutter.

The tritium hazard has been mentioned. Other activity problems have been the build-up of highly active dust in experimental thimbles. This appears to be from the steel of the thimble plugs, and is quite long-lived so is presumed to be Fe59 or perhaps cobalt impurity. No satisfactory solution has yet been found for this problem.

The build-up of A41 activity in thimbles is also a nuisance. Carbon dioxide purging has always been available to remove this hazard, and can in fact be effective. The standard to which it was done initially proved inadequate, and tightness of seals has had to be improved in many cases.

Fuel problems

With the experience gained with DIDO, there has been a steady evolution of fuel element design. The critical size discrepancy led immediately to a proposal to increase the U235 content of the element, having in mind an eventual increase of higher uranium isotopes (U236, U238) as recycled uranium came into use.

The initial design was known to distort at a 50 per cent. overpressure above normal conditions. Several elements were found distorted on removal from the reactor and this is believed to have been caused by:

(i) the use of three pumps for short periods while changing over from one pump to another,

(ii) pulsations in the coolant circuit noted occasionally by the operators, and

(iii) an increase in pressure drop in the fuel elements due to a change in design of the outlet at the top.

These possible causes have now been eliminated and the fuel elements have been strengthened.

The charge into and discharge of fuel from DIDO has so far proceeded smoothly, and experience shows it possible to change 25 fuel elements in 18 hours.

Future trends in utilisation

Several principles have emerged from this year's experience with DIDO. First is the need to conserve space both in and around the reactor. Inside the reactor, all users must plan their experiments on the smallest possible scale to increase available flux. Outside the reactor, floor-space is valuable, and any equipment which can be outside the reactor shell and make necessary contact with the inside by means of safe seals to the shell should be so placed.

To increase space in the reactor and provide fast-neutron irradiations a new fuel element is being developed. This element contains the same amount of fuel as the others but is arranged in an annulus so that a 2in. diameter experimental tube is available in its centre. This arrangement is much to be preferred to the use of hollow cylinder convertors cooled by the experimental ordinary water circuit. This same principle of designing experiments to dispose of their heat to the heavy water rather than use the experimental circuit is urged on all rig designers. It allows greater neutron fluxes to be obtained, and considerably simplifies the safety circuitry.

Two other important requirements in rig design have also emerged. One is the exploitation of gamma-heating in those cases where high temperatures are required and electrical or other heating would add complication and use space. The other which can be coupled with the first is some degree of flexibility in heat removal. It is often required to produce a particular temperature in a specimen, but it is difficult to predict the actual temperature with sufficient accuracy. In such cases, it is highly desirable to be able to set up and adjust cooling conditions after insertion in the pile to take up errors in predicted performance.

Ensuring safe operation

It has always been necessary to design, build, and operate a nuclear reactor before a complete appreciation of the safety problems is possible. With the build-up of experience with many different types of reactor, it becomes possible to consider these problems at earlier stages. A committee has been set up to ensure a careful check on the safety of the DIDO design, to get the maximum amount of relevant information out of its operation, and to ensure that experimental equipment is safely designed. The committee includes experts in each of the fields necessary.

The responsibility for the safe operation of DIDO rests with the Chief Pile Operator. He sets out his instructions to his operators in standing orders, and he accepts or rejects experiments for DIDO, according to whether he considers them safe or not. The safety committee has been set up to assist the Chief Pile Operator by scrutinising his standing orders and, from detailed knowledge of the reactor, advising on their adequacy. In addition, the safety committee scrutinises every experimental proposal in detail and gives him advice on their suitability. In this way the Chief Pile Operator never has to take a decision on accepting or rejecting an experiment without the best advice available.

To make this system work, the prospective user of DIDO is forced to think out his experiment to the end in considerable detail. In addition to satisfying the safety requirements this saves money otherwise spent on unsafe or inadequate equipment. It also ensures that experience is fed back to a central organisation, where it can be used to improve design and operating technique.

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The Control and Instrumentation of the Reactor HIFAR

By G. Page*

A brief description of the reacting core of HIFAR is given, together with some data on the reactivity balance. The principles of the coarse, fine and safety control systems are outlined. Mention is made of the various installed thermal neutron flux measuring channels. The electrical safety and interlock circuits and the ways in which they influence the operation of the reactor are discussed. An outline is given of the other associated instrumentation such as health monitors and general industrial instruments.

Introduction

HIFAR is a high flux thermal reactor, moderated and cooled with heavy water. The design and construction of the reactor is described elsewhere by Roberts (1958).

It uses 2.5 kgm. of highly enriched uranium 235 as fuel, and is designed to run at a maximum heat power of 10 megawatts, when the maximum thermal neutron flux is 10^{14} neutrons per sq. cm. per sec. The reacting core, approximately cylindrical in shape, is located centrally in an aluminium tank containing the heavy water reflector. There are 25 vertical fuel elements in a 4, 6, 5, 6, 4 array arranged on a 15 cm. lattice pitch. The maximum burn-up of the fuel is approximately 20 per cent., the average being 10 per cent. The average rating of the fuel is 4 kW. per gm. The minimum number of fuel elements for divergency is 11 (see Watson-Munro (1958)).

The excess reactivity of the cold unpoisoned, undepleted reactor containing only horizontal experimental facilities and 25 fuel elements is approximately 15 per cent. The reactivity absorbed by the experimental facilities is approximately 4.5 per cent. and is made up of absorption in the vertical thimbles in the heavy water, including voids (approx. 3.3 per cent. total), that in the horizontal thimbles in the heavy water, including voids (1.0 per cent. total), that in the vertical and horizontal holes in the graphite, (0.2 per cent. total), giving an overall total of approx. 4.5 per cent.

If the average temperature rise of the heavy water is 30° C., this will account for the absorption of approximately 1.0 per cent. of reactivity. Long-lived fission product poisons (mostly Sm 149) absorb approximately 1.2 per cent. of reactivity at equilibrium. Xenon poisoning at equilibrium is approximately 3.6 per cent. of reactivity, giving a total of approximately 5.8 per cent. of reactivity absorption.

For more information on the reactor physics of HIFAR, see Hicks (1957).

The control system

The control system was purchased from H. M. Hobson Ltd., of Wolverhampton, United Kingdom.

Because of mechanical design problems associated with water and gas rotating seals in either the walls or bottom of the reactor aluminium tank, it was decided to instal and operate the neutron absorbing control system through the radiation shielding plug in the top of the aluminium tank. The general layout of the top of this plug is shown in Figure 1.

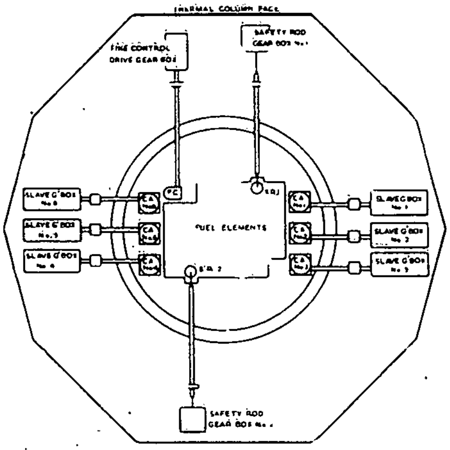

FIGURE 1.

The control system is divided into four parts:

(i) Combined coarse control and shut-down system.
(ii) Fine control system.
(iii) Safety system.
(iv) Partial dumping of the heavy water reflector.

The coarse system

The combined shut-off and coarse control system consists of a total of six signal arm type cadmium absorbers (see Figures 1 and 2). These are placed three on opposite sides of the reactor core in such a way that four of the arms move in the spaces between rows of fuel elements, the other two being outside, but close to the core. The arms are moved from the horizontal down to an angle of 56° from the horizontal. In this latter position a total of 11,500 sq. cm. of cadmium is inserted into the reactor core, causing an absorption of approximately 25 per cent. of reactivity. Because the maximum excess reactivity of the core is about 15 per cent., a considerable safety factor has been provided.

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* Australian Atomic Energy Commission Research Establishment. Manuscript received March 17, 1958.

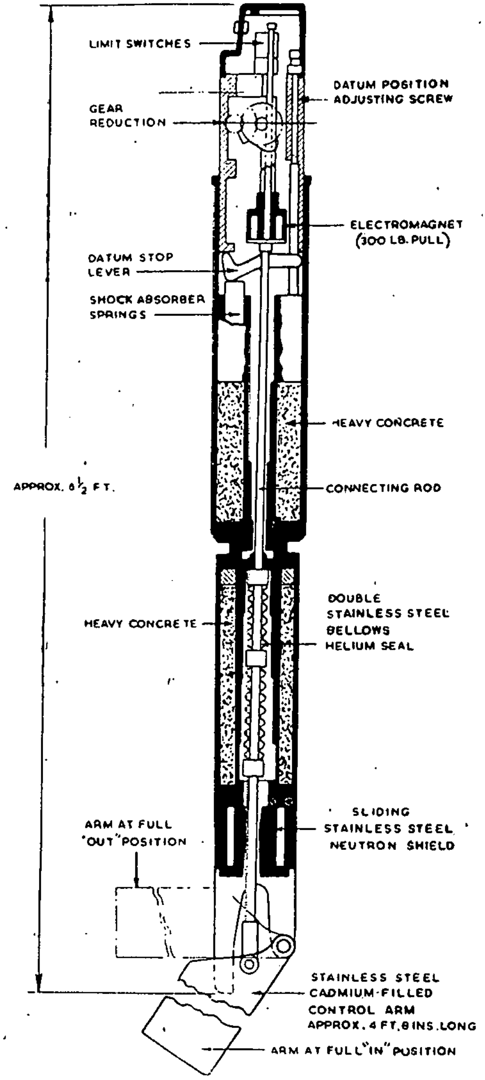


FIGURE 2.

The arms do not need any special cooling, because they are all immersed in the circulating heavy water.

The six control arms are mechanically coupled to six slave gearboxes, which are driven by a selsyn synchronous link transmission system from a single master gearbox in the control room (see Figure 3). "Slow-in" or "slow-out" electric motors move the six control arms through their full 56° travel, in or out respectively, in 25 minutes. An additional "fast-in" motor is provided to drive in the arms in five minutes.

In the case of an emergency, all the control arms can be simultaneously released electromagnetically to drop into the core under gravity in approximately 0.8 seconds. Under these conditions, ring springs are used to absorb the kinetic energy of the arms.

The whole coarse control system is designed with sufficient accuracy so that the position of each arm is repeatable to one minute of arc. Coarse and fine indication of the position of the arms is provided by a magslip system with a sensitivity of approximately 1/100 of a degree of arc of the arms.

A coincidence magslip system is also provided,

(i) to give meter indication of misalignment of up to two minutes of arc between control arms,

(ii) to shut down the reactor if the misalignment exceeds one minute of arc between any arms.

Fine control system

Fine control of the power level of the reactor is achieved with a moveable vertical rod containing 205 sq. cm. of cadmium (see Figure 4) and located just outside the reacting core (see Figure 1).

The rod can be driven in and out at a continuously variable speed through a gearbox by a special 1/10 h.p. two phase servomoter (see Figure 5). A D.C. permanent magnet tachogenerator on the same shaft as the motor provides feedback information for the speed control equipment, which consists of a combined electronic and magnetic amplifier. Provision has been made in the speed control system for the later addition of equipment for the automatic control of the reactor power level by means of fine control rod position.

At maximum speed, 61 cm. travel of the rod is covered in 20 seconds. By careful design the friction and inertia of the system has been kept as low as possible, in order to allow the rod to be moved fast enough to compensate for reactor surges.

The maximum reactivity absorbed by the rod is 0.17 per cent.

Since the rod moves in a "thimble," light water cooling is necessary to remove several kilowatts of heat resulting from neutron and gamma absorption.

Safety rod system

To safeguard against an unforeseen gain in reactivity, particularly during a reactor shutdown, two vertical safety rods are provided (see Figure 6). They are located just outside the reacting core (see Figure 1), and each is capable of inserting 800 sq. cm. of cadmium into the reactor core, corresponding to the absorption of 1 per cent. of reactivity, i.e., a total of 2 per cent. of reactivity for both rods.

Each rod is connected by means of a stainless steel cable to a gearbox containing an electromagnet which, on release, allows the rod to fall into the reactor under gravity in 1½ seconds. The kinetic energy of the rod is absorbed by a cam and spring system without "snagging" of the cable.

Even though the safety rods move in "thimbles" in the reactor, no special cooling is required because the rods are normally right out of the reactor or right in the "shut-down" reactor. The heat generated in each rod is no more than 15 watts.

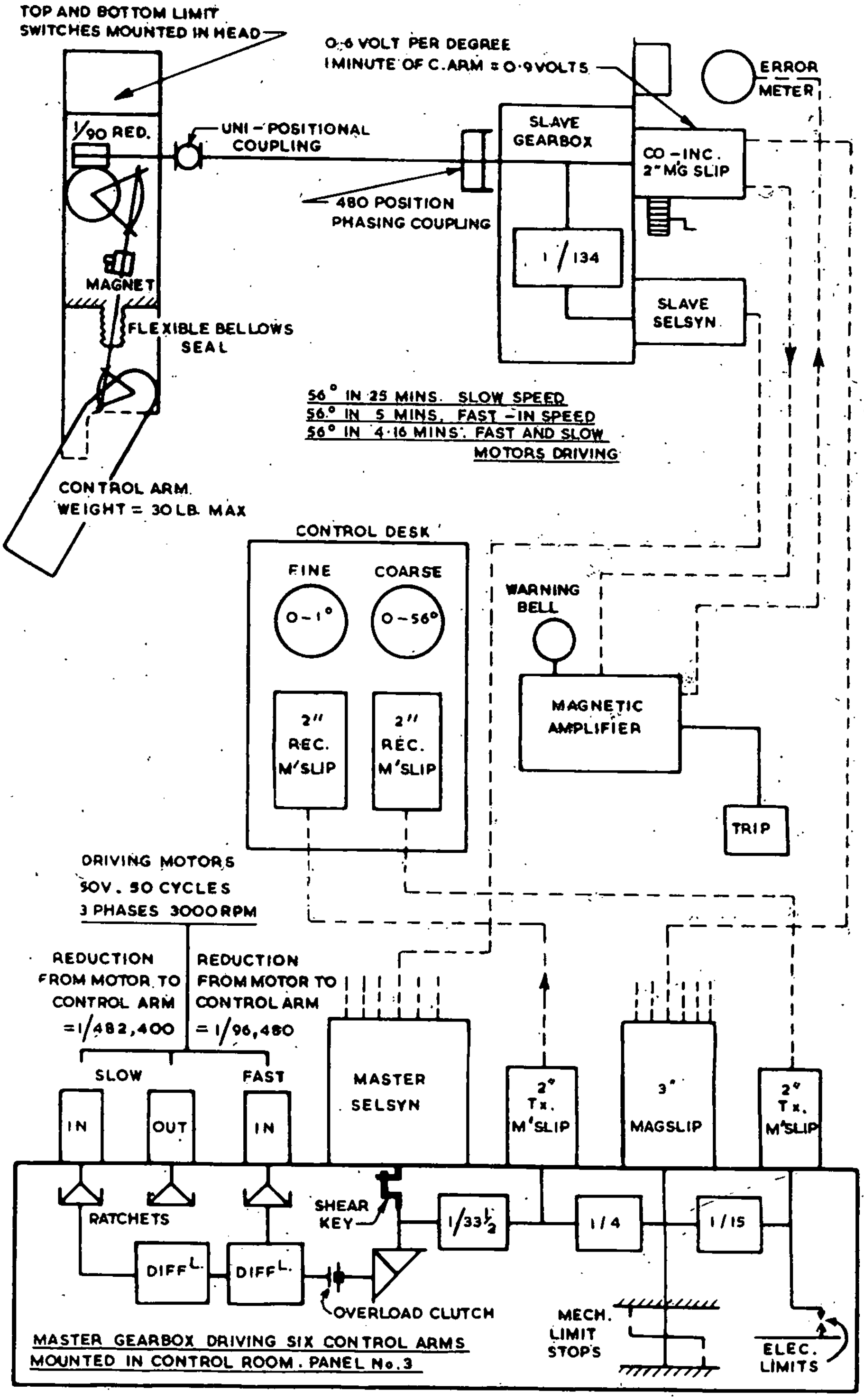

TOP AND BOTTOM LIMIT SWITCHES MOUNTED IN HEAD
0.6 VOLT PER DEGREE
1 MINUTE OF C.ARM = 0.9 VOLTS
ERROR METER
1/90 RED.
UNI-POSITIONAL COUPLING
SLAVE GEARBOX
CO-INC. 2" MG SLIP
480 POSITION PHASING COUPLING
MAGNET
1/134
SLAVE SELSYN
FLEXIBLE BELLOWS SEAL
56° IN 25 MINS. SLOW SPEED
56° IN 5 MINS. FAST-IN SPEED
56° IN 4.16 MINS. FAST AND SLOW MOTORS DRIVING
CONTROL ARM.
WEIGHT = 30 LB. MAX
CONTROL DESK
FINE
COARSE
0-1°
0-56°
WARNING BELL
2" REC. M'SLIP
2" REC. M'SLIP
MAGNETIC AMPLIFIER
TRIP
DRIVING MOTORS
50V. 50 CYCLES
3 PHASES 3000 RPM
REDUCTION FROM MOTOR TO CONTROL ARM =1/482,400
REDUCTION FROM MOTOR TO CONTROL ARM =1/96,480
SLOW
FAST
IN
OUT
IN
MASTER SELSYN
2" Tx. M'SLIP
3" MAGSLIP
2" Tx. M'SLIP
RATCHETS
SHEAR KEY
1/33½
1/4
1/15
DIFFL.
DIFFL.
OVERLOAD CLUTCH
MECH. LIMIT STOPS
ELEC. LIMITS
MASTER GEARBOX DRIVING SIX CONTROL ARMS MOUNTED IN CONTROL ROOM. PANEL No. 3

FIGURE 3.

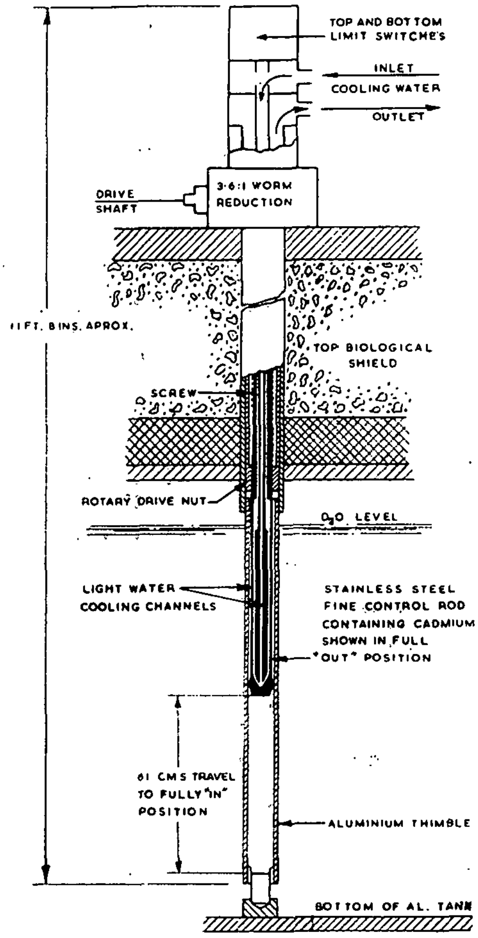


FIGURE 4.

The partial dump facility

A further emergency method of removing reactivity from the reactor is by reducing the thickness of the heavy water reflector above the fuel elements, thereby allowing neutrons to leak out of the reacting core.

A stainless steel tank is available for the "dumping" in 150 seconds of the top 2ft. of the heavy water in the reactor aluminium tank. This is equivalent to a loss of reactivity of about 3 per cent. at a rate of about 1 per cent. per minute.

A push button in the control room breaks the circuit to an electromagnet holding closed a gravity operated valve controlling the heavy water flow in a 3in. diameter pipe to the "dump" tank.

No more than 2ft of heavy water can be "dumped," because at no time must the heavy water level fall low enough to uncover any part of the actual fuel elements. This is because gas convection cooling of part of an "old" burnt-up fuel element is not sufficient to remove the fission product heat, so that serious damage could occur to the fuel elements.

Control summary

Absorption of the control systems:

Coarse and shutdown system	25%
Fine system	0.17%
Safety system	2%
Partial heavy water dumping	3%
Approximately	30%

Xenon poisoning

With all thermal reactors having fluxes of 10^{14} n/sq. cm./sec., and higher, the large thermal neutron capture cross-section of two of the fission products, xenon 135 and samarium 149, have always to be borne in mind. This is discussed in detail by Cox and Walker (1956), Glasstone (1956) and Glasstone and Edlund (1956).

In the case of HIFAR, at equilibrium the xenon 135 absorbs 3.6 per cent. of reactivity and the samarium 149, 0.86 per cent. When the reactor is shut down after prolonged operation at 10 MW the xenon 135 builds up as its parent, iodine 135 (half-life, 6.6 hr.) decays, thereby absorbing more and more reactivity until, about 10 hours after shut-down, a maximum reactivity of over 25 per cent. is absorbed. Because this is more negative reactivity than can be offset by all the fuel elements, it is impossible to start the reactor under these conditions. The xenon 135 concentration then decays away (half-life 9.4 hr.) until at about 36 hours after the shut-down it is possible to withdraw the coarse control arms and make available sufficient reactivity to enable the reactor to diverge again.

Flux measurement

Three pairs of holes penetrate the biological shield as far as the graphite reflector. They are located symmetrically on either side of the thermal column. Suitable holes have been left in the boral lining (Roberts, 1958), to allow a reasonable flux of thermal neutrons to be available for measurement.

Into these holes are inserted boron-containing ionisation chambers, types RC1 and RC2 as described by Abson and Wade (1956).

The RC1 chambers are filled with enriched boron trifluoride and have a thermal neutron sensitivity of 1.27 x 10^{-14} amps/n/sq.cm/sec. and a gamma-sensitivity of 9 x 10^{-12} amps/r/hr.

The RC2 chambers are coated with boron 10, and filled with hydrogen to a pressure of 15cm. of mercury and have a thermal neutron sensitivity of 1.7 x 10^{-15} amps/n/ sq.cm./sec., and a gamma-sensitivity of 3.3 x 10^{-13} amps/r/hr.

Each ionisation chamber can be moved approximately ± 12 cm., providing adjustment in flux of a factor of about three.

Reactor power error meter (E. K. Cole type 1462A)

The ionisation chamber is an RC2, which is placed in the top left hole where the neutron flux is between 1.7 and 5.2 x 10^{10}n/sq.cm./sec. at 10 MW power. A lead "muff" surrounds the chamber in order to reduce to a minimum any current due to gamma-radiation, particularly when the reactor is shut down.

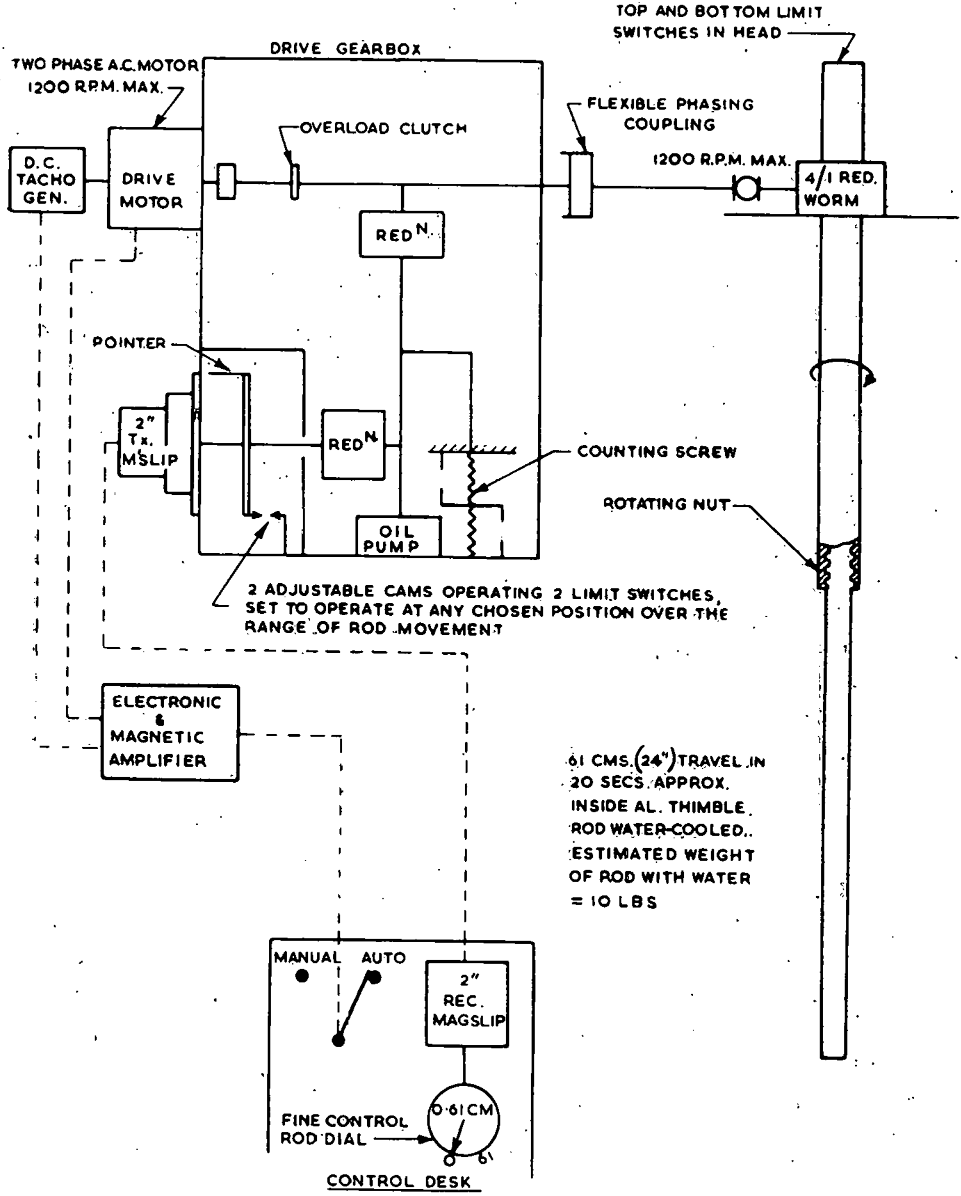


FIGURE 5.

In this instrument, effectively, the current from the ionisation chamber is backed off by the current from a battery. The former current is proportional to reactor power and the latter is the "demanded" power. The error between the two is displayed on a centre zero meter of high sensitivity. The demanded power is variable in 100 KW steps from 100 KW to 12 MW.

The stability of the centre zero meter is a measure of the stability of the operating power level of the reactor. A signal is available from this equipment for future use in the automatic control of the reactor power level.

Multi-range linear recording of nuclear power

The ionisation chamber is an RC2, which is placed in the top right hole where the neutron flux is between 1.7 and 5.2 x 10^{10}n/sq..cm./sec. at 10 MW power. A lead "muff" is used here also.

In this instrument, the D.C. ionisation current is fed into a special high impedance mechanical chopper followed by A.C. amplification with display on a fast linear pen recorder (2 seconds full-scale deflection). This is a system made by Geo. Kent Ltd., of Luton.

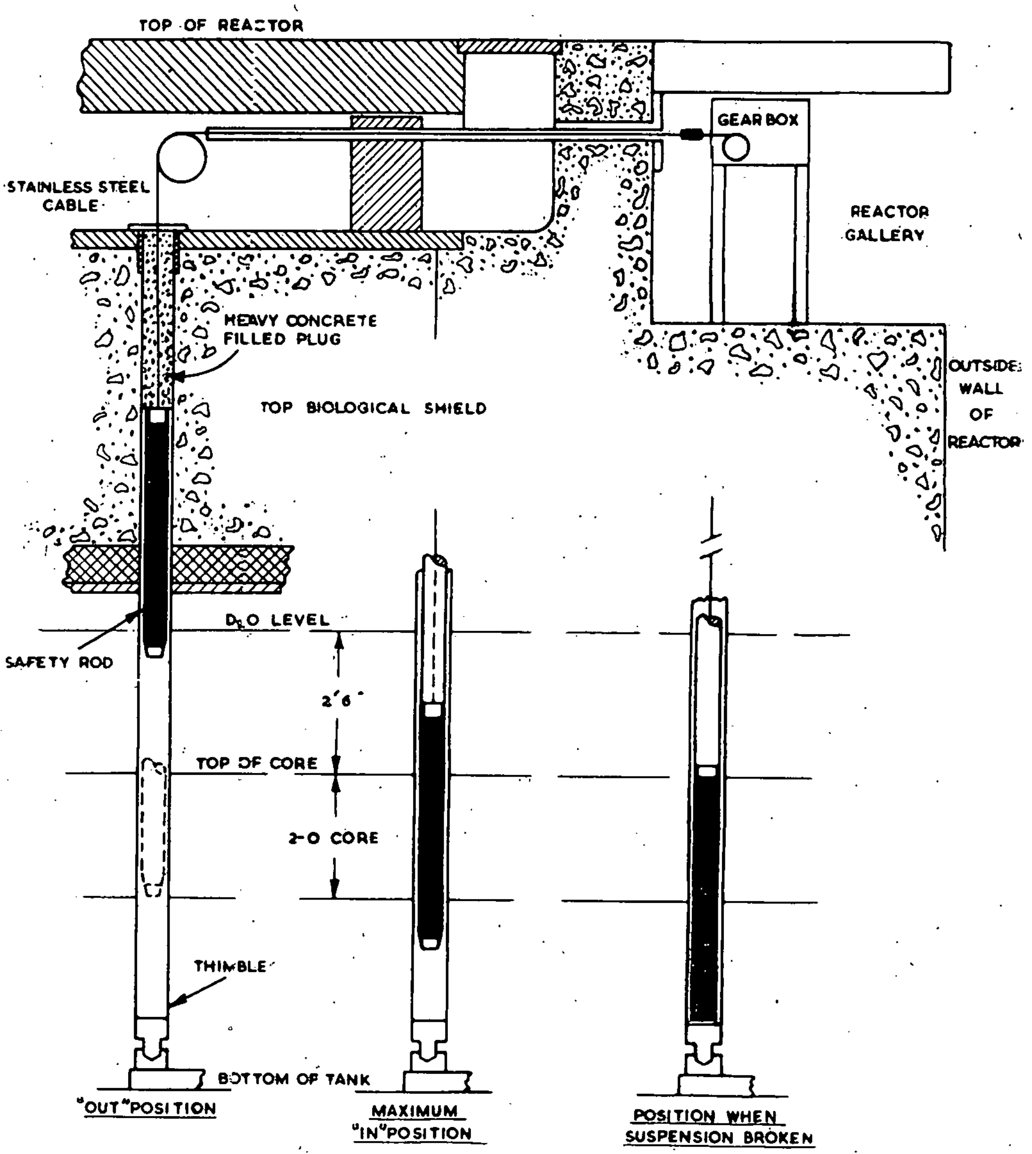


FIGURE 6.

Five ranges are provided for full scale deflection of 1.5 KW, 15 KW, 150 KW, 1.5 MW and 15 MW.

Log power and reactor period meter (E. K. Cole type 1513A)

The ionisation chamber is an RC2 which is placed in the middle left hole where the neutron flux is between 1.1 and 3.5 x10^{10}n/sq.cm./sec. at 10 MW power. A lead "muff" is also used here.

The D.C. ionisation current is fed into a special feedback D.C. amplifier with a logarithmic characteristic such that the output voltage is proportional to the logarithm of the input voltage over six decades. This is presented on a high speed pen recorder scaled from 10 W to 20 MW on a special logarithmic chart.

The log output of the amplifier is also fed into another special amplifier where it is electrically differentiated and amplified, thereby giving an output that is proportional to the reactor period, i.e., the time taken for the power of the reactor to increase or decrease by a factor $e = 2.718$.

This is presented on a meter scaled in reactor doubling time from —20 secs. through infinity to +5 seconds. Also included in the reactor period amplifier is a relay circuit which can be preset to control the reactor if the period is too short either on positive or negative periods. A signal is also available from the reactor period amplifier either to feed a recorder or to be used with other equipment for the automatic start-up of the reactor.

Single range linear measurement of nuclear power with mechanical power/time integration

The ionisation chamber is an RC1, which is placed in the bottom left hole where the neutron flux is between 1.8 & 6.3 x 10^9n/sq.cm./sec. at 10 MW power. A lead "muff" is not used here, as one is not interested in trying to accurately measure low nuclear power under shutdown conditions.

The D.C. ionisation current is fed into a special high impedance mechanical chopper followed by A.C. amplification and display on a fast linear pen recorder (two seconds full scale deflection) scaled 0 to 15MW. Mechanical power/time integration is presented on dials in megawatt hours.

High flux reactor shut-down amplifiers (Isotope Development Ltd. Type 1461A)

Four RC1 ionisation chambers are placed in the bottom right hole where the neutron flux is between 1.8 and 6.3 x 10^9n/sq.cm./sec. at 10 MW power. Each is independently adjustable over about ± 12 cm.

The D.C. ionisation current is fed into a D.C. feedback amplifier, which has been carefully designed to incorporate "fail safe" features. This amplifier operates relays which shut down the reactor when the nuclear power exceeds a pre-set value. It also operates a meter scaled in percentage of trip level at which the reactor is operating.

This general type of reactor instrumentation has been discussed by Gillespie (1956).

Safety of the reactor

More than 200 relays are employed in circuits to ensure, as far as humanly possible, that the reactor is operated in a safe manner. In almost all cases, fault and warning conditions open-circuit contacts and de-energise relay coils in "fail to safety" arrangements.

Mention will first be made of warnings and fault conditions.

Some forty "WARNING" conditions are indicated by lights in the control room. To prevent a later automatic reactor shutdown, it is necessary to take immediate corrective action. Typical of these conditions are:

(i) Reactor helium gasholder less than 10 per cent. full.
(ii) Reactor helium gasholder greater than 90 per cent. full.
(iii) Instrument compressed air supply less than 30 p.s.i.g.
(iv) Too high gamma-activity in the heavy water, etc.

If the warnings are ignored, sooner or later a condition will develop that will cause an automatic reactor shut-down. This is brought about in the following ways:

Control reversal

The coarse control arms are driven fast (13.4 minutes of arc/sec.) into the reactor. If the fault is cleared, then it is possible to reverse the motion of the control arms and move them out again to the previous operating position. The nine conditions that initiate a control reversal are:

(i) Control arms all at top limits, i.e., horizontal;
(ii) heavy water plant room doors open;
(iii) reactor helium gasholder less than 5 per cent. of full capacity;
(iv) no heavy water flow down overflow pipe;
(v) fine control rod secondary coolant flow less than 75 per cent. of normal.
(vi) fine control rod primary coolant temperature greater than 80°C.;
(vii) only two (out of three) cooling water pumps running;
(viii) heavy water flow through reactor less than 90 per cent. of normal;
(ix) only one (out of two) heavy water pumps running.

Trip

As was mentioned above, the coarse control arms are lifted through electromagnets. When any of the 16 conditions listed below occurs, then these electromagnets are de-energised, allowing the coarse control arms to drop into the reactor under gravity in about 0.8 seconds from the horizontal position, and proportionally less time from other angles. At the same time, the fine control rod is driven fast into the reactor core.

(i) Greater than one minute of arc misalignment between the six combined coarse control and shut-off arms.
(ii) No. 1 and No. 2 reactor helium gasholder protection valves closed.
(iii) Fine control rod primary coolant flow less than 75 per cent. of normal.
(iv) Only one (out of three) cooling water pumps running.
(v) Heavy water outlet temperature greater than 58°C.
(vi) Heavy water reactor inlet/outlet temperature difference greater than 8°C
(vii) Heavy water flow less than 30 per cent. of normal.
(viii) No heavy water pumps running (out of two).
(ix) Failure of mains supply to reactor building.
(x) Two out of four flux trip channels high.
(xi) Dump valve open.
(xii) Safety rods No. 1 and No 2. not up.
(xiii) Reactor power doubling time less than eight seconds.
(xiv) Emergency reactor shutdown buttons pressed.
(xv) Low head of cooling water at cooling towers (indicates a broken pipe).
(xvi) Low level of water in cooling tower basin.

Even if the fault is cleared immediately after the reactor has been tripped, it is necessary to wait for the magnets to be driven down to pick up the control arms. Then it is possible to withdraw the coarse control arms from the core only at the slow rate of 56° in 25 minutes. Thus, a spurious trip can cause a delay of up to 30 minutes before the reactor is back to its original operating condition.

Complete shut-down

The reactor is tripped as above, but in addition the safety rods are dropped, the inlet and extract building ventilation fans are stopped and the inlet and outlet seal valves in the ventilation system are flooded.

The conditions that initiate this type of shut-down are:

(i) Reactor helium pressure greater than 12in. of water gauge.

(ii) Centre fuel element surface temperature greater than 98°C.

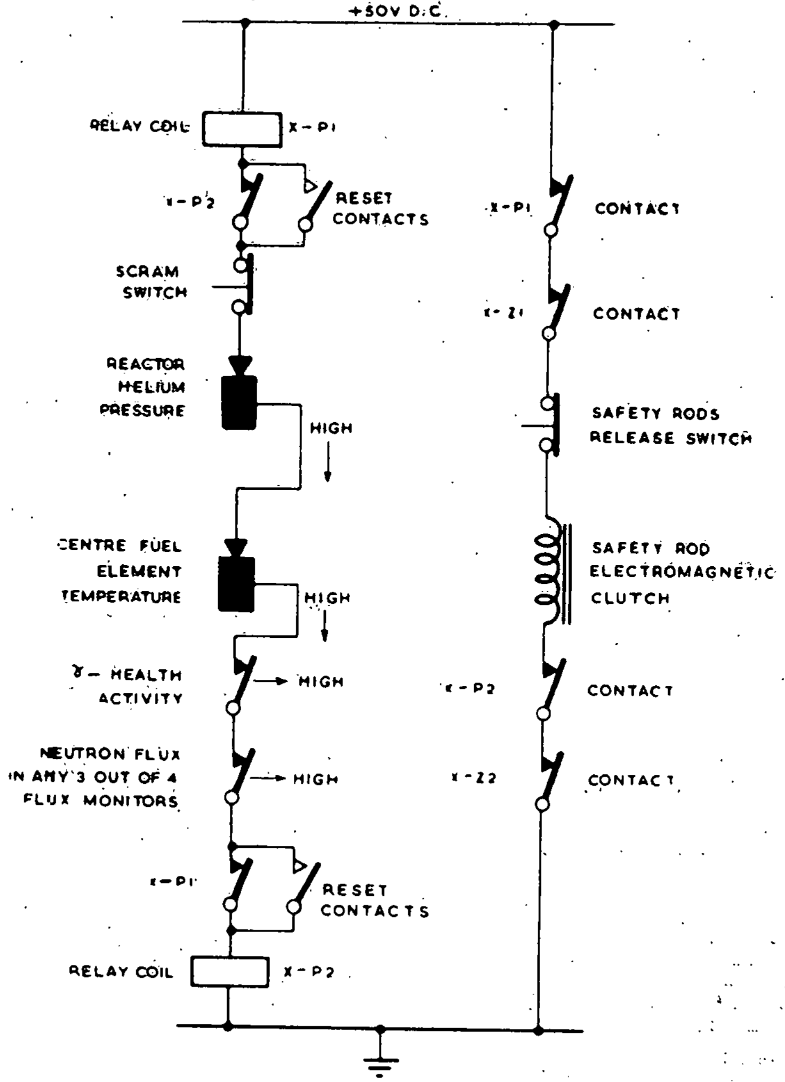
(iii) High gamma activity by control room.

(iv) High gamma activity in No. 1 or No. 2 extract filters in ventilation system.

(v) Three out of four flux trip channels high.

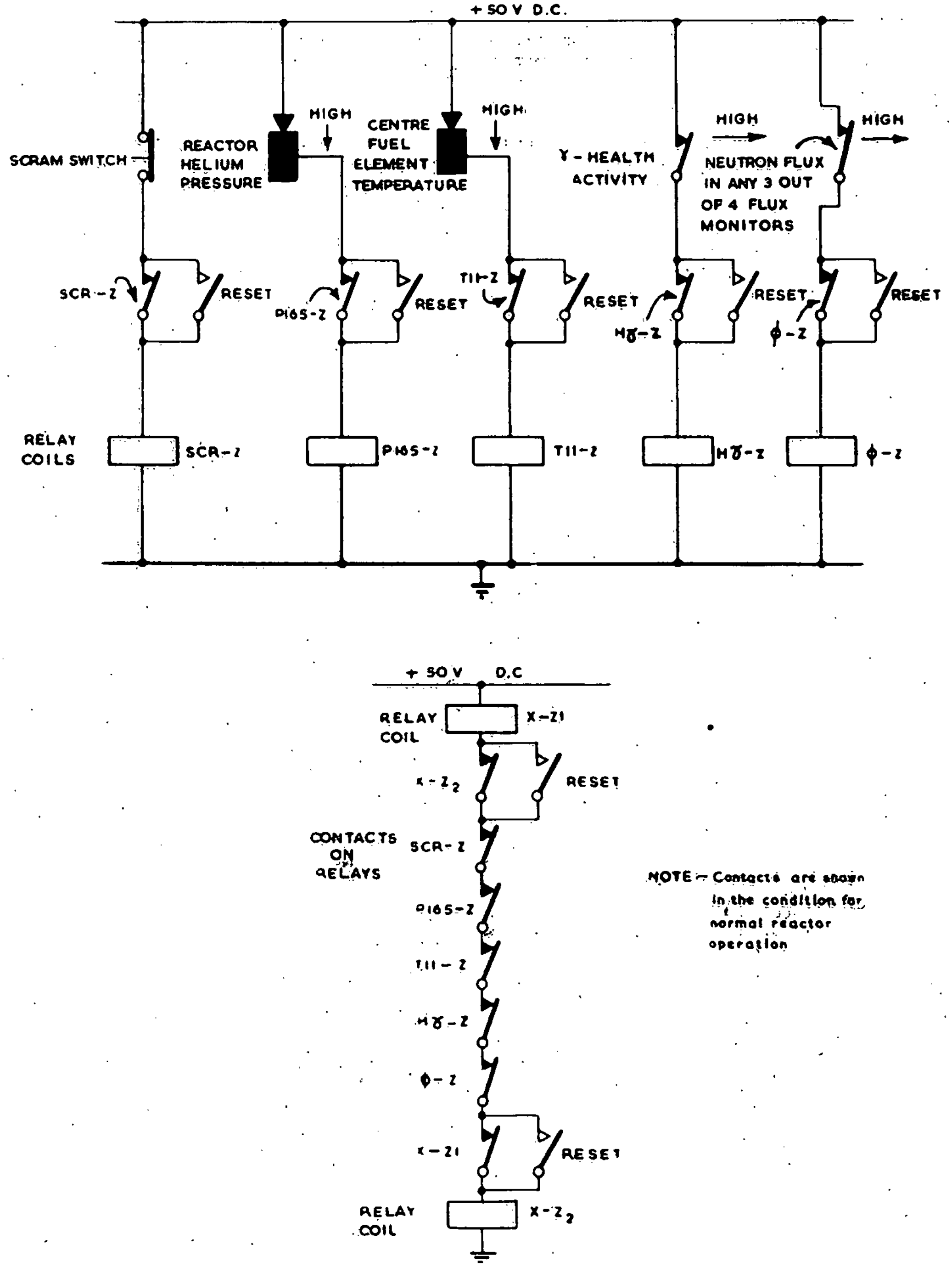
Scram

This is initiated by a push button in the control room, and also by one in the emergency control room. In addition to shutting down the reactor as for a complete shutdown, the scram button also sounds off eight sirens located in the reactor group of buildings. This is to warn people to leave the reactor building and follow the "scram" drill. The scram sirens can be temporarily stopped by a spring-loaded switch in the control room in order to allow the public address system to be used. The scram condition can be cancelled and reset only with a special key normally housed in the emergency control room.



NOTE — Contacts are shown in the condition for normal reactor operation

FIGURE 7.



NOTE – Contacts are shown in the condition for normal reactor operation

FIGURE 8.

Safety when shut down

When the reactor is shut down for maintenance or loading or unloading experiments, the safety rods are held out of the core and will be automatically dropped in if for any reason the shut down power should rise to 10 kW, or if a reactor power doubling time of less than eight seconds should occur.

Relay circuits

The basic principles of the relay circuitry are illustrated in Figures 7 and 8. (In this case it is a simplified version of the complete shutdown system.) The relay power supply is + 50-volt D.C., and most of the relays are G.P.O. Type 3000, made to inter-service specifications. The circuits are all designed to incorporate failure to safety features. For each condition that initiates an automatic shutdown of the reactor there are provided at least two independent change-over contacts that are incorporated in the "primary" and "secondary" guard circuits respectively.

Referring to Figure 7, a typical "primary guard" circuit, it will be seen that it is divided into two main sections. At the left there is the "monitoring" chain, and at the right the "operating" chain.

Consider the "monitoring" chain. The electrical contacts on the measuring instruments are all connected in series. It will be noted that the fault condition open-circuits the chain. At either end and in series with the chain there are the coils of the two monitoring relays X-P1 and X-P2. It is necessary to have both relays so that an earth fault part way down the chain will operate one or the other relay. Holding contacts from relays X-P1 and X-P2 are also inserted in series with the chain, so that restoration of a fault condition does not automatically complete the chain. To energise X-P1 and X-P2 relay coils, the reset contacts must be momentarily closed by pressing the "TRIP RESET" button in the control room.

Now consider the "operating" chain. To release any of the reactor neutron flux control mechanisms, it is necessary to de-energise the appropriate electromagnets. Here, one of the safety rods is illustrated. Contacts from both the primary and secondary guard chain monitoring relays, X-P1, X-Z1, X-P2, X-Z2, are connected in series with the safety rod electromagnet so that the opening of any of these contacts breaks the circuit.

Referring to Figure 8, a typical "secondary guard" circuit, it will be seen that this again is divided into two main sections. When the electrical contacts on the measuring instruments are closed (i.e., normal reactor operating conditions), each contact operates an individual relay through a relay holding contact with reset contacts. For instance, when the "SCRAM" button is pressed, SCR-X relay coil is de-energised and the SCR-Z holding contacts are open circuited so that on releasing the button, relay coil SCR-Z is not re-energised until the "TRIP RESET" button is pressed. Additional contacts on these relays are used to operate lights, bells, etc., in order to quickly localise faults.

One set of contacts from each relay is connected in series to form the secondary guard monitoring chain with monitoring relay coils X-Z1 and X-Z2 in a very similar way to the primary guard monitoring chain already described.

Health instrumentation

Permanently installed in suitable locations are 16 gamma health monitors and six fast neutron monitors.

Gamma-Monitors (Isotope Development Ltd. Type 1529A)

Each of these consists of two parts, an ionisation chamber and preamplifier, which can be mounted anywhere around the reactor, followed by a logarithmic amplifier which is mounted in or near the control room.

The ionisation chamber is made entirely of materials of low atomic number in an endeavour to make it an equivalent air-wall so that dose-rate readings are independent of energy over a wide range of gamma-ray energies. The output current is proportional to gamma-flux, the sensitivity being 6 x 10^{-12} amps for a dose rate of 10 mr./hr.

The ionisation current is fed into a D.C. feedback amplifier with a logarithmic characteristic to give a five decade indication of dose rate on a meter scaled approximately logarithmically from 0 to 10^5 mr/hr. The zero of the logarithmic scale is achieved by attaching to the wall of the ionisation chamber a weak gamma-source equivalent to a dose rate of 1mr/hr and calibrating the meter to give true readings of any further gamma-radiation.

An output is provided to drive a recorder and the output of 13 of the installed instruments is displayed on recorders in the control room.

Two relay trip circuits are provided. One is a low-level adjustable trip, usually set at about the international health tolerance level of 7.5 mr/hr. This gives only a warning in the control room if the gamma level is exceeded. The other trip is a high-level one, and can be set anywhere between 10 and 10^5 mr/hr. For certain locations, two-head units are installed alongside each other and this high-level trip is set to shut-down the reactor if the gamma level is high in both units at the same time.

Fast neutron monitors (E. K. Cole Ltd. Type 1463A)

Each of these consists of two parts, a proportional counter sensitive to fast neutrons and a preamplifier which can be mounted anywhere around the reactor, followed by a linear ratemeter, which is mounted near the control room.

The proportional counter is lined with polythene, a hydrogenous material with which the fast neutrons interact to produce energetic recoil protons which are detected in a methane/argon filled proportional counter. The detector responds to fast neutrons in the energy range 0.15 to 15 MeV, and the energy response is adjusted to be proportional within ± 20 per cent. to the international biological dose per neutron.

The ratemeter provides a stable EHT supply for the proportional counter. The ratemeter is a rather versatile instrument, and for this application the integrating time is set at 20 sec., the gain at 500 and the counting rate meter at 3 counts per sec. full scale. The discriminator is set at 30 V, which greatly reduces the gamma sensitivity of the equipment.

A relay trip circuit is incorporated, and can be preset to operate at any desired level, or on failure of certain parts of the circuit.

The approximate overall sensitivity of the fast neutron detector is:

Sensitivity to fast neutrons (0.15 to 15 MeV) = 0.9 counts/sec. for maximum permissible level.

Sensitivity to radium gammas = 0.1 counts/sec./r/hr.

Sensitivity to slow neutrons = 0.002 counts/sec. for maximum permissible level.

The slow neutron sensitivity can be reduced to zero by surrounding the counter with cadmium sheet, 1mm. thick.

Industrial instrumentation

This was supplied by Geo. Kent, of Luton, Eng.

The term industrial instrumentation, as used here, covers the measurement of temperature, fluid pressures, flows, levels, etc.

In both the heavy water and reactor helium circuits, all parts of instruments in contact with the fluids must be made of either approved types of stainless steels, e.g., 18/8/1, or high purity aluminium, with glass being permitted in one or two special cases. Another requirement is that the majority of both circuits must be pressure tight to withstand both a pressure of 30 p.s.i.g. without leakage, and a vacuum of 100 microns of mercury with negligible leakage. These restrictions alone limit the types of instrumentation that can be used.

Temperatures are measured either by Heracus type platinum resistance thermometers, or by the Rototherm type of instrument inserted into stainless steel pockets.

Pump pressures are indicated on Schaeffer type diaphragm gauges.

Pivoted stainless steel floats, magnetically operating contacts through non-magnetic stainless steel diaphragms, are used to provide liquid level information to automatically stop and start pumps, light warning lights, etc.

Pneumatically operated differential pressure transmitters (made by the Foxboro Co.), using the force-balance principle, are used in conjunction with suitable orifice plates or Dall tubes (improved type of short venturi tube) to measure liquid flows. These same transmitters are also used to measure heavy water levels in large tanks.

Gas flow in the reactor helium circuits is measured with Rotameter type instruments.

All the above instrumentation applies only to the heavy water and reactor helium circuits. The other liquid coolant circuits use demineralised water, except the main cooling tower circuit, which uses ordinary mains water. Hence the instrumentation is conventional.

Temperatures are measured either with Rototherm type thermometers or by mercury in steel thermometers.

Pump pressures are indicated on either diaphragm or Bourdon tube gauges.

Stainless steel bellows are used to operate electrical contacts to provide liquid level information.

Liquid flows are measured by orifice plates and differential bellows instruments, or by Rotameter type instruments.

Other special instrumentation

External heavy water leaks

All the stainless pipes containing heavy water are joined by flanges, usually tongued and grooved, with neoprene gaskets. Any heavy water leaks will occur from these flanges. It is necessary to detect these leaks immediately, both because of a possible tritium health hazard, and also because of the high cost of heavy water (about £150 a gallon).

Accordingly, some 450 flanges have electrical sensing elements fitted and these are connected in groups to a 100 channel detector unit.

The sensing element consists of a strip of filter paper wrapped around the clean edges of a mating pair of stainless steel flanges. The metal side of a strip of paper-backed aluminium foil is next wrapped around the filter paper. A wide neoprene band with "quick release" buckle is stretched over the whole to clamp it securely together and keep dirt out. Electrical contact is made on to the aluminium foil, and normally the resistance to ground (i.e., the flange) is of the order of at least 10^9 ohms.

If this resistance falls to 100,000 ohms or less because of a leak of heavy water wetting the filter paper, a simple radio valve operated relay is de-energised, thereby giving a warning of a leak. By a chart and a switch system the faulty flange can quickly be located from outside the heavy water plant room.

Leakage of heavy water into light water

The only place where this can occur is in one of the three heavy water/light water heat exchangers. It appears that the most sensitive method of detecting these leaks is by taking advantage of the short-lived nitrogen 16 activity formed in the heavy water when the reactor is running at full power. It is a fast neutron reaction O16 (n,p) N16, the nitrogen 16 having a half life of 7.3 sec. emitting a 10 MeV beta-particle and a 7 MeV gamma-ray.

Into a water by-pass line across each heat exchanger is inserted a sodium iodide crystal and scintillation head to detect the 7 MeV gamma-ray from any nitrogen 16 that will have been carried through a leak by the heavy water. To achieve maximum sensitivity, it is necessary to install lead shielding around the detectors, particularly because by the time the water reaches the detector, up to two half lives of the nitrogen 16 may have elapsed due to the flow velocities.

The head unit feeds into a linear ratemeter (E.K.-Cole Type 1463A) identical to that used with the fast neutron detectors mentioned above. Due to the high gamma-energy to be detected the discriminator level can be set up to reduce the background count rate by a considerable fraction.

Isotopic purity of the heavy water

To reduce any reactivity loss in the heavy water, it is necessary to continuously monitor the light water content. This is done by an infra-red absorption apparatus, the TRI-NON, manufactured by the Perkin-Elmer Corp., U.S.A.

The basic principles of the instrument as used for hydrocarbon gas analysis are described by Woodhull et al. (1954).

The infra-red radiation from a suitable filament is split into two parts and "chopped" at 13 cycles/sec. by a rotating shutter. Into one path is introduced a continuously flowing heavy water sample about 0.1mm. thick.

Because all the light water in the nearly pure heavy water is in the form of HDO, which has a distinct infra-red absorption band at a wavelength of about 2.8 microns, energy is absorbed in this path. Into the other path a wedge attenuator is driven by a servomechanism controlled by the detector. This is a chamber containing ammonia vapour, which also has an infra-red absorption band at about a wavelength of three microns. Energy from both paths falls alternately at 13 cycles/sec. on to the detector, which experiences a fluctuating temperature change due to energy absorption at three microns, unless both beams are of the same energy. Temperature changes are translated into electrical capacity changes which operate the servomechanism adjusting the attenuator until both beams are of equal intensity. The isotopic purity is a function of the position of the attenuator which drives an electrical potentiometer, the output of which actuates a linear potentiometric pen recorder in the control room with a scale range of 99.0 to 100 mole per cent. D_2O.

For general information on infra-red methods of heavy water isotopic analysis see Gaunt (1953).

Gamma-activity of the circulating heavy water

One of the gamma-health monitors mentioned above is located as nearly equidistant as possible from the four heavy water outlet pipes of the reactor aluminium tank. It indicates on a recorder in the control room and detects both any induced gamma-activity in the coolant (Laurence 1956) and also any large leakage of fission products into the heavy water coolant from a defective fuel element.

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SECTION 4

Basic Sciences

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The Sydney Air Shower Experiment — Paper 1

The Experimental Apparatus

By D. D. Millar*

A description is given of an array of detectors used in the experimental investigation of the properties of air showers of size 10^5 to 10^7 particles. The method of recording the data obtained from these detectors is also described. This is done on punched paper tape in a form suitable for handling by an electronic computer.

Introduction

Since the first observations by Bothe et al. (1937) of the coincident arrival of several cosmic ray particles at the earth's surface, the study of cosmic ray air showers has been actively pursued in many laboratories. As a result of such work the basic features of the phenomenon are now well understood. A high energy nuclear-active particle of the primary cosmic ray flux enters the earth's atmosphere, where it suffers a collision with a nucleus in the air. In this collision are produced secondary nuclear-active particles, nucleons and mesons, capable of further nuclear interactions. As a result of such primary, secondary, and succeeding collisions there develops through the atmosphere the nuclear-active cascade. Also produced in the nuclear interactions are π^0 mesons, which, decaying into gamma rays, initiate electron-photon cascades. Such electron-photon cascades generated throughout the atmosphere by the nuclear-active cascade account for the presence of the electron component of the air showers which constitutes all but a few per cent of the ionising particles detectable in the showers in the lower atmosphere.

The above chain of events is common to the history of all cosmic ray particles impinging on the earth's atmosphere. However, only the high energy particles produce air showers detectable in the lower atmosphere. The cascades produced by particles of lower energy are attenuated and may die out before they are detected, leaving only a residue of μ-mesons sparsely distributed over wide areas where their association in time of arrival cannot be readily detected.

If one wishes to study problems of the ultra-high energy cosmic ray particles, of energies several orders of magnitude larger than can be produced by laboratory accelerators, one must turn to observations on the extensive air showers. Direct observations of such particles is not feasible because of their low rate of incidence. The multiplication of particles which arises in the air shower however, and the spreading out of these particles, mainly caused by their Coulomb scattering in the air, permits the results of such high energy events to be readily detected in the lower atmosphere, and with appreciable frequency, by having a sufficiently large surface area covered by detecting apparatus.

Such air shower studies pursue two main problems. The one is of cosmological importance, and involves the search for possible anisotropies in the directions of arrival of the air shower primaries on the one hand, and the search for a high-energy cut-off in the incident cosmic ray energy spectrum on the other. Present indications are that such anistropy is small if it exists at all for showers of size greater than 10^5 particles (Clark, 1957) and that the cosmic ray energy spectrum continues without appreciable change in form from energies of 10^{10}eV to around 10^{18}eV (Clark et al., 1957). For such work large showers, implying high energy primaries, must be recorded and this has led to the development of giant air shower detectors with dimensions of hundreds of metres across.

The other type of study for which the air showers provide the observational data is concerned with the characteristics of the ultra - high energy nuclear interactions ($> 10^{15}$ eV) from which the showers arise. In this type of work, for which the Sydney apparatus was constructed and for which there is not the same need to pursue the giant air showers, one must infer the characteristics of the high energy interactions from the properties of the detected showers. Since the atmosphere forms an absorbing layer of 1045 $\mathrm{gm/cm}^{-2}$ thickness, and the geometric mean free path for nuclear interactions is of order 90 gm. cm^{-2} in air, the air shower is in general detected only after many mean free paths. In consequence, one is reduced to tackling the problem by assuming the form of the high energy interactions, the cross-sections, the multiplicity, the types and the energy distributions of the secondary particles, and on the basis of such a model deduce by cascade theory processes the characteristics of the model air shower in the lower atmosphere. Such model calculations have been carried out by Olbert and Stora (1957), and by Ueda and Ogita (1957). The work of these last authors has been used for comparison with the experimental data which is presented in an accompanying paper by Lehane (1958). It should be pointed out, however, that other models of the nucleon-nucleus collisions may equally well be postulated, which by suit-

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* University of Sydney, School of Physics. Manuscript received March 17, 1958.

SECTION 4, SHEET 1, BACKS ITSELF

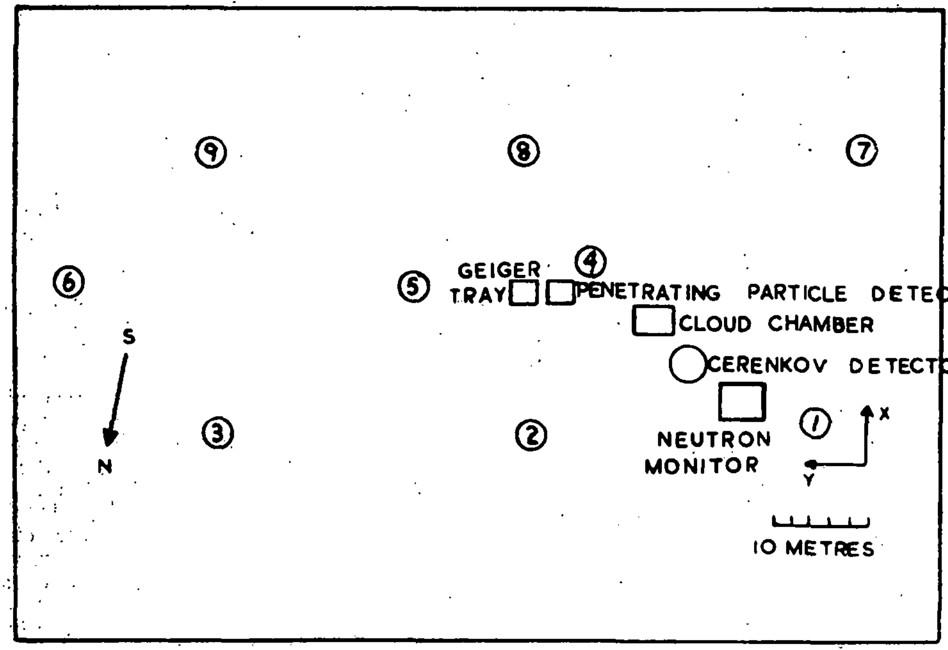

FIGURE 1:—The air shower array.

able adjustment of parameters could fit the experimental data.

It should also be pointed out that all such theories may predict the development of an air shower initiated by a particle of energy E at the top of the atmosphere. Unfortunately, the experimentalist does not observe showers according to this criterion. He observes showers of a certain size and these may have been initiated by a particle of energy E_1 at the top of the atmosphere, or by a particle of energy E_2 making its first collision considerably lower in the atmosphere. The fluctuations in the development of an air shower from a primary particle of given energy are likely to arise principally from this source of fluctuation in depth of the first collision. Such fluctuations should certainly be taken into account in comparing the results of theoretical computation with observation.

The Sydney experimental array

The Sydney apparatus consists of an array of nine liquid scintillation counters, each of 0.23 sq. metres area and liquid depth 10cm., arranged as shown in Figure 1. They are distributed within an area of 70m. by 30m. and the spacing between detectors is approximately 20m. These detectors provide the basic information which is recorded on each shower—namely, the shower size, the position of the shower core, and the direction of incidence of the shower. With this basic information are correlated the data obtained from the other detectors. The method of determining the first two quantities from the local particle densities at each scintillator and by fitting these to an assumed, and experimentally substantiated, shower structure function, is dealt with in the paper by Wallace (1958). The direction of incidence of the shower is determined by the method of Bassi, Clark and Rossi (1953) by measuring, with millicrosecond techniques, the time intervals between the front of shower particles hitting four of the scintillators, numbers 2, 3, 8 and 9 in Fig. 1. These four (see Fig. 1) scintillators in coincidence normally provide the triggering pulse indicating the arrival of the air shower, although an alternative triggering pulse provided by the coincident discharge of a number of Geiger counters out of three trays in the vicinity of the cloud chamber has on occasion also been used.

A tray of 40 Geiger counters and of area 0.4 sq. m. is also included, in order to compare the local particle density as measured by the scintillators with that measured by the Geiger counters. This comparison, the results of which are described by Wallace, enables us to conclude that the estimate of density given by the scintillators is not very much different from that of Geiger counters. The showers detected by our scintillator array can therefore be compared with those detected by other workers using Geiger counter arrays.

Also included is a Wilson cloud chamber of 65 cm diameter and 25 cm. illuminated depth. This has been used to provide a comparison between the fast timing determination of air shower directions and the mean directions of tracks photographed in the chamber. The result of this comparison is mentioned by Malos (1958).

Two devices are included in the array which afford experimental data on the μ-mesons and the nuclear-active particles in the showers. The first of these is a penetrating particle detector consisting of three trays of individually hodoscoped counters embedded in a lead pile. From the response of this detector information has been obtained, and is presented by Lehane (1958), on the μ-mesons and the high-energy nuclear-active particles. The second device is a neutron detector whose prime function is the continuous recording of the flux of low energy nuclear-active cosmic ray particles. It has been included as part of the air shower array to give information on that portion of this flux which is part of the incident air showers. Preliminary results from this last detector confirm similar observations by Marsden and Doran (1956), but further observations are required in order to take into account adequately the inherent fluctuations in the response of such a detector.

Lastly, the array contains a Cerenkov detector consisting of a parabolic mirror which focuses the Cerenkov light emitted by an incident air shower on to the photocathodes of seven photomultipliers. The magnitude of the light pulses detected by such a device may be expected to be related to the size of the air shower at altitude above the ground level detecting equipment, so that the attenuation of individual air showers in the atmosphere could in principle be determined if this relation were established. The results reported by Brennan (1958) indicate the importance of knowing the direction of incidence of the shower and the angular distribution of the particles in the shower, before this method of determining shower size at altitude may be feasible.

The recording of the data

The cloud chamber is photographed stereoscopically upon arrival of a detected shower. These photographs are later re-projected on to a ground glass screen to determine the directions in space of the individual particle trajectories. The time intervals between the arrival of the shower front at the four scintillators used for direction finding are measured by photographing the screen of an oscilloscope on which are displayed the four scintillator pulses (Malos, 1958).

All other information is recorded automatically on punched paper tape in a form suitable for handling by Silliac, the electronic computer in the Adolph Basser Computing Laboratory. The recording circuits are gated by a triggering pulse, provided as mentioned above, of resolution time about 5 micro-sec. Within this gating time, pulses from the photomultipliers of the nine scintillators and of the seven-element Cerenkov detector, also from the addition circuit of the unshielded Geiger tray, are accepted by stretching circuits which convert these pulses into D.C. voltage levels stored on polystyrene dielectric condensers.

These stored voltage levels are then converted one at a time into a binary number in the range 0 to 40, covering a voltage range 0-60V, by one volt steps from 0 to 20V, and 2V steps from 20 to 60V. The gains of the circuits are arranged so that one volt equals the average pulse height produced by a single relativistic μ-meson traversing a scintillator, and this level is assumed to correspond to one air shower particle detected by the scintillator. The binary numbers thus formed are punched on to paper tape. Other information, such as the time of occurrence of the shower and its serial number, are also punched on the tape in binary form.

The information from the hodoscoped Geiger counters in the penetrating particle detector are punched on the tape more directly. The pattern of holes punched out gives a direct visual presentation of which of the Geiger counters has been traversed, and which have not, by ionising particles.

The recording of all data from a single shower occupies about 2ft. of tape, and takes about five minutes to be punched out. The tape carrying one day's records, of about 60 showers, is fed into Silliac with a program tape which converts pulse heights into particle densities, according to the gains of the individual scintillators. These gains are measured daily by counting the rate of pulses above the one scintillator particle level, and comparing this with a previously determined pulse height distribution. The gains of the stretching circuits are checked at intervals of two weeks. The overall uncertainty in the conversion from recorded pulse heights to density of particles caused by instrumental error is less than 10 per cent., which is less than would be the Poissonian fluctuations in numbers of particles traversing a scintillator.

Discussion

The apparatus which has been described above was designed for studies of air showers of size 10^5 to 10^7 particles initiated by primaries of energies estimated on current theories to be of order 10^{16} to 10^{17} eV. The design of the detecting array and the method of recording the data have both been influenced by the location of the experiment—on the one hand the positioning of individual detectors has been largely determined by the design of the roof of the School of Physics building, while on the other, the method of recording owes much to the presence within the building of the electronic computer, Silliac.

The apparatus enables one not only to study the average properties of showers which may be classified according to size, core location or direction of incidence, but also, with increasing numbers of events recorded, the fluctuations in these properties about the average values may be investigated.

In some cases the results obtained with different detectors, for example, the cloud chamber and the fast-timing apparatus used for the determination of the shower directions, may be directly compared, and insight thus obtained into the nature of the response of each detector and into the nature of the air showers themselves.

Acknowledgements

It is a pleasure to acknowledge the large part played in the design and operation of this experiment by the other members of the air shower team: M. H. Brennan, W. M. Layson, J. A. Lehane, J. Malos, M. H. Rathgeber, C. S. Wallace and M. M. Winn. My thanks are also due to Professor H. Messel for his continual stimulus in this work, also to him and to the Nuclear Research Foundation within the School of Physics of the University of Sydney for the provision of excellent experimental facilities.

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The Sydney Air Shower Experiment. Paper 3:—

The Zenith Angle Distribution of Air Showers

By J. Malos*

The zenith angle distribution for showers in the size range $10^5 = 10^7$ particles has been obtained experimentally. The shower direction is calculated from the relative arrival times of the shower front at four of the nine scintillators used in the Sydney air shower experiment. The various contributions to the error in the zenith angle, θ, are discussed. The angular resolution in θ is about ± 4°. The zenith angle distribution is used to determine the effect of atmospheric absorption of showers in the size range $10^6 = 10^7$ particles. The results are in agreement with those of other workers.

Introduction

The rate at which air showers are detected by a given apparatus changes with variation in altitude, variation in barometric pressure at a fixed altitude and variation of zenith angle. All these effects are related in that they are produced by changes in the thickness (in units gm. cm. $^{-2}$) of the air traversed by the shower before detection. Study of these effects in the lower atmosphere indicates that the observed frequency of shower detection decreases exponentially with increasing depth of atmosphere with a practically constant attenuation length. An accurate knowledge of the effects of atmospheric absorption allows certain characteristics of the development of the air shower to be inferred.

The zenith-angle distribution for showers in the size range -0^5-10^7 particles has been obtained. From this a value for the absorption length is obtained and compared with previous results.

The arrival directions of showers as determined by the fast timing method (Bassi et al. 1953) have been compared with observations using a cloud chamber.

Method and experimental arrangement

Bassi et al. (1953) showed that at a given instant most of the shower particles lie in a flat disk perpendicular to the direction of propagation of the shower axis and moving with a velocity near that of light. The relative arrival times of the shower front at three detectors is, therefore, in principle, sufficient to determine the direction of incidence of a shower. In the Sydney air shower experiment described by Millar (1958), four of the nine scintillators are used for this purpose. The four detectors lie in a horizontal plane and are arranged in a rectangular array 31 metres by 26 metres.

The direction cosines of the plane of the shower front are given by l and m, and its velocity of propagation is taken as equal to the velocity of light c. A least-squares analysis leads to the following values of l and m,

$$l = \frac{c\ (t_2 - t_3 + t_1 - t_4)}{2a} \qquad (1)$$

$$m = \frac{c\ (t_2 - t_1 + t_3 - t_4)}{2b} \qquad (2)$$

where a = 26 metres and b = 31 metres, and t_1, t_2, t_3, t_4 are the arrival times of the shower front at each of the four scintillators.

The zenith angle Θ is given by the following relation:

$$O = \sin^{-1}\ {}^{1}\ (l^2 + m^2)1/2 \qquad (3)$$

A measure of the error in each measurement is given by

$$\epsilon = \frac{t_2 - t_1 + t_4 - t_3}{2} \qquad (4)$$

In the determination of the relative arrival times of the shower front at each of the four scintillation detectors, the pulses produced in the four photomultipliers by a shower are separated in time by delay lines. After amplification, the pulses are displayed on the screen of a fast oscilloscope. Arrival of a shower is determined by coincidence of pulses in the four scintillators within a resolving time of 200 millimicro sec. Discriminators in each scintillator channel ensure the presence of. one or more particles in each scintillator.

An event is recorded by photographing the oscilloscope trace with an automatically controlled camera. At the beginning of a day's run the sweep speed is calibrated by photographing a 100 Mc/s signal. After every ten shows a millimicrosecond pulse generator introduces a fast pulse into the preamplifiers of each of the four channels, thereby simulating a vertical shower and affording a frequent check on the stability of the sweep speed. These check pulses also allow a determination of the artificial delays which are introduced in three of the channels. The point where each pulse commences to rise is measured.

Experimental results

In Figure 1 is a plot of ϵ for all the 707 recorded showers of determined size. The mean is located at —0.7 millimicro sec. and the standard deviation for the distribution of ϵ is

$\sigma(\epsilon) = 7.2$ millimicro sec.

This corresponds to a resolution of the order of ±5° for vertical showers.

Fluctuations in the measured relative positions of the artificially produced pulses determine instrumental and reading errors as being not greater than 2 millimicro sec.

This indicates that the major contribution to the error ϵ comes from the thickness of the shower disk and the shower front not being a plane. A check on 62 of the showers, in which

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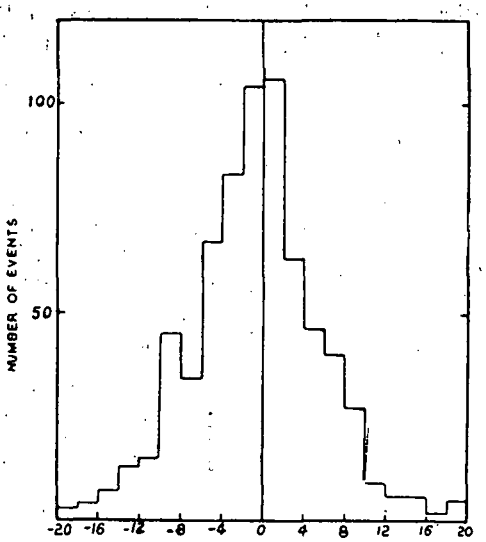

FIGURE 1:—Distribution of error, ε.

three of the detectors each had recorded a minimum of at least three particles and the other had recorded only one particle, indicated that excessive ε was associated in general with a delay in the arrival of the single particle. This delay is consistent with a shower disk thickness ~ 5 metres as is also the value obtained for σ(ε). This is consistent with scattering of the electrons in a shower.

Comparison with cloud chamber observations

The direction of the shower axis obtained both by the fast timing method and measurement of the directions of the shower tracks in a large Wilson cloud chamber (Brennan et al. 1957) have been compared. Forty-nine showers have been recorded for which a direction of incidence has been measured by the fast timing method at a time when only three scintillators were in use and which also gave a direction in the cloud chamber. There is good agreement within the experimental errors. Our later results, using four scintillator pulses, indicate that the previous results using only three pulses were not subject to appreciably larger error than are quoted above.

Zenith angle distribution

The resolution has been improved to ±4° for purposes of determining the zenith angle distribution by rejecting showers with ε > 10 millimicro sec. A total of 604 showers satisfied this criterion and were of size $\geq 10^5$ particles (Wallace, 1958). The zenith angle distribution of these showers is used to obtain the effect of atmospheric absorption of showers in the size range 10^5 to 10^7 particles.

For this analysis we assume, as in Clark (1957), that the absolute intensity has a power law dependence on shower size N and depends exponentially on x, x being the atmospheric thickness traversed by the shower, and measured in the direction of the shower axis. The number of showers of size > N detected at an atmospheric depth x is then

$$K(N,x) = K_o {\frac{N}{10^6}}^{-\Upsilon} e^{\frac{-(x-x_o)}{\Lambda}} \qquad (5)$$

where the slight variation of the exponent, γ, with size is ignored and γ is taken as equal to its value 1.53 for N = 10^6 particles, xo = the vertical depth of the atmosphere and is taken as 1045 gm. cm. $^{-1}$, $x = \frac{xo}{\cos \Theta}$ and Λ is a measure of atmospheric absorption.

From equation (5) it follows that

$$\frac{1}{\Lambda} = \frac{-\delta \ln R}{\delta x} \qquad (6)$$

where R = the measured rate per steradian.

A histogram plot of the number of showers nθ, at intervals of Θ 5° is made. $\frac{n\theta}{\sin \Theta}$, where Θ is taken at the mid-point of each interval, is then a measure of the rate of showers per steradian. Λ is obtained from the slope of the plot of the logarithm of $\frac{n\theta}{\sin \Theta}$ against x — xo as in Figure 2.

The value of Λ = 129 ± 12 gm. cm. $^{-2}$ is obtained from the slope of the line which is fitted to the data X-Xo × 200 grm. cm. $^{-2}$. Data for X-Xo > 200 gm. cm. $^{-2}$ is suspect because of the decrease in resolution with increasing Θ and the rise in the probability of detection of a shower of given size with increas-

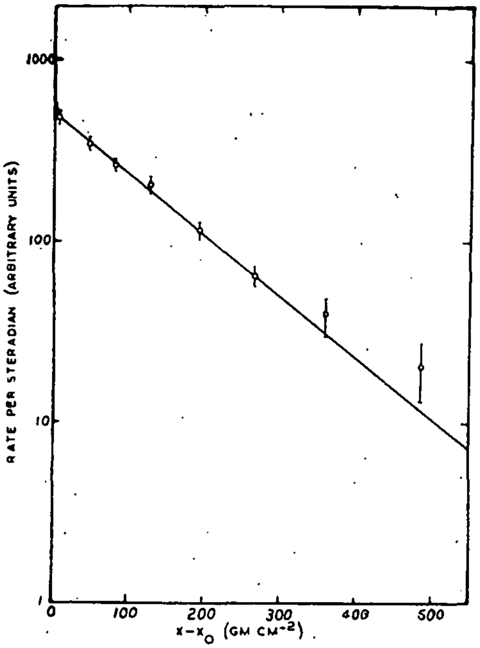

FIGURE 2:—Relative rate of detection of showers versus atmospheric depth obtained from the observed distribution in zenith angle.

ing zenith angle. Errors in θ tend to broaden the zenith angle distribution. The overall effect is slightly to increase the absorption length Λ. Allowing a figure of 5 per cent. (Clark, 1957) for this effect we have

$$\Lambda = 122 \pm 12 \text{ gm. cm.}^{-2}.$$

This value may be compared with the value of 133 gm. cm. $^{-2}$ obtained from both variation in the rate of showers with altitude and barometer coefficients at altitudes up to 3,500 m. (Greisen, 1956). Clark (1957), using a zenith angle distribution, determined by a similar experimental technique to ours, obtains a value of $\Lambda = 107 \pm 11$ gm. cm. $^{-2}$ for showers of the same range of sizes. Clark et al. (1957) obtain a still lower value of $\Lambda \sim 95$ gm. cm. $^{-2}$ for showers of larger N.

Differences in the above results are not yet known to be significant; they may be caused by a systematic error so far unresolved.

If one assumes that the number of particles in a shower decreases with depth exponentially so that

$$N(x) = N(Xo)e^{\frac{-(x-xo)}{\lambda}} \qquad (7)$$

where N(x) and N(xo) are the numbers of particles in the shower at depth x and xo respectively and λ is the attentuation length, then from (5) and (7),

$$\lambda = \gamma\Lambda \qquad (8)$$

This quantity λ so defined is then of physical significance, and affords information on the effectiveness of the role of the nuclear-active cascade in sustaining the life of the shower in the lower atmosphere. However, it should be pointed out as has been done by Miyake (1958), that the possibility of large fluctuations in the depth of the primary collision initiating the shower can lead to an interpretation of the experimentally measured quantity Λ which depends strongly upon the mean free path for the primary interaction, rather than upon absorption of the particles of an individual shower alone.

Conclusions

The zenith angle distribution of air showers of size $\geq 10^5$ particles has been determined and is in reasonable agreement with other observations on barometer coefficient and altitude dependence of air showers. The statistics determine whether there is any variation of the zenith angle distribution with shower size.

Acknowledgements

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Free Radical Measurements in the Radiolysis of Liquid Hydrocarbons

By K. H. Napier and J. H. Green*

Molecular iodine labelled with iodine 131 has been used as a detector for the radicals produced in the initial ionising event hydrocarbon liquids under high vacuum. Alkyl iodide mixtures were formed during irradiation by a Sr90—Y90 beta source. The mixtures were analysed by vapour phase chromatography. Experimental results for pentane-iodine mixtures are discussed.

Introduction

The use of molecular iodine labelled with iodine 131 as a radical scavenger was first used by the Durham group (Durham, Martin and Sutton, 1949). The principle of the method is that iodine is homogenously mixed in the liquid or gas to be irradiated, but in small quantity, so that iodine itself does not absorb the radiation or interfere with fast reactions between primary radicals within the particle tracks. Energy transfer between the "hot" radical and a heavy atom is fast, and the assumption is that each collision between a radical and iodine results in an organic iodide. The mixture of iodides is then usually analysed by adding carriers and fractionally distilling (Williams and Gevantman, 1952). Although the point has not yet been checked, it is felt that the organic free radical, diphenyl-picryl-hydrazyl (DPPH) is probably a more efficient scavenger than iodine. This suggestion is made by Fessenden and Schuler (1957), when they were comparing their results for cyclohexane with those of Prevost-Bernas et al. (1952), who used DPPH. Bevington (1955) concluded, however, that DPPH is only about 70 per cent. efficient as a scavenger. DPPH is a total radicle scavenger in the sense that individual radicals cannot be distinguished. This is true also for the method of Dainton (1948), in which the initiation of polymerisation of acrylonitrile in aqueous solution is used as a measure of radicals produced by the absorption of ionising radiation in water.

The advantage of iodine for radical catching and for identification of the radicals rests on two main assumptions. First, that the scavenging is completely efficient: even if it is not, the proportions of the iodine-labelled products should be the same. However, the second assumption is that all reactions of the type $R + {}^*I_2 \rightarrow R{}^*I$ are equally likely, and that the iodinated products are themselves stable.

Evidence for the first assumption was provided by McCauley and Schuler (1957), who compared the amount of iodine reacted with the amount of iodine which was combined in alkyl iodides formed. The assumption that all reactions are equally likely is, of course, an approximation based on the large mass difference between iodine and any radicals formed (at least in lower hydrocarbons). It has been necessary also to determine whether or not the concentration of dissolved iodine interfered with the radiolysis of the solvent hydrocarbon. Schuler (1957) has shown that iodine has no disturbing effect on the yield of hydrogen gas in the decomposition of cyclohexane, provided that the iodine concentration was below 10^{-4}M. However, the hydrogen yield is a relatively insensitive detector of such a disturbance. Thus, it has been found that larger concentrations of iodine than 10^{-4}M decrease the hydrogen yield. This effect can only arise in the removal of hydrogen atoms (formed by splitting C-H bonds) by iodine, presumably with the formation of appreciable amounts of hydrogen iodide. This point is extremely interesting, because hydrogen iodide has not yet been reported as one of the active products of radiolysis, but we find indications of large amounts of HI in the gas chromatograms.

This paper presents the first results of an investigation into the radiolysis of pentane-

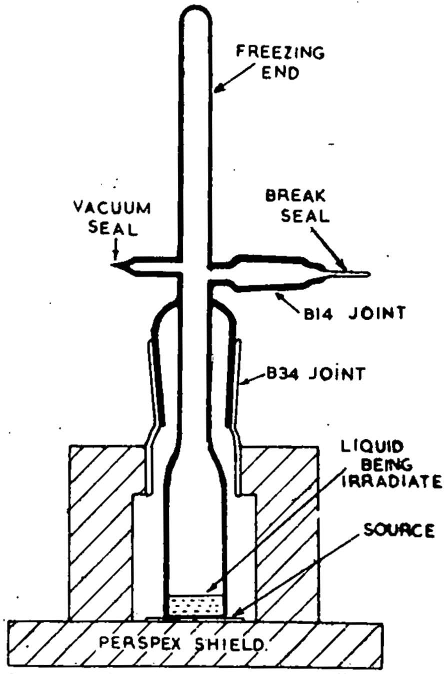

FIGURE 1: Source housing and cell.

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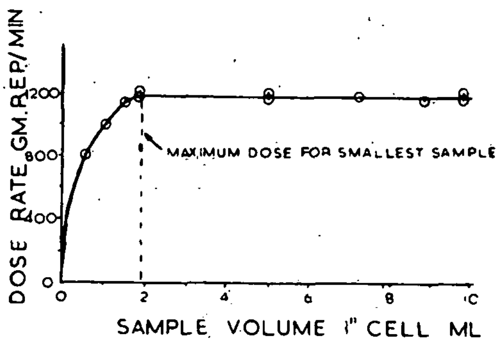

FIGURE 2.

iodine mixtures. Gas chromatography has been adopted for the determination of the labelled alkyl iodides which are formed, because it is undoubtedly the most sensitive and potentially the most accurate method available.

Experimental

The source available for the radiolysis was a 500 mC Sr90—Y90 source, sealed on a flat silver strip. The active area measures 1 cm. by 2 cm. The cells for use with this source were designed as flat bottomed cylinders 1in. in diameter, the bottom of the cell being as thin as possible so that only a small amount of radiation would be lost by absorption. (Figure 1.)

The source was shielded in a perspex box, with walls 1in. thick, one of them being removable to allow installation or removal of the source. A B34 standard socket was set in a hole in the centre of the top of the box. The sample cell could be lowered through the socket, and fixed in position by an attached B34 cone fitting into the socket. The cells were of such a dimension that the bottom was just above the source.

Lead shielding was also provided to absorb any bremsstrahlung by reaction of the beta particles with the source and housing. The 1in. diameter cells allowed complete coverage of the active area, for most efficient use of the source. There was a constant geometry when each cell was oriented in a set direction.

Calibration runs were made with ferrous sulphate solution to determine the dose received. The G value for the process was taken as 15.5 for ferric iron oxidised in 0.8N sulphuric acid as described by Haybittle, Saunders and Swallow (1956). At first consistent results could not be obtained. However, when the ferrous sulphate solution was stirred by passing a fine stream of air through it during irradiation, reproducible results were obtained. (No effect was detected by passing air through solutions without irradiation for similar times to those of the exposure.) Samples of various sizes were now irradiated, and, if the results were plotted as total dose in gm. rep. against the volume of the solution, the curve shown in Figure 2 was obtained.

It seems that all the beta radiation is absorbed in the first 0.4 cm. of solution, and, therefore, it is no advantage for this work to irradiate samples of greater volume than 2 ml., because the total absorption of radiation is not increased.

The organic solutions for irradiation were prepared under high vacuum. Attempts to prepare iodine labelled with iodine 131 were unsuccessful if the iodide solution was dried with potassium dichromate under vacuum, when the iodine should be liberated on heating. Because only small amounts of iodine (0.1 mgm.) were required for each run, a large fraction was usually lost by adsorption on the walls and stopcock grease of the vacuum system.

Samples for irradiation were finally prepared by forming the iodine by oxidation of 0.1 mgm. of iodide and 20 microcuries of iodine 131 with acid and potassium iodate. The iodine was extracted into the pentane and the solution was washed with water and dried over phosphorous pentoxide. The pentane was of C.P. quality, untreated in any way except in the drying and degassing steps in vacuo. The mixture which resulted contained less than 10^{-4} molar iodine. The iodine solution was transferred to an irradiation cell and frozen by liquid oxygen. Then by successive pumpings, degassings and freezings in the normal manner, the cell was sealed off under vacuum.

The samples were irradiated for various times from 200 to 1,000 minutes. For 1 ml. samples the dose rate was 1,100 rep/min., giving a total dose ranging from 2×10^4 to 10^5 rep. After the irradiations, there remained a mixture of various hydrocarbons, hydrogen and products containing iodine. It is only for the iodine products

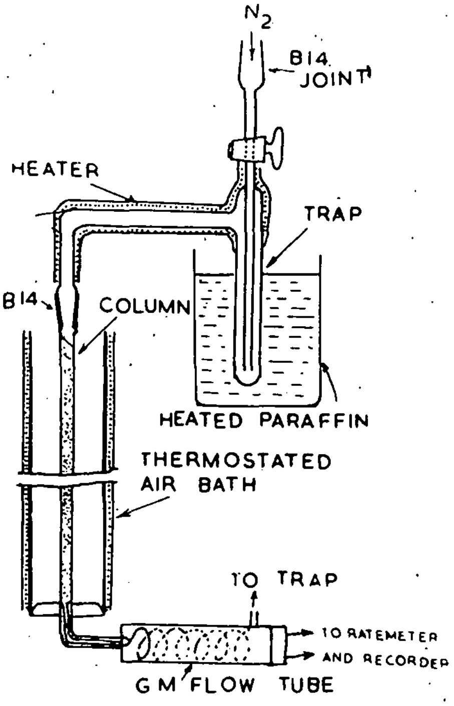

FIGURE 3: Active gas chromatography.

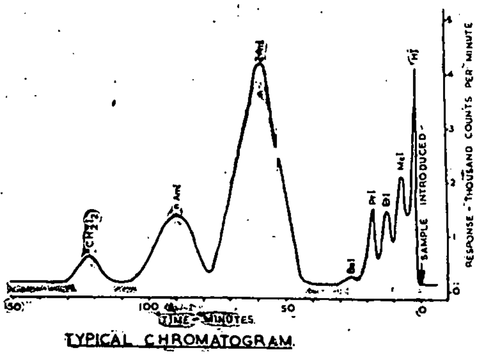

FIGURE 4: Typical chromatogram.

that the mixtures were analysed. The analysis for hydrocarbon products is difficult without a mass spectrometer.

Analyses were performed by gas chromatography according to the scheme shown in Figure 3. A column of silicone oil on firebrick was used, with nitrogen as the eluting gas, at temperatures of 50°C. to 100°C. The detector was a flow Geiger-Mueller tube (Type F10 Twentieth Century Electronics), in which the gas flows through a fine spiral in the centre of the tube. This tube was connected to a ratemeter coupled to a chart recorder. Only portions containing iodine were now detected, in amounts much less than 0.1 mgm. This method not only gives a very sensitive qualitative analysis, but also the peak areas are directly proportional to the amount of labelled iodide present.

The reaction products from the cell were transferred to the column by incorporating a break-off seal on the cell by which the contents could be reintroduced into the vacuum system. The seal was broken, and the contents were distilled into a trap cooled in liquid oxygen. Any non-condensible products could be pumped by a Toepler pump into a small calibrated capsule so that their volumes could be measured. This fraction would be mainly hydrogen and methane. The trap retaining the condensible products was now removed from the vacuum line and connected to the column by a ground glass joint. The contents were evaporated from the trap by placing a heated paraffin bath (at 100°C.) around the trap. When evaporation was complete, the nitrogen stream was turned on. Flows of about 30 ml. per minute were used. The large sample of the hydrocarbon present reduced the efficiency of the column. This in turn reduced the resolution of the lower fractions, but did not affect the later fractions. The various components in the chromatogram were identified by calibration of the column with samples of known alkyl iodides, using a thermal conductivity bridge as detector.

However, because free iodine left unreacted will not pass through the column, but will exchange activity with a second sample, a new column was needed for each run. An inactive sample of methyl iodide, which is passed through a column after an active run, will appear active at the detector. This exchange could lead to enriching of the lower fractions more than the later fractions because the rate of exchange is faster for the lower fractions, and thus give erroneous results. Each was calibrated after a run by passing inactive methyl, ethyl and propyl iodides through the column with a conductivity gauge as the detector.

Results and discussion

A typical chromatogram is shown in Figure 4. Repeated analyses have so far been consistent to within 5 per cent., and only those for pentane-iodine mixtures are presented here. We are not yet satisfied that we can give accurate G values for the various iodides because it is difficult to compare the counting efficiency of the tubes used for the initial mixture and for the final vapour fractions. The results are therefore given as percentages of the total iodides eluted at column temperatures up to 100°C. The results for pentane are given in Table 1.

TABLE 1.—ANALYSIS OF PRODUCTS OF RADIOLYSIS OF IODINE-PENTANE MIXTURES

IODIDE	PERCENTAGE OF TOTAL	
Methyl	8a	24b
Ethyl	4	30
n-Propyl	3	6
n-Butyl	1	5
sec-Amyl	50	—
n-Amyl	17	22
Methylene	4	13
Hydrogen	11	3

b: Results of Williams and Gevantman (1952).
a: Present work.

Williams and Gevantman (1952) have reported their results for pentane-iodine vapour mixtures. These were obtained by carrier-distillation, which is not as useful as the chromatographic technique. Their results show that the yields of methyl, ethyl, and amyl iodides are about the same and that propyl, butyl, and methylene iodides are present in much smaller, approximately equal amounts. Our results are not at all similiar for the six iodides mentioned as one may see in Table 1.

However, the two outstanding features of our results are the presence of a very large amount of secondary amyl iodide, larger even than the normal iodide, and the presence of appreciable amounts of hydrogen iodide.

Of all the iodides formed, methylene iodide is the only one which requires some secondary process for its production. All the others can clearly be formed by a primary process only, and this fact lies at the basis of the scavenger method. The primary process resulting from the excitation and settling down of the molecule after radiation energy is absorbed, is the fission of C-C and C-H bonds. The C-C bonds are much less readily broken than C-H bonds, as the large percentage of amyl iodides clearly shows. It is also clear that C-H bonds break more readily in the 1 and 2 positions than in the others. (See below.)

Although Dewhurst (1957), on the basis of his study of hydrocarbon products of the radiolysis of n-hexane, considers that C-C bond rupture at positions 2 and 3 is the most likely, we cannot see any connection between this suggestion and the primary radical yields. In our experiments, breakage of the first C-C bond predominates to give 8 per cent. CH_3I.

It is only recently that the presence of large amounts of secondary alkyl iodides have been found in the radiolysis products. For example, McCauley and Schuler (1957) find G values of 1.0 and 2.1 for normal and secondary butyl iodides respectively in the decomposition of butane-iodine mixtures. Dewhurst (1958), using saturated (approx. 0.03 M) inactive iodine in n-hexane, finds G values of 0.70 and 2.60 for normal and secondary hexyl iodide. In our experiments, using very small concentrations of labelled iodine, and analysing by vapour phase chromatography with carrier-free organic iodides, we find percentages of 17 and 50 for normal and secondary amyl iodide. These results show clearly that C-H fission occurs primarily at the first and second carbon atoms. However, it is certainly feasible, that all hydrogen abstraction leading to amyl iodides could occur at the first carbon atom, followed by a very fast settling down of the molecule, which makes the second carbon atom reactive. Secondary amyl iodide would result from this rearrangement. If this is not feasible, then C-H breakage at the second carbon atom must lead to the large amount of secondary amyl iodide found.

The production of hydrogen iodide in hydrocarbon-iodine mixtures, which is indicated on our chromatograms, has not been reported up to the present. In the decomposition of alkyl halide vapour, the formation of a trace of HI was noted by Williams and Gevantman (1952). If the radiolysis of liquid mixtures is taken to completion, i.e., until there is no remaining free iodine, and if HI is as good a scavenger as iodine as claimed by Hanrahan and Willard (1957), then we should not expect to find HI in the products. In our experiments, no more than about three-quarters of the initial labelled iodine was consumed in the radiolysis, and we might expect to find some hydrogen iodide in the analysis.

There are many points which need clarification, and we intend to continue with a rigorous examination of the pentane-iodine system using vapour phase chromatography, and with pentane alone, using mass spectrometric analyses.

Acknowledgements

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The Chemical Fate of a Radioactive Atom

By D. L. Baulch and J. F. Duncan*

The full text of this paper is being submitted after the Geneva Conference on the Peaceful Uses of Atomic Energy, 1958. A summary of the paper is given here.

Summary

In a radioactive process in which the daughter atom is a different element from the parent, the decay produces a change in the nuclear field strength attracting the electrons. Since the energy levels of the parent and daughter are different, expansion (as in alpha-decay) or contraction (as in beta-decay) of the electron shells about the nucleus will occur. If this rearrangement is slow by comparison with the time scale of the nuclear event (10^{-12} sec.) electronic excitation results. The degree of excitation may be quite large, of the order of 100 eV for an atomic number of 50, and sufficient to cause considerable ionisation. This may initiate chemical reactions which make it difficult to interpret the chemical effects of nuclear recoil.

If one wishes to use the radioactive process to study the chemical behaviour of such unusual species as $(CH_3NHO)+$ or $Pr^{4}+$ (which can quite easily be made by radioactive methods) it is essential to know how much excitation is produced in any particular case, the chemical consequences and how they may be controlled.

Electronic excitation must always take place in an isolated atom or molecule undergoing a radioactive change involving a change in charge. But it does not follow that in some circumstances either (i) electronic readjustment may not take place before chemical reaction can occur, or (ii) the electronically excited state may not be stable and/or chemically unreactive.

If, in the former case, conditions can be controlled so that electronic excitation is lost before the bond between the radioactive atom and its parent molecule is affected, then the chemical behaviour of the daughter atom will be the same as if the original decay had taken place without electronic excitation.

Evidence from the following sources is considered in detail to ascertain when this might occur:—In (a) ionisation by gamma-radiation; (b) charge production in radioactive decay; (c) internal conversion; (d) radiative neutron capture; (e) beta-gamma decay. It is shown that examples are known in which excited states may initiate chemical reaction, may be stable, or may be de-excited before chemical reaction can occur.

An electronically enriched environment is most likely to allow isothermal readjustment of excited states to take place during radioactive decay; heavy atoms are also to be preferred, since the degree of excitation is then likely to be small.

Some results obtained for the beta-recoil yield of ThC from lead oxide surfaces on which a carrier-free mixture of ThB/C has been deposited are presented and discussed in this context. We show unequivocally that the surface chemistry plays an important part in determining the fate of a daughter atom, even under conditions which are most favourable to the elimination of electronic excitation.

The conclusions arising from this work are considered in relation to the problem of obtaining (a) chemical binding energies from recoil induced by beta-decay; (b) recoil energies of nuclear physical interest and (c) radioactive and stable isotopes by nuclear recoil.

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Manuscript received March 17, 1958.

The Sydney Air Shower Experiment — Paper 5:

Cerenkov Radiation from Cosmic Ray Air Showers

By M. H. Brennan*

Some of the characteristics of the Cerenkov light emitted by the particles in the air shower during its passage through the atmosphere are studied, using a light receiver in association with an array of scintillation counters. It is shown that, although no functional relationship can be established between the size of the Cerenkov pulse and the distance, R, of the light receiver from the shower core, the maximum height of the pulse decreases with increasing R. It is also found that the light may be emitted at angles of at least 30° to the shower axis. From the angular distribution of the Cerenkov light the angular distribution of the electrons in the shower with energies above 21 Mev is obtained. Finally, it is suggested that, for a given angle between the shower axis and the axis of the acceptance cone of the light receiver, the size of the Cerenkov pulse depends primarily on the integrated particle flux in the acceptance cone.

Introduction

The study of cosmic ray air showers has been handicapped because observations on individual showers could reasonably be made only at one level in the earth's atmosphere. Hence, information on the longitudinal development of the showers during their passage through the atmosphere has had to be obtained from the average behaviour of showers detected at various altitudes.

However, a method by which the development of individual showers could possibly be studied has been developed by Jelley and Galbraith (1953, 1955). In a series of experiments at Harwell and the Pic du Midi, Jelley and Galbraith established that the Cerenkov radiation emitted by the particles of an air shower during their passage through the atmosphere could be detected against the background night sky illumination level by a light receiver, consisting of a parabolic mirror with a photomultiplier at its focus. This result was later confirmed by the Russian workers, Nesterova and Chudakov (1955).

The early experiments of Jelley and Galbraith were confined to simple apparatus, thus making the interpretation of results difficult. In particular, it was not possible to study quantitatively the dependence of the height of the Cerenkov pulse on shower size and on the location of the core of the shower with respect to the light receiver. Also, in the analysis of their results Jelley and Galbraith found it necessary to assume that most of the light detected by their receiver was emitted by electrons close to the shower axis and with trajectories close to the direction of the shower axis. It was on the basis of this assumption that the light receiver was used in a search for possible point sources in the high energy cosmic ray flux. (Jelley and Galbraith, 1955.)

The purpose of the present experiment is to investigate in a more quantitative manner the various characteristics of the Cerenkov radiation from air showers. We shall, in fact, show that there is no simple relationship between the size of the Cerenkov pulse and the distance of the shower core from the light receiver, and also that the light is emitted at angles of at least 30° to the shower axis.

Apparatus

In the present experiment the light receiver is located near one end of the array of scintillation counters used in the Sydney air shower experiment (Millar, 1958). This allows us to study showers of size $\sim 10^5$ to 10^7 particles whose cores fall up to $\sim$ 60 m. from the light receiver.

The light receiver consists of seven 2 in. diameter photomultipliers (EMI type 6260) clustered about the focus of a 44 in. diameter parabolic mirror, which has a focal length of 18.6 in. This arrangement results in seven acceptance cones, each with a half angle of $\sim 3°$, clustered about the vertical.

High tension to the photomultipliers is supplied through potentiometers from a central power supply. The unit is designed in such a way that at no time is there an anode current greater than 100 microamperes in any one of the seven photomultipliers. This ensures stable operation of the detector, and allows us to operate despite large fluctuations in the light intensity which occur when cloud passes over the detector. As a further precaution, the hut in which the detector is housed has a roller blind roof which is closed by a time switch one hour before sunrise.

The pulses from the photomultipliers are amplified in separate preamplifiers, and then fed through coaxial cable to the central recording

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station. The manner in which the pulse heights are recorded has been described elsewhere (Millar, 1958).

Since the E.H.T. supplied to the photomultipliers varies so that the anode currents remain less than 100 microamperes, provision has to be made for recording the value of the E.H.T. This is done continuously on a chart recorder and also at the time of the shower by recording the instantaneous value of the E.H.T. on the paper tape used to record the pulse heights. Thus, all pulse heights can be normalised to their value at 1,000 V. high tension. Pulses of normalised height less than 0.6 V. are not accepted. This level, which is dictated by the high night sky luminosity, corresponds to ~ 6,000 photons incident on a 2 in. diameter photocathode, after reflection from the parabolic mirror.

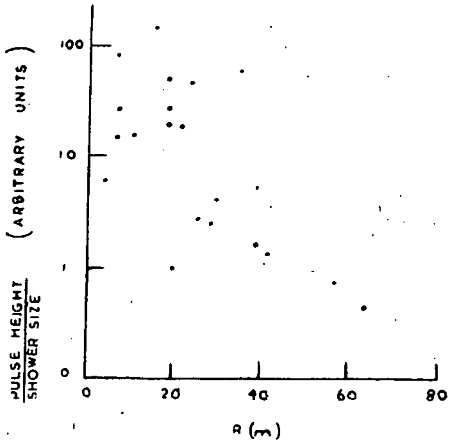

FIGURE 1:—The dependence of the reduced Cerenkov pulse height on the distance, R, of the light receiver from the shower core.

Radial dependence of the Cerenkov pulse

The radial dependence of the Cerenkov pulse was investigated by using the response of the scintillator array to yield an estimate of the size, N, of the shower and the distance, R, of the light receiver from the core of the shower. In this first series of experiments only the central photomultiplier was used, and events analysed only when the high tension supplied to the photomultipliers indicated that there was no cloud overhead.

Two hundred and six showers were recorded for which core location and shower size could be obtained. Of these, 22 were accompanied by a pulse from the central photomultiplier of normalised height greater than the minimum acceptable level. The ratios, C/N, of Cerenkov pulse height to shower size as a function of R are shown in Figure 1 for these 22 events.

It is evident that no functional relationship between C/N and R could be established from this figure. However the maximum value of this "reduced pulse height" is seen to decrease with increasing R.

For 56 of the 206 events analysed above, one of the off-centre photomultipliers was also in operation. The acceptance cone of this photomultiplier was inclined at 7° to the vertical, and directed towards the centre of the array of scintillators. Of the 56 showers, 16 (29 per cent.) were accompanied by a pulse from this photomultiplier. This is to be compared with the corresponding figure of 11 per cent. for the central photomultiplier. We shall see later that this difference in detection efficiency may be understood qualitatively in terms of the integrated particle flux in the respective acceptance cones.

Angular distribution of the Cerenkov light

The angular distribution of the Cerenkov light about the shower axis is of considerable interest since it is capable of yielding information on the angular distribution of the electrons which emit the light. It is also necessary to know this distribution before results obtained with the light receiver can be properly interpreted. Accordingly, a series of investigations has been commenced in the Sydney experiment which will eventually lead to a well determined angular distribution of the Cerenkov light. Preliminary results of these investigations are presented here.

The direction of the shower axis was determined to within a few degrees by a timing technique which is described in an accompanying paper (Malos, 1958). As in the investigations on the radial dependence of the Cerenkov light, only the central photomultiplier was used. Since the acceptance cone has a half angle of only 3° and the axis of the cone is vertical, we may identify the angle of emission of the light (with respect to the shower axis) with the zenith angle of the shower axis.

Of 157 showers for which the shower size, core location and direction of incidence could be determined, 20 were accompanied by a Cerenkov pulse in the central photomultiplier.

Let us first classify the events according to the sign of H, the height at which the distance between the shower axis and the axis of the acceptance cone is a minimum. That is, H is positive if the shower passes "over" the detector before striking the ground, and negative if it strikes the ground before reaching the detector. We then find that the fraction of showers which were accompanied by a Cerenkov pulse is

0.20 for H positive and

0.11 for H negative.

This result again points to the importance of the integrated particle flux in the acceptance cone in determining the response of the light receiver.

In order to extract some information about the angular distribution of the Cerenkov light with respect to the direction of the shower

TABLE 1.—THE FRACTION OF SHOWERS ACCOMPANIED BY A CERENKOV PULSE AS A FUNCTION OF ZENITH ANGLE.

Zenith angle (degrees)	0—10	10—20	20—30	$\geq$ 30
Fractional rate for C/N $\geq$ 10	0.50—0.67 (6)	0.11—0.42 (2)	0.03—0.23 (1)	0.06—0.19 (1)
$\geq$ 20	0.50 (6)	0.05—0.16 (1)	0.03—0.17 (1)	0.06—0.19 (1)
$\geq$ 40	0.17 (2)	0.05 (1)	0.03 (1)	0 (0)
$\geq$ 80	0.08 (1)	0.05 (1)	0.03 (1)	0 (0)
Total number of events	12	19	30	16

The upper limits to the fractional rates are obtained by assuming that pulses of height not greater than the threshold value (0.6 volts) have, in fact, a height equal to the threshold value. Lower limits to the fractional rates are obtained by assuming that such pulses have zero height. The figures shown in brackets are the actual number of Cerenkov pulses recorded.

axis, we have considered only those showers where the skew distance between the shower axis and the axis of the acceptance cone is less than 20 m. We thus restrict our observations to particles close to the axis of the shower.

Eighty-seven showers satisfied this additional requirement, and of these 10 were accompanied by a Cerenkov pulse. Results for these showers are presented in Table 1, in which limits are given for the fraction of showers accompanied by a Cerenkov pulse for various ranges of zenith angle.

A most interesting feature of these results is the fact that Cerenkov light may be emitted at an angle of at least 30° to the shower axis. As we shall show in the next section, this is entirely consistent with the theoretically predicted angular distribution of the electrons in a shower. It is also in accord with numerous photographs of shower electrons in cloud chambers which have been obtained in the Sydney experiment and elsewhere.

Finally, it is possible to obtain some information on the form of the angular distribution of the light. To do this we have calculated upper and lower limits to the "average" value of the reduced pulse height (Cerenkov pulse height divided by shower size) for several ranges of zenith angle. The upper limit, $\bar{h}_{max}(\theta)$, is obtained by assuming that all events which were not accompanied by a Cerenkov pulse greater than the threshold value (0.6 V) were, in fact, accompanied by a pulse **equal to the threshold value.** The lower limit, $\bar{h}_{min}(\theta)$, on the other hand, was obtained by assuming that all such pulses had zero pulse height. The results of these calculations are presented in Table 2. Also shown is $\bar{h}(\theta)$, the arithmetic mean of $\bar{h}_{min}(\theta)$ and $\bar{h}_{max}(\theta)$. The form of $\bar{h}(\theta)$ is consistent with an expression of the form

$$\bar{h}(\Theta)\ \alpha\ \Theta^{-0.7\ \pm\ 0.3}$$

The significance of this result is discussed in the next section.

Discussion

The results presented in the previous sections clearly show the need for an independent and accurate determination of the direction of the shower axis in experiments using light receivers of the type described. This is particularly true if the detector is to be used in investigations

TABLE 2.—AVERAGE REDUCED PULSE HEIGHT (ARBITRARY UNITS) AS A FUNCTION OF Θ, THE ANGLE OF EMISSION RELATIVE TO THE SHOWER AXIS.

θ (degrees)	0—10	10—20	20—30	30—40
$\bar{h}_{min}(\Theta)$	23.6	6.6	2.9	1.3
$\bar{h}_{max}(\Theta)$	26.4	17.0	12.7	13.3
$\bar{h}(\Theta)$	25.0	11.8	7.8	7.3

on the longitudinal development of the showers.

It is also evident from the results presented in the preceding section that it is not correct to assume, as Jelley and Galbraith did in their early work, that the Cerenkov light is only emitted by particles travelling parallel to the shower axis. This result is quite reasonable when we consider that in electron-photon cascade theory the root mean square angle of scatter of shower electrons above 21 MeV, the threshold for Cerenkov emission at sea level, is ~ 10°.

A further point of interest is the form of the angular distribution of the Cerenkov light, which we have shown to be proportional to $\theta^{-0:7}$. Because the angle of emission of the

θ

Cerenkov light is small compared with the scatter of the electrons in the shower, we may identify this distribution with the angular distribution of electrons in the shower with energies greater than 21 MeV. Thus, our results, for distances less than 20 m. from the shower axis, are consistent with an angular distribution of the shower electrons of the form

$$N(\Theta)\,d\Theta \propto \Theta^{-0\cdot 7}\,d\Theta.$$

A distribution of this form is predicted by the electron-photon cascade theory (Greisen, 1956).

Finally, it is interesting to enquire how far existing air shower theory may be used to interpret the results of the present experiments. The only theoretical attempt which has been made to describe the Cerenkov light accompanying an air shower is that of Holdansky and Zhandov (1954), who make a number of assumptions which are certainly not valid for events such as those discussed here, where the shower passes within ~ 100 m. from the light receiver. A more profitable method of approach has been suggested by the author (Brennan, 1957), who has shown that the intensity of the light pulse, for a given angle between the shower axis and the axis of the acceptance cone, depends primarily on the average value of the electron density in the acceptance cone integrated over all heights. This approach enables one to take accurate account of the radial and angular distributions of the electrons in the shower.

Evidence which supports this approach may be found in the results quoted on the comparison between the responses of the two photomultipliers used in obtaining the radial dependence of the Cerenkov light. There it was found that the photomultiplier which was directed towards the array of scintillators, and hence saw on the average a higher integrated particle flux, responded with higher efficiency than did the central photomultiplier.

Acknowledgements

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Radiochemical Method for the Detection of Fast Neutrons

By Reginald Mills*

The possibility of using Fe56 (n,p) Mn56 nuclear reaction as a threshold detector for fast neutrons has been investigated. Radiochemical separation of the induced manganese activity allows a large quantity of iron to be irradiated, thus making the method sensitive to low density fluxes. The reproducibility of the method has been tested by irradiation with accurately-monitored fast neutron fluxes. The practicable lower limit of sensitivity has also been calculated.

Introduction

The nuclear reaction Fe56 (n,p) Mn56 has an energetic threshold of 3.0 MeV, but owing to the Coulomb barrier only neutrons of energies above $\sim$7.5 MeV are effective in producing the reaction. It should be utilizable therefore for the detection of neutrons with energy in excess of 8 MeV. The exposure of iron foils to a flux of neutrons would be the simplest method of using such a detector, but the low cross-section for the process (110 millibarns at 14 MeV) renders it fairly insensitive. In this investigation, the feasibility of using a large quantity of iron and extracting and concentrating the induced maganese activity has been explored. With this technique the sensitivity of detection should be considerably extended.

Experimental

Chemical separation procedure

The chemical isolation of the induced activity involves the use of carrier techniques to separate and concentrate manganese from the iron substrate.

In an initial series of experiments, compacted iron powder was used for the target material, but proved unsatisfactory for two reasons. Since approximately 400 grams of the powder was used in each irradiation, the dissolution and processing of the material involved difficult and lengthy chemistry. Elapsed time, from the end of irradiation to the final counting of the Mn56 activity, was of the order of five hours or two half-lives of the radioisotope. The second difficulty arose from the fact that appreciable manganese impurities are usually associated with iron and iron compounds. For example, "Analar" ferric chloride contains about 0.1% manganese and commercial iron powder undoubtedly has a higher percentage of the impurity. The presence of this manganese affects the accuracy of the detection process in two ways. Slow neutron activation of the impurity may produce Mn56, so losing specificity for the threshold energy. Secondly, the radiochemical yield, which involves comparing the analysis of the extracted manganese relative to a fixed amount of added carrier, cannot be stated with any precision.

The target system selected to obviate these difficulties consisted of a chloride complex of iron dissolved in isopropyl ether. Grahame and Seaborg (1938) have shown that in a water-ether system, manganese is completely retained in the aqueous phase when the ether is shaken with hydrochloric acid. The troublesome manganese impurity can therefore be virtually removed. In addition, the separation is relatively simple and rapid if the ethereal solution is used as the starting material. Isopropyl ether will absorb about 160 grams of iron per litre of solution from 8N hydrochloric acid, and it was found convenient to use about two litres of solution. The use of moderating liquid such as isopropyl ether has the disadvantage that the fast neutron flux may be appreciably attenuated in contrast to the use of iron powder, where the effect would be very much smaller. This attenuation effect can, however, be minimised by having the containing vessel in the form of an annular section. In our experiments, where the width of the container was 4cm., the calculated maximum attenuation is less than 10 per cent. The chemical isolation procedure which is outlined below is relatively simple, and takes about one hour only.

After irradiation, a known amount of manganese chloride carrier (*ca.* 30 mgm) is added to the ethereal solution. It is then shaken with 500 ml. portions of 8N hydrochloric acid. In this step, essentially all of the manganese is extracted into the aqueous layer plus a small fraction of the ferric chloride complex. The latter is removed by shaking with a fresh portion of isopropyl ether which has been saturated with 8N hydrochloric acid. The acidic aqueous layer is neutralized with strong sodium hydroxide, and manganese hydroxide is precipitated. The precipitate is filtered, washed, and dissolved in 1N sulphuric acid. A little sodium bromate is added, and, upon boiling for 5-10 minutes, manganese dioxide is precipitated. The solid is filtered off, washed with acetone and dried in an oven. It is then ground, transferred to trays and placed under an end-window Geiger counter. After counting, the precipitate is analysed for manganese content by the potassium iodate method (Vogel 1951), and this analysis is compared with the weight of carrier added, so giving the radiochemical yield. In our experiments, this yield averages 80%.

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The original ether solution can, of course, be used for a large number of irradiations, but it has to be topped up with a small amount of fresh solution each time. The small loss of iron during the initial extraction with acid has been measured and averages 0.3%. However, an average retraction in volume of the ether solution by about 0.6% occurs also, and this has a compensating effect, so that the iron concentration remains reasonably constant. The above effects can probably be neglected for most purposes.

Irradiation procedure

Solutions of ferric chloride in isopropyl ether were irradiated with fast neutron fluxes to test the reproducibility of the method, and also to enable an estimate of its absolute sensitivity to be made. These neutrons were obtained as products of the nuclear reactions Li7 (d,n) and H3 (d,n) He4, the latter reaction giving a monoenergetic beam of 14.1 MeV neutrons. Cockroft-Walton H.T. sets were used to produce the above reactions. For the Li7 (d,n) process, the flux was monitored by a Hornyak scintillation button and, in addition, by iron disks. In the H3 (d,n) He4 case, the recoil alpha particles were counted in a scintillation counter with known geometry, which enabled the absolute flux to be determined precisely. For these irradiations, the ethereal solution containing the iron was contained in a polyethylene bottle and placed about 30 cm. from the neutron-emitting target.

Results

In all experiments the activities measured had pure half-lives of ~2.6 hours, emphasising the method's complete insensitivity to gamma and slow neutron activation.

Data from the test irradiations are given in Tables 1 and 2 below.

TABLE 1.

Irradiation by fast neutrons from Li7 (d,n) reaction:—

Run No.	Activity (cpm).	
1	13355	Standard
2	12581	deviation
3	12597	±2%.
4	12830	

TABLE 2.

Irradiation with 14.1 MeV neutrons from H3 (d,n) He4 reaction:—

Run No.	Activity (cpm).	
1	2593	Standard
2	2751	deviation
3	2610	±4%.
4	2517	
5	2589	
6	2404	

In both tables the activities have been normalised against a monitored count, and have been corrected for decay and radiochemical yield.

Discussion

Examination of the data given in the above tables shows that this detection technique is capable of determining fast neutron events with a relative precision of ±4%. In fact, the precision appears to be limited mainly by that of the independent monitor. Since the recoil alpha monitor gives a record of the absolute flux of fast neutrons, an estimate can be made of the lowest flux that can be practicably measured with the system. If two litres of solution (containing 400 grams of iron) are subject to an incident flux of 80 neutrons/cm.^{2}/sec. for 1 hour, then a counting rate of 10 c.p.m. will be obtained using an end-window Geiger counter. The radioactive sample is placed in close proximity to the window of the counter so that the counting geometry is slightly less than 2π.

Some advantages of the method may be summarised as follows:

(a) It is a threshold detector and records only neutrons with energy in excess of 8 MeV.

(b) It is sensitive to very low density fluxes.

(c) It is completely insensitive to gamma and slow neutron activation and can therefore be used, unshielded, close to particle accelerators.

(d) It requires no special instrumentation and is inexpensive.

There are also some obvious disadvantages.

(a) It does not give instantaneous indication of flux, and some hours must elapse before the data are obtained

(b) It gives only relative measurements of flux, although, when calibrated, it should be able to give absolute data.

Acknowledgements

I wish to record the helpful advice of Dr. J. Carver, who originally suggested the development of the method.

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SECTION 5

Associated Techniques

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Some Temperature Effects of Geiger-Muller Tubes

By R. E. Belin*

A brief survey of reported temperature effects in Geiger tubes is followed by a description of experiments carried out on organic quenched tubes with oxide-coated cathodes. The count rate effects observed are associated with the semi-conductor oxide film on the cathode since they are absent in tubes with cathodes cleaned by reduction in hydrogen at 400°C. They are also absent in halogen quenched tubes. In all tubes repeatable changes in threshold voltage with temperature variations and plateau improvements with a temperature rise may be associated with the processes of sorption.

INTRODUCTION

During a radio-activity survey of the geothermal areas of New Zealand in 1951 the author observed an increase in counting rate above normal background, when an alcohol-argon tube was subjected to a temperature of 150°C. The counting rate was observed to decrease with time. This tube, as well as others, had been used extensively at 100°C., in the field, without any changes being observed in their operating characteristics.

In view of an extended program in the geothermal area and because of the inconsistency of available reports it was decided to evaluate the characteristics of both organic and halogen-quenched tubes at elevated temperatures.

Halogen-quenched tubes at elevated temperatures have received little attention. Clark (1955) has shown that with special cathode preparation they can be operated up to 200°C. with little change in characteristics.

The effects of changes of temperature on self-quenching Geiger tubes have been investigated by numerous workers and with alcohol-argon tubes inconsistent results have been reported. Parkash and Kapur (1950) and Loosemore and Taylor (1950) showed that in all cases within the temperature range 20 to 100°C. the threshhold voltage rose, the plateau shortened, and the plateau slope increased. Fujioka, Kita and Minakawa (1951) reported that a poor tube outgassed at 800°C., before filling, showed improved characteristics with increase in temperatures until at about 200°C. the tube went into a continuous discharge. For a tube with a good plateau at room temperature the plateau improved up to 79°C., then began to deteriorate. Plateaux taken at various temperatures after such a tube had been raised to 105°C. were variable and showed some deterioration from previous heating. Blanc and Viste (1955) showed that the threshold voltage rose linearly with temperature up to 120°C. and thereafter parabolically. Kimura (1950, 1951) and Puri and Gill (1956) observed that an increase in temperature produced an increase in the background counting rate. Kimura (1951) showed that the counting rate increase was proportional to the rate of rise of temperature and dropped to normal following a slight drop in temperature. Plateaux and threshold voltages before and after remained unaffected. If a tube was raised to and held at a certain temperature the counting rate decreased to normal exponentially with the square root of time. Puri and Gill (1956) suggested that the counting rate phenomena as well as threshold voltage changes were closely associated with adsorption and desorption of the tube gases.

CHOICE OF GEIGER TUBES AND METHODS OF MEASUREMENT

Kimura's (1951) observations on thin-film field emission (Malter 1936) of oxide-coated cathode surfaces prompted an investigation of two types of organic-quenched Geiger tubes: (1) with oxide-coated cathodes and (2) with pure metal cathodes.

For type 1 tubes, copper and stainless steel cathodes were selected while for type 2, copper cathodes cleaned by hydrogen reduction at 400°C. were chosen. For the halogen-quenched tubes, type 3, the cathodes were of 18/8 low-carbon stainless steel or high-purity aluminium.

The organic-quenched tubes were filled with 1 cm. Hg of ethyl alcohol and 9 cm. Hg of argon; ethyl formate replaced the alcohol in some tubes. A typical filling for the halogen-quenched tubes was 0.06 cm. Hg of bromine and 27 cm. Hg of argon; here the procedure of Le Croissette and Yarwood (1951) was adopted. For the organic-quenched tubes, at temperatures up to 150°C., all-metal construction was employed; for experiments above 150°C. the cathodes were enclosed in a glass envelope. All tubes, after evacuating to a pressure of 10^{-3} mm. Hg and just prior to filling, were completely flushed with the inert gas at a pressure of about 20 cm. Hg.

Threshold voltages at constant overvoltage (actually at constant pulse height) and the counting rate were continuously monitored during the experiments. Plateaux at constant temperatures were measured using a plateau plotter. A cathode ray oscilloscope was used for visual examination of the Geiger pulses.

Sudden temperature changes were effected by immersing the tube, during operation, into

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a constant-temperature bath, either oil or water. An air oven was also used.

RESULTS

The organic-vapour-quenched tubes with oxide-coated cathodes exhibited the following effects:

(i) Upon heating the tube for the first time to a temperature T ($T \leqq 150$°C.), the background counting rate rose to some high value, N_0, above the natural background counting rate. The value of N_0, was dependent upon the temperature rise.

(ii) If the tube temperature was maintained at T, the count rate N_1, at time t, above natural background followed the relation

$$N_1 = N_0 t^{-a} \qquad (1)$$

where "a" was found to be approximately 0.8, if t is in minutes. The time taken, at a temperature of 100°C., for N_1 to become small and insignificant was usually around 4 hours, although this was found to vary, as N_0 was different for each tube. This count rate variation occurred **once** only in the life of the tube. (A tube whose count rate has become normal at the natural background counting rate will subsequently be referred to as "seasoned.")

(iii) A further increase of temperature above T, but still below 150°C., produced a further increase in counting rate which also behaved as set out in paragraphs (i) and (ii) above.

(iv) The temperature of a seasoned tube could be varied anywhere between ambient (20°C.) and the seasoning temperature without any changes in counting rate.

(v) When the temperature was raised to a constant value between 150°C. and 230°C., the counting rate behaviour was as described above, but it never quite returned to the natural background counting rate value. The value to which it returned was dependent upon the temperature above 150°C. Small pulses of constant amplitude (about 1 mV at the Geiger threshold) were observed. Their threshold voltage was several hundred volts below the Geiger threshold. Their amplitude and rate seemed dependent upon the tube voltage.

(vi) At 230°C. the counting rate of these tubes suddenly rose to an extremely high value, as if the tube had broken into a continuous discharge. A tube whose temperature had exceeded 230°C., when again at ambient temperature, behaved normally except that the plateau had become shorter and steeper than before. The plateau quality at ambient temperature became worse the longer the tube temperature was maintained at or above 230°C.

(vii) When the tube was first raised to temperature T the threshold voltage, V, began to rise according to the empirical relation

$$\frac{dV}{dt} = \frac{K}{t^b} \quad \text{for } t > 0, \qquad (2)$$

where b was found to approximately. = 1. t is in minutes. K was dependent upon the temperature rise as well as other apparently variable factors within the Geiger tube. The permanent threshold voltage rise was found to vary from tube to tube and to be in the range of 10 to 50 V.

Combining equations (1) and (2)

$$N_1 = C\left\{\frac{(dV)}{(dt)}\right\}^{a/b} \quad \text{for } t > o \qquad (3)$$

Thus N_1 is a maximum when $\frac{dV}{dt}$ it a maximum.

(viii) For a seasoned tube, the temperature could be raised from ambient to the seasoning temperature T ($T \leqq 150$°C.) with only a small increase in the threshold voltage. This returned again to normal when the temperature returned to ambient. When T = 100°C. the threshold voltage increment, with respect to the threshold voltage at ambient temperature, was approximately 5V and was proportional to the temperature rise. The rate of change of these voltage changes appeared to follow an equation similar to (2).

(ix) The plateau lengths of all seasoned tubes rose linearly with temperatures up to 100°C., and above this the improvement rate decreased.

(x) Raising the temperature of the anode to a red heat left the counting rate unaffected.

Figure 1 is a typical record showing the manner in which the threshold voltage varies with time, at a constant temperature, and its relation to the counting rate. Each pip gives the summation of 1000 counts. After a short sojourn at ambient temperature, between time 92 and 160 minutes, the tube temperature was again raised to 150°C. and the threshold voltage rose to the same value as at time, 92 min., just before the temperature was dropped.

It and the counting rate continued to follow equations (1) and (2), until 200 min., when there was an unaccountable increase in the counting rate associated again with a further threshold voltage increase. Note the manner in which the threshold voltage drops after a sudden lowering in temperature at 92 min. The threshold voltage of a seasoned tube, when suddenly raised in temperature, follows a curve which is the reciprocal of that shown between 92 and 160 min.

The hydrogen cleaned copper cathode tubes exhibited the following effects in common with the oxide coated cathode tubes:

(a) The continuous discharge at 230°C.

(b) The small variations in threshold voltage with temperature.

(c) Linear plateau improvement with rise in temperature.

(d) No effect when the anode was raised to a red heat.

These tubes gave no change in counting rate over the temperature range 20° to 230°C.

The maximum temperature at which the halogen tubes were examined was 150°C. Of the

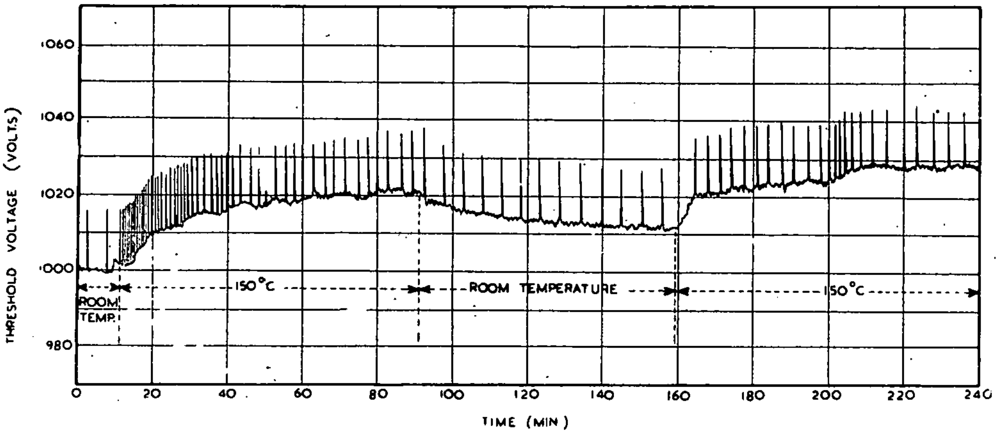

FIGURE 1.

above effects only the following were observed:

(a) Variations in threshold voltage with temperature.

(b) Linear increase in plateau length with rise in temperature up to 150°C. (Belin 1957).

The increase in counting rate upon first heating was absent.

DISCUSSION

The increase in counting rate observed when the temperature of the tube is first raised, and its subsequent hyperbolic decrease with time, differ from Kimura's (1951) observation. In his case the counting rates decreased exponentially with the square root of time and he makes no mention that this effect occurs only once in the temperature history of a tube. Kimura (1951) has shown that the increase in counting rate is not due to spurious pulses generated by a preceding pulse.

Since the hydrogen-cleaned copper cathode tubes do not show the changes in operating characteristics shown by tubes with oxide-coated cathodes, it is reasonable to assume that the difference can be attributed to the semiconductor oxide film. The possibility of reactions at the anode is dismissed by the anode heating experiments.

Fergusson (1953) found that rubbing the cathode surface or a brief application of a high voltage discharge through a Geiger tube produced a high counting rate which in both cases decayed hyperbolically with time. The rate was accelerated at temperatures above ambient. Kramer (1952) showed that, after metal surfaces were rubbed, electrons were given off at a rate which obeyed equation (1). He found that for metals $a = 1$.

Malter (1936), using certain oxides as targets in a cathode ray oscilloscope, found, after cutting the primary beam, that the collector current decreased hyperbolically with time. He called this thin film field emission, to which Kimura (1951) refers. Bardeen and Brattain (1949) report that the surface dipole, hence the contact potential of a semi-conductor, is altered by absorption and desorption of ions on the surface and this in turn is affected by the ambient gaseous state. They also report that electrons can become trapped on germanium containing minute traces of impurities and these subsequently escape at a rate which is exponential with time.

All these reports indicate that charged particles can become trapped on the surface of a semi-conductor, and that these may subsequently escape hyperbolically with time. This would explain the counting rate behaviour observed, provided the traps were destroyed after releasing the charged particles.

The relation between the counting rate and the rate of change of threshold voltage (equation (3) suggests a process of desorption of quenching vapour from the cathode accompanied by a change in contact potential and threshold voltage. (If quenching agent is added at a hyperbolic rate to the sensitive volume of a tube then, within limits, the rate of rise of threshold voltage will also be hyperbolic.) Calculation tends to eliminate either the quenching agent or argon from being released permanently in sufficient quantity to produce such a large increase in the threshold voltage.

The essential physical difference between the two types of organic-quenched tubes is the complete absence of the oxide surface in the hydrogen-cleaned one. Thus if oxygen, either chemically adsorbed by the oxide or from the oxide (thickness unknown), were released from the oxide-coated cathode, then all the above effects, as well as a source of ions, are explicable.

The small repeatable changes in threshold voltage accompanying temperature changes presumably result from adsorption and desorption of the quenching vapour, either organic vapour or bromine. This view is supported by Puri and Gill (1956). Desorption of a quenching agent with a temperature rise explains the improvement in plateaux of seasoned as well as all other types of tubes examined. (With all the fillings used an increase in the pressure of the quenching agent produces a longer plateau but higher threshold voltage.)

The sudden tremendous increase in counting rate at 230°C. is not satisfactorily explained by consideration of Richardsons' law for thermionic emission, Fujioka, Kita and Minikawa (1951) observed a similar effect at approximately the same temperature. They suggested that at this temperature gases are liberated from the walls of the tube. This is unlikely as the process would ultimately cease, for after 2 hours continuous discharge at a temperature greater than 230°C., there appeared to be no diminishing of the counting rate, although a small plateau still remained at ambient temperature. However, the deterioration in the plateau length after a tube had been subjected to such treatment was readily explained by a spectographic analysis of the Geiger tube atmosphere. This showed the presence of an appreciable amount of water, the amount being directly related to the length of time the tube was maintained at the high counting rate above 230°C.

CONCLUSIONS

The removal from the cathode of the oxide surface by reduction with hydrogen at 400°C. has shown that:

(a) There is little change in operating characteristics of such a tube until at a temperature of 230°C. the tube goes into a continuous discharge.

(b) The oxide semi-conductor film in the tubes with oxide-coated cathodes, in some unexplained way, is directly responsible for the count rate variation and threshold voltage increase observed in these tubes, when heated for the first time.

The improvement in plateau length with rise in temperature, and the small variations of threshold voltage accompanying temperature changes of all types of tubes—whether quenched with organic vapour, or a halogen—can be explained by sorption. Desorption gives an increase in plateau length and threshold voltage.

ACKNOWLEDGEMENTS

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Inaccuracy of the Colloidal Gold Method for Determining Liver Blood Flow

By J. McRae, M. Playoust, R. Boden and C. R. B. Blackburn*

Methods of determining liver blood flow are reviewed briefly, in particular the method using the blood disappearance rate of injected radioactive gold colloid (Au 198). This method assumes a high extraction by the liver at one passage. Because of anomalous results, an experiment was designed to measure the extraction in humans, using simultaneous estimations of radioactivity of blood from the brachial artery and from the hepatic vein following intravenous injection of the radiogold colloid. The maximum extraction rate varied between 62 and 15 per cent. This method of determining liver blood flow using the colloid at present available from Harwell is unsound. It is suggested that there may have been a change in the colloid particle size since the first experiments with the material some years ago.

INTRODUCTION

Bradley and his co-workers (1945) introduced the bromsulphalein (BSP) method for measuring hepatic blood flow. This technique involves catheterisation of the hepatic vein, and the maintenance of a constant peripheral blood level of the dye by an infusion at a constant rate. Simultaneous blood samples are then taken from the hepatic vein and from a peripheral artery, and the BSP concentration determined. Hence, with the knowledge of the rate of BSP infusion, the hepatic blood flow can be calculated using the Fick principle.

This method has been quite a valuable one, but is tedious both for the subject and for the observers. There also remain some doubts about variation in the hepatic vein BSP content depending on the exact position of the catheter. In patients with severely damaged livers, the BSP extraction is too low for the method to be used at all.

Dobson and Jones (1952) described a method of estimating hepatic blood flow using intravenously injected colloid particulate matter (in this case chromic phosphate labelled with P32). Briefly, the method consists of measuring the radio-activity of serial blood samples after intravenous injection of the labelled colloid, and plotting the results on semi-logarithmic paper as a function of time. A multi-component exponential curve is obtained, the initial portion of which closely approximates to a simple exponential which may be represented by the equation $C = C_o e^{-kt}$ where C is the concentration at any time, Co is the initial concentration and k is a constant. If it is assumed that all the colloidal particles are removed by the liver and the spleen at one passage, the slope of this straight line represents the fraction of the total blood volume perfusing the liver per unit time, k being termed the disappearance constant. k may be derived from the equation

$$k = \frac{\log_e 2}{t_{\frac{1}{2}}}$$

where $t_{\frac{1}{2}}$ is the half time of disappearance, and the hepatic blood flow is then estimated by multiplying the blood volume by k.

There remains the question whether the colloid particles are removed by the liver (or spleen) at the first opportunity. Using chromic phosphate, the overall efficiency of removal was of the order of 90% using the dog, rabbit, mouse and chicken, (Dobson et al. 1952, Sheppard et al. 1951, and Little et al. 1953). Dobson and Warner's result for k in 29 normal young men was 0.287 ± 0.007 minute^{-1}, giving a liver blood flow of 1.5 — 1.8 litres/minute (Dobson et al. (1953)) which corresponds well with the figure obtained using the BSP method of Bradley et al. (1945) and Sherlock et al. (1950).

However, the preparation of labelled chromic phosphate of suitable particle size is a laborious procedure, and it seemed to Vetter, Falkner and Neumayr (1954) that a further simplification would be to use a colloid readily at hand such as radio-active gold (Au198), which is supplied from the Radiochemical Centre, Amersham, England. The average particle size is not determined on each batch, but is stated from electron microscopy to be 200-250 A° with a small percentage of particles as small as 50 A° and as large as 600 A°. Because this is a gamma-emitting isotope, external counting over the liver may be used as an additional method of determining hepatic blood flow. Also, instead of serial blood samples, it is possible to obtain the blood disappearance curve using a counter placed between the legs.

In three cases only (one cirrhotic and two normals) these investigators estimated the extraction rate directly by hepatic vein catheterisation, and these had extractions of 87, 78 and 81 per cent. respectively for the first five minutes following injection of the colloid. They noted that these figures were in striking agreement with the results of animal experiments using chromic phosphate (Dobson and Jones 1952) and those of liver perfusion experiments with radiogold performed by Little and Kelly (1953).

We have attempted to use the radiogold method for the determination of hepatic blood flow, but because of a number of anomalous results, it was decided to verify the alleged high extraction rate of the colloid by the liver.

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* Clinical Research Unit, Royal Prince Alfred Hospital and the Department of Medicine, University of Sydney. Manuscript received, March 27, 1958.

METHOD

Five patients were studied by placing a Cournand arterial needle into the brachial artery, and a cardiac catheter into the hepatic vein via the median cubital vein with the aid of fluoroscopy. Tracings were taken as a routine for both free and occluded hepatic vein pressures.

Between 10 and 15 microcuries of Au198 were injected intravenously, and serial blood samples were taken from both hepatic vein and brachial artery. The samples were withdrawn simultaneously, with the proviso that allowance was made for the delay in appearance of the hepatic vein blood at the end of the catheter. In most of the cases samples were taken at 2, 3, 5, 7, 10, 15, and 20 minutes after the colloid injection.

The whole procedure was combined in three of the five cases with the determination of liver blood flow using the BSP extraction method. The position of the catheter in the hepatic vein was confirmed in all cases by fluoroscopy.

The patients investigated were as follows:—

Patient 1. Female, aged 32, with established cirrhosis.

Patient 2. Female, aged 21, with progressive hepatitis.

Patient 3. Male, aged 32, with gastric ulcer —(no clinical or biochemical suggestion of liver disease; normal BSP retention, normal flocculation tests, etc.).

Patient 4. Male, aged 40, with alcoholic cirrhosis.

Patient 5. Female, aged 30, with refractory anaemia (no clinical or biochemical suggestion of liver disease; normal BSP retention, normal flocculation tests, etc.).

RESULTS

In the five patients tested the maximum Au198 extraction varied from 62 to 15 per cent. The two normals were 31 and 61 per cent., while the three with liver disease were 21, 15 and 32 per cent. respectively. In four of the patients (1, 2, 3, and 5) the maximum extraction was the one calculated from the first pair of blood samples; in none of these was the extraction observed to remain constant for any period of time. The extraction in the fourth case was relatively constant for about 10 minutes (actually, it rose a little at first).

After a varying period of time (7 minutes to more than half an hour) the extraction fell to zero.

The figures which we obtained for the extraction of the gold colloid are shown on the accompanying graph.

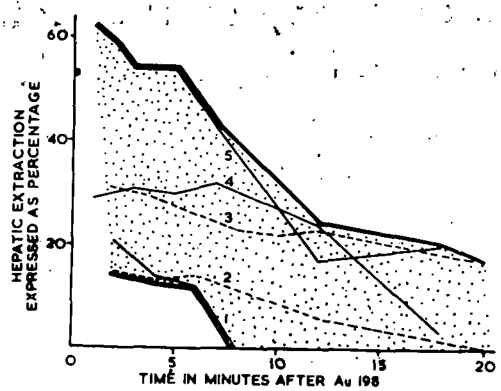

FIGURE 1:—Graph showing the extraction of colloidal gold by the liver. The results for the five individual patients are represented by the thin continuous and interrupted lines, while the shaded area indicates the whole range of values obtained.

DISCUSSION

It seems from the above results that the extraction of colloidal gold is considerably less than the 80 per cent. reported by Vetter and his co-workers. In other words, most of the colloidal particles are not removed by the liver at one passage, as has been assumed by other investigators.

Furthermore, the extraction rate in our series varied from person to person, so that a constant correction factor could not be introduced to permit an estimate of hepatic flow. Even in the one subject there is no certainty that the extraction will remain constant for some 8-10 minutes after the injection of the colloid, this being the part of the curve which is used for the calculation.

The fall in the extraction to zero after 20 minutes or so was not unexpected, because this has been reported by other investigators (Dobson and Jones, 1952; Dobson et al. 1953; Little and Kelly, 1953). It is assumed that this is due to residual very small colloid particles which are not accepted by the liver's phagocytes.

It is not known at this stage whether the low early extraction which we have demonstrated is due to a change in the particle size of the colloid prepared at Harwell; enquiries are at present being directed along these lines. In any case, it seems that the preparation of colloid at present available is unsuitable, and that determinations of liver blood flow using this material are gross underestimates.

We propose to determine if there is a significantly lower extraction of colloid by diseased livers compared with normal ones. If this is found, it will throw doubt on the possibility of ever finding a suitable colloid which will give accurate results for hepatic flow in the presence of liver disease.

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The Localisation of Placenta using Radio-isotopes

By E. P. George* and D. McGrath†

A method of determining the position of the placenta using an isotope tracer is described. The method relies on the ability to distinguish the increased activity over the placental blood pool. The method has been tried in 12 cases, in which one erroneous result was obtained. The radiation exposure to mother and foetus is calculated and found to be less than 0.1 r. The value of the method is critically discussed in relation to other established methods of placental location.

INTRODUCTION

Placenta praevia still ranks high as a cause of maternal and foetal death, and in its management accurate information concerning the placental position is obviously of prime importance. Three methods of localisation are available to the obstetrician

(i) clinical judgment.
(ii) low voltage diagnostic X-ray filming.
(iii) tracer radioisotope studies.

In the first method, the clinician forms his impression of the placenta position from observations of the posture and movement of the foetus, the occurrence of ante-partum haemorrhage, etc.

In the second method, developed in 1949 by Reid (1949a, 1949b) three routine films are exposed using low kilovoltage X-rays — recumbent antero-posterior, erect lateral, and recumbent soft-tissue lateral. Direct visualisation of the soft tissue shadow of the placenta is generally obtained. Frequently, this is helped by calcification which sometimes occurs in the placenta.

In the third method, developed by McClure, Browne and Veall (1952) a small amount of radioactive isotope is mixed in the maternal blood stream. External scanning of the abdomen with a nuclear radiation detector is then used to locate the concentration of radioactivity in the placental blood pool.

It was decided to investigate the X-ray and radio-isotope methods in order to determine their value in the management of placenta praevia. In this paper a preliminary report is given of the radioisotope technique only, because the necessary X-ray equipment is not yet available.

EXPERIMENTAL METHOD

In the work of McClure Browne and Veall in London, radioactive sodium (Na^{24}) was used. Because this isotope has a half-life of 14 hours, it was not available for use in Sydney. Some other tracer mixing freely with the blood stream and easy to detect had to be chosen. Supplies of radio-iodinated human serum albumin (RIHSA) were made available by the Radiochemical Centre, Amersham, through the Commonwealth X-Ray and Radium Laboratory. It was decided to use this material, because of its ease of detection, and because the radio-iodine, being protein bound, would not be rapidly absorbed by the maternal or foetal thyroid. In order to ensure even further a minimisation of thyroid uptake 131, Lugol's iodine was administered orally several hours before the test.

Initially, 50 microcuries of RIHSA were injected into an antecubital vein. Later, this was reduced to 25 microcuries. After allowing 10 minutes for adequate mixing in the blood stream, the abdomen was scanned, with the patient lying on a special scanning couch. This consisted of a shielded scintillation counter which could be moved over calibrated scales in the horizontal and vertical planes.

The counter was surrounded with a lead shield, 1in. thick. At the sensitive end of the counter, in front of the sodium iodide crystal, was a cylindrical collimator 3in. in diameter and 3in. long, down the centre of which was a 1in. diameter hole.

Readings of the counting rate were then taken on a counting rate meter on a grid with 2cm spacing. The interval was dropped to 1cm. in regions of high counting rate, or where its rate of change with distance was high.

Contours of equal counting rate on the grid were then drawn (isocount curves), and the position of the placental blood pool was identified with the point of maximum activity. A typical set of isocount contours obtained in this way is shown in Figure 1. From these contours, it was concluded that in this case the placenta was normally placed at the top of the fundus uteri, a conclusion that was later confirmed at delivery.

The anterior/posterior location of the placenta was then investigated by taking readings at various distances over the region of high counting rate. For this purpose, the directional collimator was removed. Using the inverse square law, it was then possible to get some indication of the A/P location. This was also checked in some cases by taking a series of readings with the collimated counter placed horizontally at the side of the patient, and pointing inwards towards the mid-line. The counter was moved vertically in a plane containing the predicted placental centre. A typical result obtained by this method is shown in Figure 2. This diagram indicates a posterior location of the placenta.

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RESULTS

Eleven cases have been investigated so far by this method. This admittedly small number represents our first clinical trials of the method. The results were sufficiently promising to warrant an extension of the work to improve the statistical accuracy of the results, and this has been arranged. In the meantime, it was thought this interim report would be of interest.

In these 11 cases, the localisation with RIHSA was carried out any time following the 32nd week of pregnancy. The cases where then followed through to delivery, and the exact location of the placenta determined:

(i) by actually seeing the placenta in situ at Caesarian section. This occurred in two of the 11 cases, or

(ii) by inserting the hand into the uterus and determining the site of the placenta immediately after the birth of the baby. This was done in six cases.

The remaining three cases were delivered vaginally, but manual exploration of the uterus was not done. The results in these three cases were therefore unproven. The results are summarised in Table 1.

TABLE 1.—SUMMARY OF RESULTS

Assessment	No. of Cases
Correct	7
Incorrect	1
Unproven	3
Total	11

In the one case classified as incorrect, the degree of error was very small. The isotope result was definitely placenta praevia with the placenta slightly to the right of the mid-line. At Caesarian section the placenta was defined to be central placenta praevia.

The three unproven cases were delivered vaginally, indicating the absence of placenta praevia. In none of these cases was placenta praevia indicated isotopically, and thus these three results are not inconsistent.

RADIATION HAZARDS

In the application of isotopic tracers in pregnancy, it is important to consider the radiation hazards involved. These are of two kinds:—

(i) thyroid irradiation due to selective uptake of iodine 131 liberated from the RIHSA by albumin metabolism, and

(ii) general body irradiation.

The uptake of iodine in the maternal thyroid was checked by taking frequent readings of radioactivity over the patient's thyroid gland. Due to the pre-medication with Lugol's iodine, the uptake was found to be always less than 1 per cent. of the administered dose. With an administered dose of 25 microcuries, this therefore means an upper limit of 0.25 microcuries uptake in the thyroid. The foetal uptake in the thyroid will be less than this by an amount which depends on the permeability of the placental barrier to iodine. From these figures, it follows that the upper limit to the irradiation received by maternal and foetal thyroid may be taken as 0.25r. From the known amount of iodine 131 administered, it follows that the mean level of the whole body irradiation is 0.05 roentgens, and this will also be the dose to the maternal gonads. Vascular organs, such as liver and spleen, will receive higher levels than this, up to 0.2r.

Because the albumin does not cross the placental barrier to any appreciable extent, the foetal irradiation both to whole body and to gonads will be reduced by a factor of about 10 times, and will be of order 0.005 r—of the same order as that received from natural background radioactivity in one week.

These figures indicate that provided the thyroid uptake of detached iodine 131 is adequately blocked with Lugol's iodine, or any other recognised method of administering iodine, the radiation hazards are negligible.

DISCUSSION

In their pioneer work on the isotope method, McClure, Browne and Veall (1953) obtained an accuracy of 85 per cent. correct predictions. In this work Na24 was used, which necessitated rapid scanning of the abdomen because of the rapid diffusion of sodium from the circulation. The scanning was done with an uncollimated Geiger counter. These workers have made improvements in technique along the lines described in this paper and Veall (1957) has now reported an accuracy of over 90 per cent. Our own results are not inconsistent with this figure, though clearly more results are required in order to make our figure more precise. Trials have just started on a more extensive series of cases, which will be reported in due course.

The accuracy obtainable with X-ray placentography is quite high. In a recent paper Watson et al. (1957) reported an accuracy of 94 per cent., similar to the figures of Percival and Murray (1955) who recorded an accuracy of 96 per cent., while Hartly (1954) reported an accuracy of 98 per cent. These figures clearly leave little room for improvement, and no claim has been made that the isotope method is more accurate than placentography. Indeed, it will be hard to equal the accuracy attainable in placentography.

On the other hand, there are many cases which are unsuitable for placentography, such as patients with marked hydramnios or an undiagnosed soft tissue pelvic tumour. Also, Donald (1955) has reported that multipara with lax abdominal walls are unsuitable.

In attempting to assess the possible place in the future of the isotope method of placental localisation, we have formed the opinion that placentography will probably prove of greatest value, with the isotope method playing a supplementary role in those cases unsuitable for placentography, or where the X-ray diagnosis is in doubt for any reason. This opinion is partly based on the fact that while technical complexities of each method are comparable, there are a great many trained radiologists for every clinical isotopist or medical physicist. However, we believe there is a definite place for the isotope method, and that a combination of the two techniques could see the accuracy of placental location pushed very close to the 100 per cent. mark.

In this regard it is of interest to note that with modern counting methods, the amount of isotope used can be cut down to such an extent that the radiation dose can be made very small compared with that used in placentography. In arriving at a figure for the latter technique, we have assumed that the dose received is similar to that in pelvimetry. According to the M.R.C. report (1956), we find that the dose to foetal gonads in pelvimetry is 2.7 r in one hospital known to reduce irradiation to a minimum. This is a good many times greater than that in the isotope method, and might be taken as an indication for the isotope method if at any time it was felt desirable to reduce the foetal irradiation to an even lower level than that attained generally as present.

CONCLUSIONS

The following conclusions have been drawn:

(i) Isotopic localisation of placenta is a safe procedure.

(ii) The method described has good accuracy.

(iii) The accuracy is not likely to be better than that reported for placentography.

(iv) Provided the thyroid gland is blocked by the administration of stable iodine, the radiation dose received in the isotope method is lower than that in placentography.

(v) At present placentography will probably be the method of choice, with the isotopic method a useful reserve procedure for those cases not suitable for placentography, or where the X-ray result is in doubt.

ACKNOWLEDGEMENTS

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Use Of Radioactive Tracers in Cardiac Investigations

By E. P. George, J. Hickie, F. A. Rocke and W. A. Seldon *

Using iodine 131 serum albumin, a technique is described in which by placing a counter over the chest the passage of a tracer through the heart and great vessels may be recorded. From the curve so recorded the volume of blood pumped per minute by the heart may be determined. In a series of cases, results obtained by this method have been compared with results obtained by the more usual method. The standard deviation of both determinations appears to be of order of 10 per cent. The theory of the method is presented, and the merits of the method are discussed critically. An inversion of the method in which the dilution curve of a tracer injected directly into the heart is measured peripherally has been tried. This has proved of value in the diagnosis of congenital heart disease and is briefly discussed.

INTRODUCTION

The use of the dye-dilution curve as a means of studying cardiac function was first suggested by Stewart in 1897. In this method, a small amount of dye is injected into the venous circulatory system at a convenient point. At some other point in the system the concentration of the marker is measured as a function of time. From this curve, knowing the amount of marker originally injected, it is possible to determine a number of important physiological factors, such as circulation time, cardiac output (i.e., the average rate of circulation of blood through the heart in litres per minute), pulmonary blood volume, and the shape of the curve may also be of diagnostic significance. Kinsman, Moore and Hamilton (1929) brought the method to the point of practical application in man, and it has now become one of the accepted methods of measuring cardiac output. The method is difficult to carry through, and the equipment can be somewhat involved.

The use of a radioactive marker in place of the dye was first tried by Nylin and Celander (1950), who used phosphorus 32. In this method an arterial puncture is required and a series of blood samples collected over a period of five minutes. These samples were counted with a beta-counter, because phosphorus 32 is a pure beta emitter.

With the availability of radio-iodinated human serum albumin, the iodine 131 in which is a gamma emitter, it became possible to dispense with the arterial puncture and to rely purely on external counting with a scintillation counter. (Veall et al. 1954, Pritchard et al. 1955.)

Experience with the latter method is reported in the following paper.

THEORY OF THE DILUTION CURVE

In order to obtain an expression for the form of the dilution curve, consider a chamber of volume V, in which fluid is entering and leaving at a constant rate v_c litres/minutes. Assume that in the entering stream, a volume v of some marker is introduced, and that the time it takes for this marker to flow into the chamber V is t_1 minutes. We take as the origin of time the instant when the marker first appears in the chamber. Let c(t) be the concentration of marker in the chamber. Then if the marker is uniformly mixed in V, the equation satisfied by c(t) in the time interval $t = 0$ to t_1 is

$$\frac{dc(t)}{dt} = \frac{v^{i}}{t_1 V} - \frac{c(t)v_c}{V} \quad (1)$$

with $C(o) = O$

For $t > t_1$, the equation is

$$\frac{dc(t)}{dt} = - \frac{-c(t)v_c}{V} \quad (2)$$

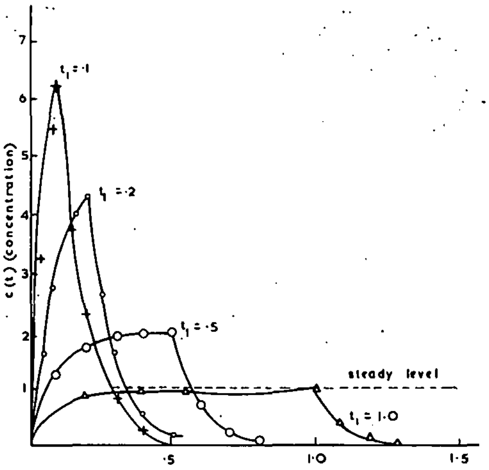

FIGURE 1:—Dilution curves, showing the effect of varying time duration of injection, t_1. Cardiac output constant = 4 litres/min.

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*St. Vincent's Hospital, Sydney. Manuscript received January 20, 1958.

The solution to equations (1) and (2) is

$$c(t) = \frac{v_i}{t_1 v_c}\left\{1 - e^{\frac{-v_c}{V}t}\right\} \quad t < t_1 \quad (3)$$

$$c(t) = \frac{v_i}{t_1 v}\left\{e^{\frac{v_c}{V}t_1} - 1\right\} e^{\frac{-v_c}{V}t} \quad t > t_1 \quad (4)$$

Typical curves calculated from (3) and (4) are shown in Figures 1 and 2 (full lines).

By integration of equations (3) and (4) it may be proved that

$$\int c(t)dt = \frac{v}{v} = \text{Area} \quad \ldots(5)$$

In actual fact, the circulatory system is a closed system. The exit port is connected to the entry port, and the marker will re-appear after the circulation time t_2, which is usually greater than t_1.

In this case, the above set of equations remains unaffected for times less than t_2. For $t > t_2$ (and assuming $t_2 > t_1$) equation (2) becomes

$$\frac{dc(t)}{dt} = -c(t)\frac{v_c}{V} + c(t-t_2)\frac{v_c}{V} \quad (6)$$

where the second term on the right hand side of the equation takes into account the effect of recirculation.

Equation (6) has been solved by taking the Laplace Transform. The solution may be expressed as a power series:

$$c(t) = \sum_{n=0}^{\infty} C_n(t) \text{ with } C_n(t) = 0 \text{ for } t < nt_2$$

$$C_n = \frac{v_i}{V}\left\{\frac{v}{V}(t-nt_2)\right\}^n - \frac{v_c}{V}(t-nt_2) \quad (7)$$

The nth term in the series expresses the effect of the nth recirculation, the nth subsidiary maximum at time $t = nt_2 + V/v$ The amplitudes of these subsidiary maxima are in the ratio; 1:0.37 : 0.27:0.22.

The effect of the recirculation time t_2 is to make a series of subsidiary maxima appear in the dilution curve beginning at $t = t_2 + \frac{V}{v_c}$

In actual practice this last refinement [equation 7] does not lead to any practical application, because not all the blood takes exactly t_2 minutes to circulate. Rather, there is a distribution in circulation times, so there is in reality a steady decline in c(t) from the first maximum to a steady level given by:

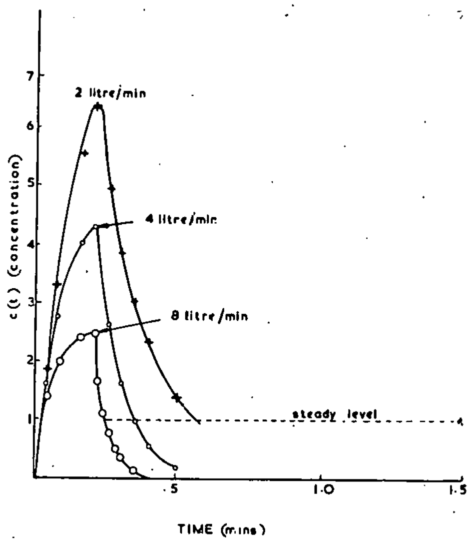

FIGURE 2:—Effect of varying cardiac output, V_C. Injection time $t_1 = 0.2$ min.

$$\text{Steady Level} = \frac{Vi}{L} \quad (8)$$

where L is the total volume of blood in the whole circulatory system, usually called the blood volume.

If in practice t_1 is made short compared with the mean recirculation time, then sufficient of the curve corresponding to the first transit can be observed so that the remainder of the curve can be extrapolated, and in this way the effects of recirculation ignored.

From equations (3) and (4) it is seen that the shape of the curves is defined by three parameters—t_1, the time for the marker to enter the chamber, v_c the rate of flow through the chamber and V, the volume of the mixing chamber.

The effect of varying the injection time is shown in Fig. 1. From equation (5) it follows that all curves have the same area. They become progressively longer and flatter as t_1 is increased. Similarly, the effect of varying the cardiac output v_c is shown in Fig. 2, with all the other parameters held constant at a typical set of values. Theoretical considerations have been presented by Newman et. al. (1951).

EXPERIMENTAL METHOD

In applying the above theory to an actual determination of cardiac output in a patient, V is the volume of the heart chambers in which dilution takes place, and v_c the rate of flow, is the cardiac output. It should be noted that in the theory v_c is assumed constant, whereas in actual practice the flow is pulsatile. However, this latter assumption does not give rise to significant error on account of the short duration of one heart beat compared with the period of complete dilution.

All of the above theoretical considerations apply whatever the form of marker used. In practice, chemical tracers and radioactive tracers have been used. The chemical tracer most frequently employed has been Evans blue dye, T1824, the concentration of which is measured by the light absorption in a series of blood samples, although other chemical tracers, such as sodium chloride and sugar, have been employed. Among the radioactive tracers, erythrocytes labelled with phosphorus 32 and iodine 131 labelled human serum albumin may be mentioned, of which the latter is to be preferred because of the simplicity of measurement.

In the latter method, a collimated crystal counter is placed over the base of the heart. The counter is connected to a ratemeter, the output of which is connected to a strip chart recorder. A small volume of iodine 131—labelled human serum albumin, is injected into an antecubital vein, and in this way the shape of the dilution curve is graphed directly, the vertical scale being, so far, arbitrary. A typical curve obtained in this way is shown in Fig. 3. The chart recorder is left running for about 10 minutes so that the steady level may also be observed. After extrapolation of the descending part of the dilution curve, the area under this portion corresponding to the first transit is determined graphically. The area and the steady level are measured on the same arbitrary scale, and thus it follows from equations (5) and (8) that

$$\frac{\text{Area}}{\text{Steady level}} = \frac{\text{Blood volume}}{v_c}$$

or that

$$\text{Cardiac Output} = v_c = \frac{\text{Blood Volume} \times \text{Steady level}}{\text{Area}} \quad (9)$$

The Blood Volume is determined in the usual way by a comparison of the activity of a blood sample and a known fraction of the injected material. It is defined by

$$\text{Blood Volume} = \frac{\text{Total counts/min. injected}}{\text{Counts/min./ml. of blood.}} \quad (10)$$

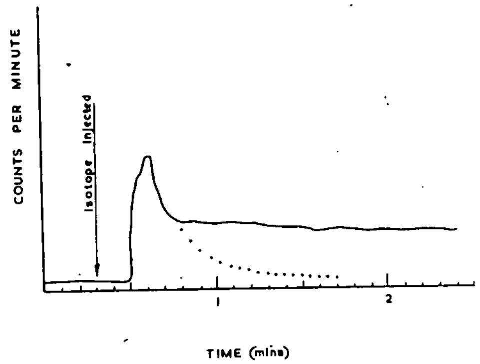

FIGURE 3:—A typical dilution curve following intravenous injection of RIHSA. The isotope was injected at the point indicated and appeared in the heart chambers 12 seconds later. The downsweep of the curve has been extrapolated as shown in the dotted line in order to estimate the cardiac output.

The equipment used by us was manufactured in Australia by Messrs. Long Industries Nuclear Engineering Laboratories, Sydney. The scintillation counter used for these measurements consisted of a circular crystal of sodium iodide, thallium activated, facing an E.M.I. photomultiplier tube type 6950. A lead collimater with an aperture of 1in. at the crystal, and 3in. at the far end, is fitted in front of the counter. The counter is placed close to the patient's chest over the base of the heart. The ratemeter is used on the range the dye was first tried by Nylin and Celander which gives integration time of two seconds.

In order to minimise the retention of iodine 131 in the patient's thyroid gland, all patients investigated by this method receive one gram of potassium iodide daily, commencing one day before and continuing for several days after the test. In order to reduce the radiation dose to the patient, attempts were made to reduce the amount of iodinated albumin to as low a value as possible. It was found that satisfactory readings could be obtained using 25 microcuries, and this value has now been adopted as a standard. This material is injected in a known volume, usually one millilitre.

Ten millilitres of blood are collected in an oxalated collection bottle. This is transferred to a plastic pot and its activity measured in a scintillation counter. An aliquot of about one-thousandth of the injected dose is assayed in another container, made up to the same volume with water. The cardiac output is then determined from equations (9) and (10).

RESULTS AND DISCUSSION OF THE CARDIAC OUTPUT MEASUREMENTS

So far some 25 determinations of cardiac output have been made with this method on patients, without intracardiac shunts which could give rise to abnormal curves. The patients concerned fall into three separate classes: (a) Fifteen patients who were being studied at the same time by the Fick method. (Cournand and Rances, 1941.). (b) Five patients undergoing left ventricular puncture in the operating theatre and (c) Five women in advanced pregnancy investigated for localisation of placenta.

From the first series it was possible to get some idea of the accuracy of the method. For each patient the ratio of the radioisotope result to the Fick result was taken. The mean value of this ration was found to be 0.95± 0.03. The standard deviation of the figures so obtained was ± 0.13. It is generally accepted that the errors associated with the Fick method are about ± 10 per cent. (Neely et al. 1954.) Our results indicate that the errors associated with the radioisotope method are of the same order of magnitude, and that any systematic difference between the results by the two methods is small, of orders 5 per cent. (Veall et al. 1954, Huff et al. 1955.)

From Fig. 1 it is clear that the highest accuracy may be expected with shorter injection time, which involves smaller injection volumes. The possibility of improving the accuracy in this way is being explored.

In the other two series of cases, no such comparison is possible, because they would not be considered for simultaneous investigation by the Fick principle. The necessary catheterisation could not be carried out in the operating theatre for the second series, nor would it be considered for the third series of advanced pregnancies. They serve to illustrate the class of patient in which the isotope investigation can play a very useful role.

The method appears to be sufficiently accurate, and is much more convenient for operator and patient than other methods. Also, it can be repeated at quite short intervals if necessary. The method appears likely to become established in routine clinical measurements of cardiac output, and would appear to be eminently suitable for research purposes where repeated observations are necessary.

DIAGNOSTIC VALUE OF THE ISOTOPE DILUTION CURVE

In the above series of cases, the curves were all of a similar pattern, and were generally consistent with those expected theoretically. From such a series it is possible for the experienced observer to get a good idea of what to expect in the normal case. In the case of congenital abnormality, such as atrial septal defect or anomalous pulmonary veins, quantitative deviations from the normal dilution curve may be expected due to excessive recirculation. The possibility of using the isotope dilution curve as a diagnostic tool for the investigation of such abnormalities has been discussed by Prinzmetal et al. (1948). The method is not always reliable, because of difficulties associated with siting the counter, location of which is critical, and it has not come into general practice. A variation of a method which overcomes this objection has been tried out and is reported briefly below.

This method is again an isotopic variant of a method established by dye-dilution methods. Swan and Wood (1953) have described a procedure for the detection of intracardiac shunts. A cardiac catheter is passed into the heart, and through it a small volume of Evans-Blue dye is passed. The concentration curve in the arterial blood is recorded as a function of time using an ear oscimeter. From a comparison of tracings obtained with the catheter tip placed in different chambers on each side of the heart, the presence, and possibly the type, of intracardiac shunts could often be detected.

The method might lend itself to adaptation to the isotope method in cases already being investigated under catheterisation. Ordinary iodine 131 as sodium iodide was used, because quantitative results were not sought. Sodium iodide was used, because it was cleared more rapidly by the patients—thus reducing radiation hazards—and also to conserve supplies of the iodinated serum albumin. The only difficulty was to find a suitable point in the circulatory system at which to scan the arterial blood. This can always be done by arterial puncture, but it was decided to avoid this procedure and to employ external counting as far as possible. After some trial and error it was decided to measure the dilution curve at the finger tips. This avoided any complication due to the venous return, and was the closest approach to the ear oscimeter technique of Swan and Woods.

The same equipment described above was used. The counter was placed over the finger tips, and in order to increase sensitivity the collimeter was removed, thus enabling the full area of the crystal to be employed. The finger tips were placed in contact with the end of the counter, being then within a few millimeters of the crystal surface.

Following saturation of the thyroid gland with potassium iodide, 25 to 50 μc of iodine 131 — labelled NaI were inserted through the catheter, depending on the number of repeated observations to be made.

A series of dilution curves obtained in this way with different catheter positions is shown in Figure 4. It will be noted that the dilution curves are now quite different in shape, presenting a fairly rapid rise in 10 to 20 seconds to

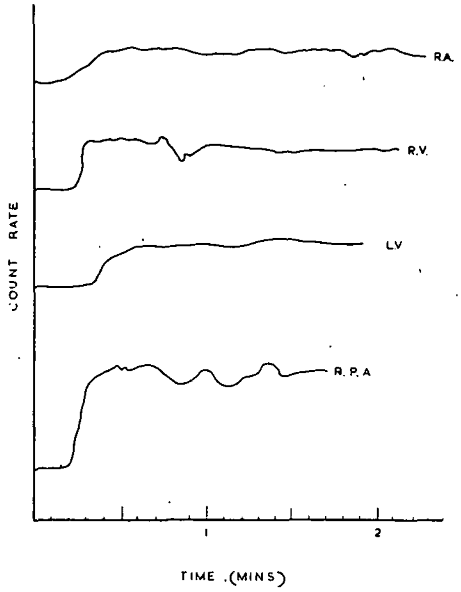

FIGURE 4:—A series of curves showing the time dependence of the finger-tip counting rate following injection through a catheter. The tip of the catheter was in various heart chambers. Reading from the top down, the injection was made into the right auricle, right ventricle, left ventricle and right pulmonary artery.

a steady value which is thereafter maintained. Curves of a similar shape have been reported by McIntyre and Pritchard (1952), who made measurements with a counter placed on the ball of the foot. The shape of the curve is now presumably largely determined by the diffusion of the marker into the vascular bed. The explanation of this particular shape of the curve, and in particular the absence of the first peak of the dilution curve is imperfectly understood. However, it is not of importance for the particular use we want to make of it.

What we have found of significance is the time interval between the injection of the marker through the catheter, and the appearance of the marker at the finger tips. In the case illustrated in Figure 4, these time intervals were:

Position of Catheter	**Appearance time (secs.)**
Right Auricle	6 with slow rise to 21
Right Ventricle	14 with rise to 20
Left Ventricle	21
R.P.A.	12 rising to 18.

These results clearly indicate a right-to-left intracardiac shunt at the atrial level, a finding which was confirmed later at operation. The method has been used frequently, often with valuable results. More results are needed before an appraisal of the value of the method can be given.

CONCLUSIONS

(i) The isotope dilution curve provides a simple and reliable method for the measurement of cardiac output.

(ii) Small volumes and rapid injection of the tracer are desirable for the greatest accuracy.

(iii) The standard error of the method is about ± 10 per cent., and any systematic error is negligibly small.

(iv) The measurements can be made in 10 minutes, and the method is particularly valuable under conditions rendering other methods unavailable.

(v) An inversion of the method in which the dilution curve of isotopic marker introduced through a cardiac catheter as measured at the finger tips is of significance in the diagnosis of congenital deformity.

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Active Handling Facilities at A.E.R.E., Harwell

By J. M. Finniston*

INTRODUCTION

The metallurgical research program at Harwell involves a study of materials which are alpha-active, e.g. plutonium, uranium 233, irradiated bismuth and irradiated materials which are generally beta-gamma-active but can also be alpha-beta-gamma. The operator working with such materials has to be shielded directly from radiation, and protected from ingestion of or contamination by aerosols or discrete particles. The nature and type of protection which has to be afforded varies with the type of material. This paper describes typical facilities for the handling of alpha-active materials, principally plutonium and alpha-beta-gamma samples, mainly irradiated uranium.

ALPHA-HANDLING FACILITIES

The handling of alpha-active materials is done in sealed boxes of steel sheet with a working face of perspex ⅜in. thick. This thickness is strong enough to carry many experimental attachments, withstand the working pressure differential and stop all alpha-particle recoils. The boxes generally operate with a controlled atmosphere (e.g. argon), because of the ease of oxidation of plutonium and the dispersion of the oxide to a very fine powder, and at a negative pressure of about one inch water gauge so that leakage is always inwards to the box. The handling of equipment and samples is done manually through sealed arm length rubber gloves let in holes bored in the perspex; this confines the operating area for a pair of gloves to roughly two 2ft. quadrants, anything beyond this requiring a separate pair of gloves appropriately placed. Services (electricity, water, special gases, etc.) are led to sealed junctions set in the boxes.

This is the basic structure of alpha-active glove boxes, and development has been by variation and modifications to facilitate handling and maintenance and improve reliability and safety. This is best illustrated by describing two types of box, i.e. the fixed box of early construction, and the free standing box now in common use.

The fixed box

Initially, the boxes at Harwell consisted of a series of cells roughly 15ft. long by 5ft. wide by 12ft. high, pitched side by side to give a total length of about 100ft. The front working face of these was of perspex panels bolted to steel stanchions, the lower front carrying service panels. A steel back face with removable steel panels forms the walls of a corridor behind the box and allows access to a frog suit area for maintenance, decontamination, etc. A view of a length of this block of boxes is shown in Figure 1.

Each cell can carry several pieces of equipment, but because certain of these had to operate in an argon filled atmosphere to prevent oxidation of samples, an inner glove box was generally built to contain the alpha-active material and equipment directly required for its manipulation; all ancillary apparatus was left in the main cell without rigid atmosphere control. The double perspex windows (one on the main box, the other on the inner box) complicated the handling arrangements by requiring a glove within a glove in certain instances. A single pair of gloves was generally set at a convenient operating height (roughly chest level) and at about 17in. separation, one foot above the working floor of the box.

The argon atmosphere at one time was supplied from bottles, but was later laid on from a specially designed liquifier through gas lines to fixed and free-standing boxes. The purity of this argon is maintained at less than 5 parts per million oxygen and 20 p.p.m. nitrogen, and is fed to the boxes to give roughly up to five changes an hour; with air leakage at 0.5 per cent. box vol./hour the argon in the box analysed about 0.1 per cent. oxygen. The argon after its passage through the box is fed back to the purification plant; where oxygen is removed with excess hydrogen by catalytic combination (on palladium) to form water, which is extracted by refrigeration. Liquification using liquid oxygen as coolant allows separation of the excess hydrogen as gas, and removes the nitrogen in solution in argon, from which it is later redistilled and vented to extract.

The gloves in these early boxes were flanged and held in position by a clamping ring. To change a glove, the ring was removed, a circlip holding the glove in position until a new one was put in place. The old glove and circlip were then pushed into the cell and the new glove clamped.

The disadvantages of the fixed cell system are that it is not economical of space, it has restricted accessibility because working is from one face and handling has to be done through two sheets of perspex, the changing of gloves is laborious and the box is difficult to maintain. Nevertheless, the cells can carry comparatively large pieces of equipment, and a wide variety of metallurgical operations have been done in such boxes; these included vacuum casting up

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*Atomic Energy Research Establishment, Harwell.
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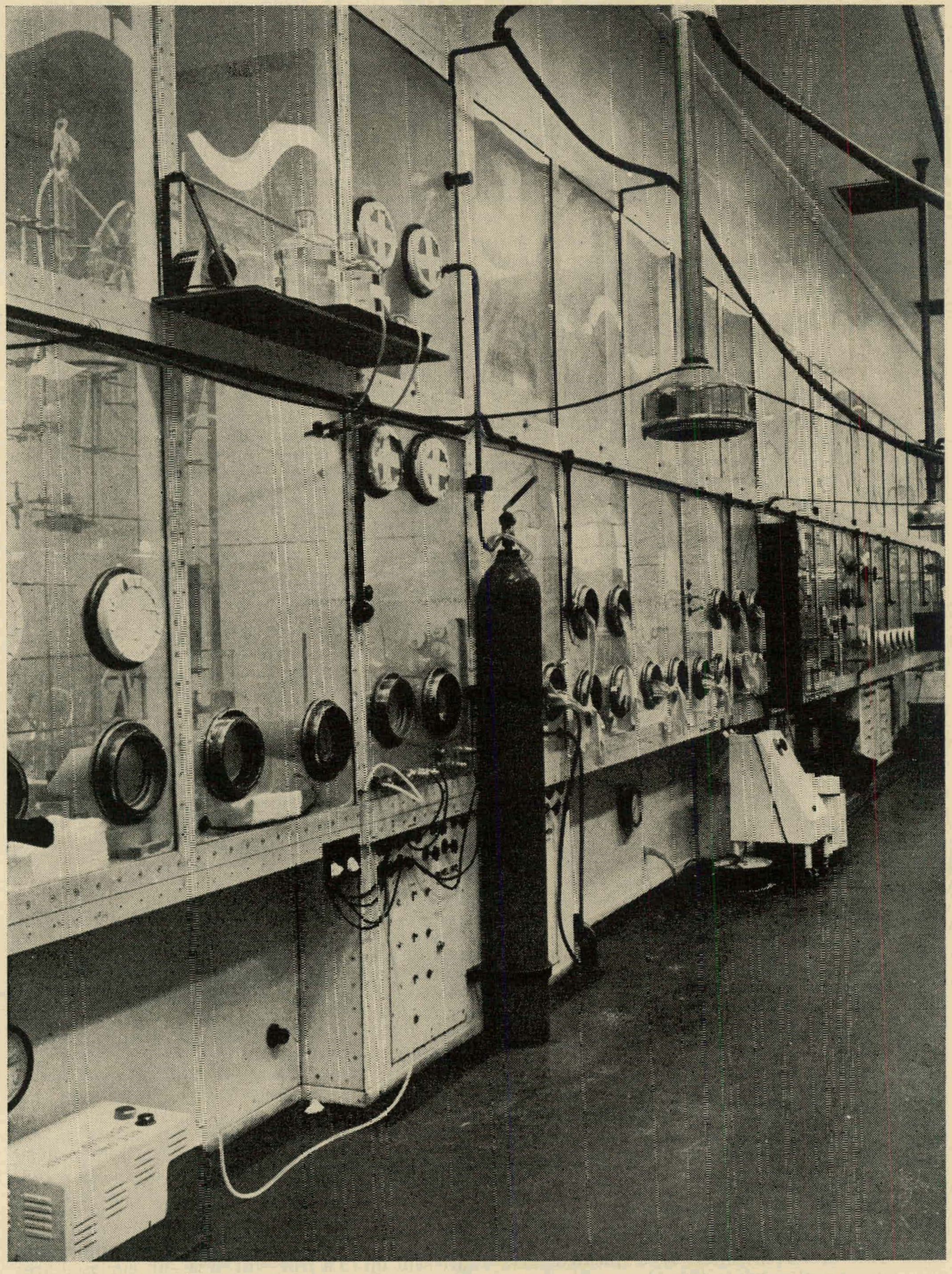

FIGURE 1:—Showing length of block fixed boxes for plutonium metallurgical researches.

to 50 c.c., extrusion of samples initially ¼in. in diameter, automatic polishing for metallography, and thermal analysis.

Free-standing boxes

The restriction of the working face in the fixed cell to one side of a box and the relative inflexibility of the arrangement for research (as opposed to routine) applications led to the concept of the free-standing box. This concept was also associated with the philosophy that the type of equipment that was put into the box should be near standard, and therefore more reliable in operation than special units under laboratory development. Such equipment would also be cheaper, spares would be readily available, and the time involved in developing new equipment would be saved. It has in fact been found that there are few experiments which do not lend themselves to the use of standard equipment in plutonium metallurgy.

One such "modern" box for vacuum-casting plutonium alloys is shown in Figure 2. Boxes were originally built around a steel angle frame to which perspex windows are bolted against rubber gaskets, forming an airtight seal. In still later models the angle frame was dispensed with, the box being made from mild steel sheet bent to form, welded and stiffened with metal strips at weak places, e.g., the base. The inside of the box is painted; a strippable paint (e.g., a vinyl polymer, epoxy resins, etc.) can be used, but has not been found necessary or satisfactory for metallurgical work. Where particular corrosive materials are involved, the box could be made of a stainless or corrosion-resistant material. The bottom of the box is constructed so that there are no corners or places where radioactive spills can lodge or segregate. The perspex windows are sealed to the mild steel sheet by an extruded rubber self-sealing strip.

The box shown is divided into two principal compartments. In the top section, the alpha-active material is melted and cast, and is argon filled therefore as before. In this type of box, leakage rates may be as low as 0.01 per cent. (cf. fixed box); this reflects in higher purity atmospheres and less call on the purification plant. The bottom section carries the ancillary equipment, and is generally run in a nitrogen atmosphere, which saves argon and reduces the fire risk. Other services to the box are served from overhead supplies of power, inert gas, etc. Piped services, e.g., electrical supplies, cooling water, active extract, etc., are passed in and out of the box through some convenient vacuum-tight sealing arrangement; these vary from taped rubber bungs to elaborate soldered fitments. To reduce the amount of alpha-active effluent to be dealt with, a cooling water is recirculated in a simple air-cooled system. To keep the contamination in the extract and argon systems as low as possible, a fireproof filter is incorporated in the box. The preferred filter is a pleated glass fibre with no absorbent; this gives a large surface area in a confined space, has low gas flow resistance, and high filter efficiency for particulates.

THE GLOVE PORT AND TRANSFER PORT

An important feature of any box is the glove and transfer port, the main problem being to ensure that at no time is the inside of the box open to the atmosphere.

A beaded form of rubber glove is now invariably used for handling alpha-active materials, and the double-beaded system of glove changing (Figure 3) is self-explanatory.

When not in use, the gloves are pulled out of the box and the ports sealed with moulded bakelite bungs. A bung in position can be seen at the back face of the box in Figure 2.

Transfer of equipment and materials to (and from) the box is achieved through p.v.c bags with an O-ring incorporated in the open neck, as illustrated in Figure 3 (b). Smooth surfaced p.v.c. is preferred to the satin surface if heat sealing is used. The bags are mounted in the same way as gloves, altough various sizes are available and ports can be made to suit.

A feature of all boxes has been the need to provide adequate storage space for equipment, not only for normal operation of the box but to meet situations if things go wrong—a working "rule" is if you think you will need it in the next three months, put it in the box. Services to these boxes are made more than adequate, and all boxes are fitted with four or more standard 5 amp. three-pin sockets, each capable of independent connection to the appropriate power supply.

A large number of these boxes can be placed in line with transfer ports between one box and the next. This allows a sequence of operations to be done in standard boxes of reasonable size.

HAZARDS

Two especial hazards against which constant vigilance is needed are fire and vacuum blow-out. The main cause of fire is due to defective electrical equipment. Where the risk is great, metal-sheathed cables are preferred, though this is not a foolproof precaution because the box may carry a corrosive atmosphere which can lead to unexpected breakdowns of the cable sheath. Electric motors are also a danger. In one box carrying a lathe, the motor has been separated from the lathe by putting it in a subsidiary box filled with nitrogen. Squirrel-cage motors are preferred to commutator types. Because argon is not such a good insulator as air where voltages above 230 are used, increased clearances are allowed.

Where vacuum systems are involved, safeguards are introduced to ensure that the differential pressure does not exceed a certain limit; in particular, in event of a leak in the box, the rotary vacuum pumps are switched off by controls before the pressure of the box falls to a dangerous level.

MAINTENANCE

When glove boxes have to be maintained they are sent up to a frog-suit area—an enclosed area where handling is done by operators working in "diving" suits. Figure 4 shows an operator in action. The boxes line up against a rubber gasket on the outer wall of the frog-suit area

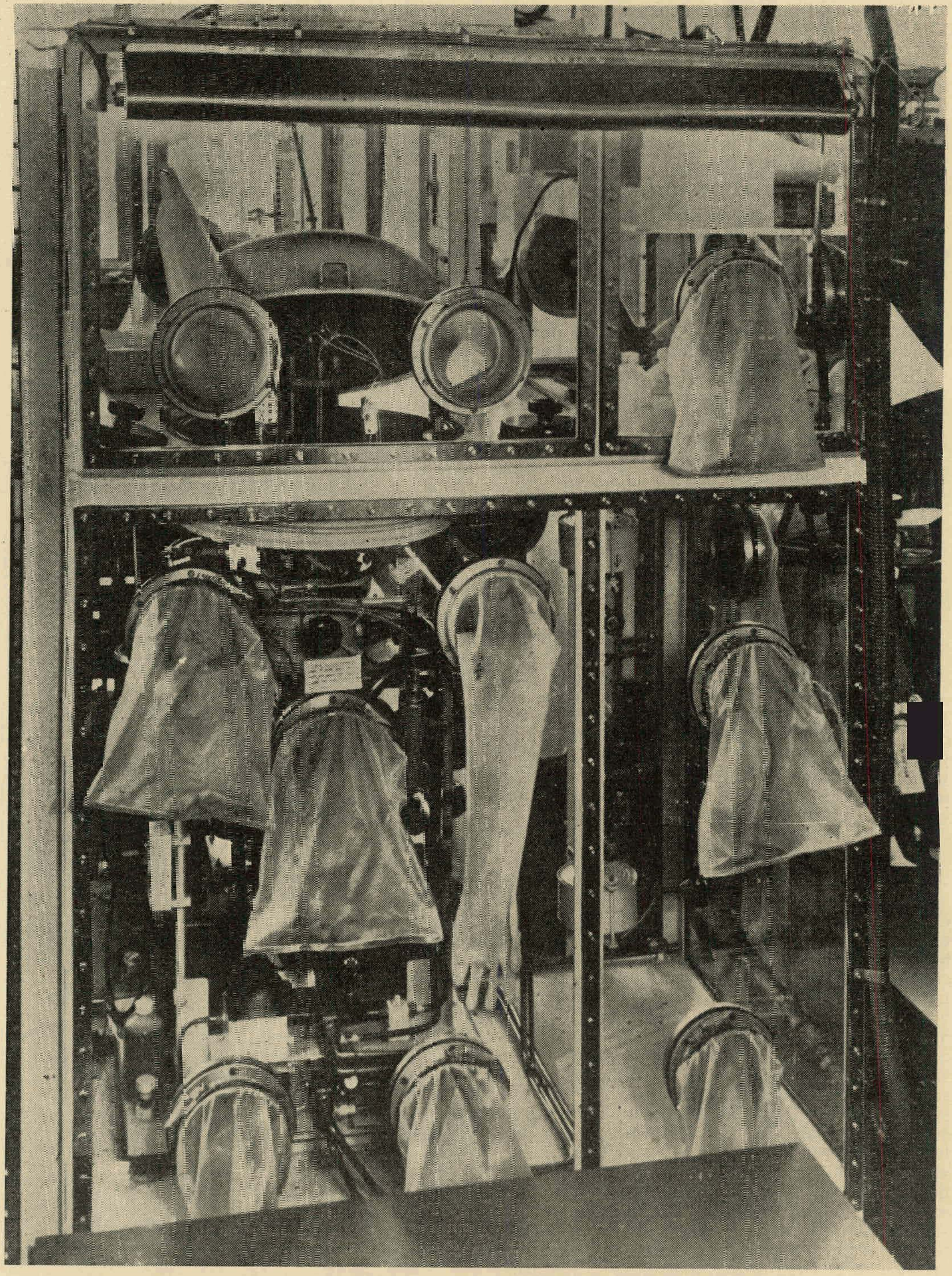

FIGURE 2:—Showing free-standing alpha-active box for metal casting.

and back on to an airtight door creating a lock. The frog-suit operator detaches the inner air lock on his side, removes the window or port from the alpha box, takes out the equipment from the box, and replaces it as desired. He finally reseals the window or metal port in this box, decontaminates the box, and replaces the air lock door. The box is then removed from the outer wall of the frog-suit area.

ALPHA-BETA-GAMMA HANDLING FACILITY

In the alpha-beta-gamma-active handling facility, the cell has not only to maintain alpha-tightness, but has to safeguard the operator against gamma-rays. This can be done by interposing either lead, concrete, or other high density material between the sample and the operator. Estimates of the thickness of wall to provide safeguards against overdose of gamma-irradiation are given in Table 1.

TABLE 1.—THICKNESS OF WALL (IN INCHES) TO SAFEGUARD AGAINST OVERDOSE OF GAMMA RADIATION.

Activity level MeV curies (1 MeV's)	Lead	Concretes			Steel or iron
	Actual*	Ordinary	Barytes	Steel Shot	
10	6	31	24	15	10½
10^2	8	40	29	18½	13
10^3	10	48	35	22½	16
10^4	12	56	41	26	18½
10^5	14	63	46	30	21½

*i.e., using standard brick sizes or combinations of standard sizes.

The first facilities at Harwell for the examination of irradiated materials was based on lead shielding, because its relative thinness compared with concrete (Table 1) allowed manipulation with ball tongs which were available, whereas the concrete cell would have needed the more complicated through-the-wall or over-the-wall manipulators, which were not to hand.

GENERAL FEATURES

As in alpha-active work, gamma boxes can be either in line or free-standing units; of these alternatives, the free-standing lead-shielded box was chosen for metallurgical work. It consists essentially of an alpha-active free-standing box as described previously, set within a lead wall surrounding it on all four sides. Because all the techniques and procedures for alpha-box construction and operation are applied in the same degree when unshielded, no further description of the alpha-box is given unless some special practice has had to be introduced. At suitable positions in the lead walls, holes are left for handling tongs and other equipment, and for lead glass windows for viewing. The boxes themselves are approximately 3 to 4ft. square and about 3ft. high. Those actually constructed contain a wide variety of metallurgical tools, hardness tester, analytical balance, lathe, arc welding equipment, metrology apparatus, furnaces for heat treatment, etc. Some of the boxes rest on concrete plinths which also support the surrounding lead shielding; in others the lead is floor-based. The top of the alpha-box is sealed by a perspex panel, and above this the lighting is arranged. Because even quite low activities can give serious "sky shine" (reflection of gamma-rays from air) all the boxes are fitted with top steel plate shielding to reduce this to a safe level.

In the design of the boxes, great attention has to be paid to the accessibility of all items of equipment for manipulation, "dead" areas which cannot be reached by tongs having to be avoided particularly. In contrast to the alpha-box, special equipment was designed and the lead-shielded box initially "tailor-made" to meet particular routine tests on irradiated samples. In later designs the flexibilty of the box has been improved by not fixing equipment in place, but there is a limitation to this determined by the ease of removal and introduction of equipment

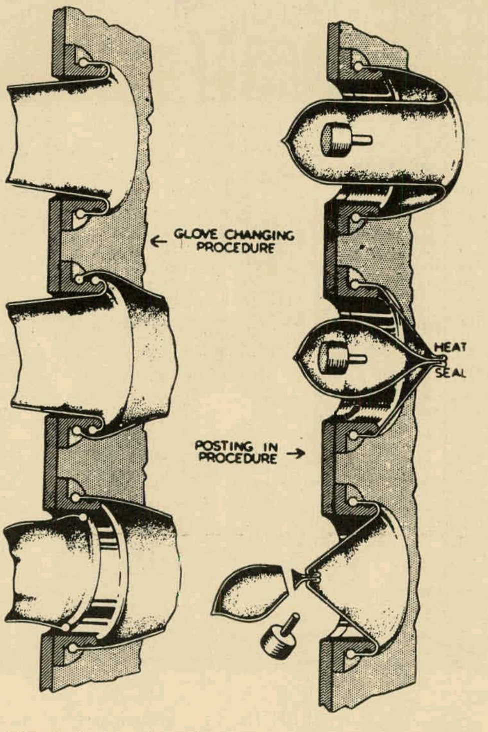

FIGURE 3:—(a) Showing method of changing gloves

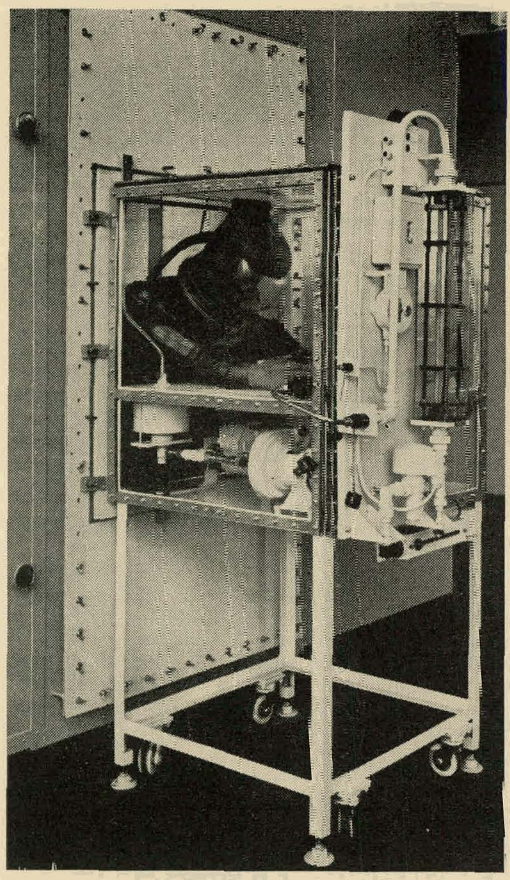

FIGURE 4:—Maintenance of alpha-active box by frog-suited operator.

via small posting ports and the reach and strength of tong units. In the following paragraphs details are given of tong units, windows, lighting, etc.

Shielding

In some of the early-type boxes, shielding was achieved by the use of one or more walls of 4in. thick lead bricks. These were of chevron design i.e., V type dovetailing on four sides so that when stacked as a wall there were no leakage paths for radiation. Two of these lead walls gave protection up to approximately 200 MeV curies. Because 1,000 MeV curie samples were anticipated, a new type of single lead brick 10in. thick, with dovetailing between each block formed by a domed surface, was developed. Each block weighs approximately 325lb. and is lifted and stacked in position by a screw-thread cast into the top of each block for attachment of an eye-bolt. This brick considerably reduces the numbers of units in each wall, and makes a more rigid (and safer) structure. Although designed for 1,000 MeV curies, tests show that the shielding is probably sufficient for 5,000 MeV curies. The top shielding to guard against "sky shine" consists of several inches of plate steel, supported from the lead walls.

Viewing

In these boxes, viewing is through lead glass windows in the lead wall, perspex panels being let into the alpha-active box opposite the windows. Some of the glass is ceria stabilised to restrict darkening by radiation on the inner face. Because lead glass has a density 6.2 to obtain shielding equivalent to that afforded by the walls, a considerably greater thickness of glass, which stands "proud" of the general shielding wall, has to be used. With a 10in. lead wall the windows are 16in. thick. The window units fit into a machined lead housing located in the lead wall. Into this housing fit several glass bricks 6in. square by 4in. thick. The high refractive index of the glass makes it suitable for viewing, but having to position the window adjacent to the tong manipulator limits the window size. In some of the earlier type boxes, top viewing by top mirror was used in addition to wall windows; with the installation of top shielding this additional viewing is now lost.

Adequate lighting is provided by tungsten filament or sodium vapour lamps fitted above the top perspex cover of the sealed box. Access for replacement of these lamps is through holes cut in the upper courses of lead bricks to permit withdrawal of the lamp and holder units.

Manipulation

Most operations inside the boxes are done with ball tong units. A typical unit is shown in Figure 5. It consists of a hollow shaft through which passes a push rod. The push rod is connected rigidly at the outer end to a pistol-type grip, and at its inner end locates inside removable "jaw" units. These jaw units are made in a variety of shapes to suit the object to be handled, and constructed so that they can be replaced remotely by a simple spring clip. At various positions round the inside of the alpha-active box are situated spare jaw units. The tong shaft is located in the lead shielding by a sphere free to move in all directions, but with restricted angle. This sphere fits into a lead housing built into the wall; it is made either of lead 5in. diameter (for handling low activities) or of uranium 7in. diameter for use with higher activities and giving about the same shielding as the 10in lead wall. Where the tong shaft passes through the shell of the alpha-active box, it is fitted with a port very similar to the standard glove port for plutonium boxes. A p.v.c. sleeve held in position by a rubber O-ring covers that part of the tong shaft which is likely to become contaminated; the inner end of the sleeve is clamped to the extreme end of the tong shaft. Without this sleeve it was found that during normal operation contamination spread along the tong shaft through the lead ball and on to the operators' hands. Poly-vinyl chloride was preferred because of its better resistance to wear and its lesser susceptibility to faults introduced in manufacture. For the detachable head an alpha-seal is made between

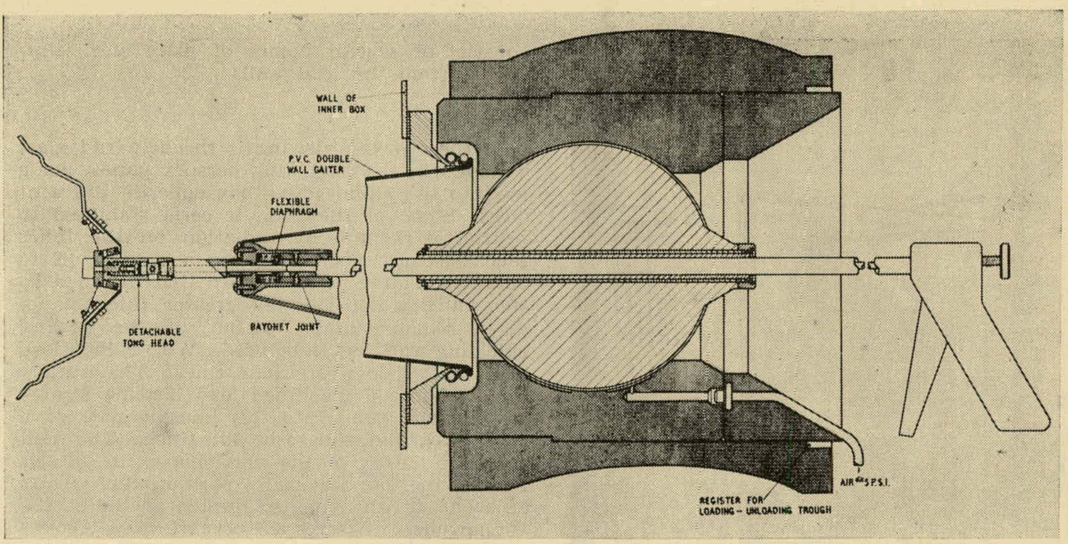

FIGURE 5:—Ball tong unit.

the shaft and the jaw unit to prevent the spread of contamination along the inside of the shaft.

Transfer

For work with highly active alpha-bearing materials it is essential that no spread of contamination occurs during posting, and therefore an adaptation of the technique for plutonium handling has been adopted. Figure 6 (in combination with Figure 3) illustrates this diagrammatically.

The lead wall is pierced by a steel tube which is sealed off at its outer end with a standard type, very long p.v.c tube; at its inner end the tube is sealed to the alpha-box. The alpha-box contains a pair of 2in. lead shutters opposite the transfer tube, so that when these are brought together they provide shielding against gamma reflection from the back wall. A lead shelf is also incorporated in the alpha-box under these ports to prevent direct radiation.

The actual posting is done as follows.

A lead coffin containing the specimen is brought up to the side of the box in line with the posting port. The inner shutters in the box are opened, and the lead plug removed to allow the coffin to be brought up against the lead wall. By means of a push rod operating through the rear of the flask and a tong unit situated in the opposite wall of the box in line with the posting port, the specimen is pulled into the p.v.c bag in the transfer tube and placed under the shelf. At stage (c) in Figure 6 the coffin is withdrawn slightly, another bag placed over the transfer port, and the first bag pulled into the alpha-box. The coffin is then withdrawn further, and the outer lead shield on the box replaced.

When posting out, the specimen is pushed into the bag which is drawn into the coffin. The inner lead shield is closed, the bag looped as shown, heat sealed and severed. The spare bag on the coffin side is tucked into the coffin, using suitable long reach tongs, the coffin door closed and the coffin removed. A new bag is slipped over the remaining portion of the first bag, the latter released into the box, and the outer shield replaced.

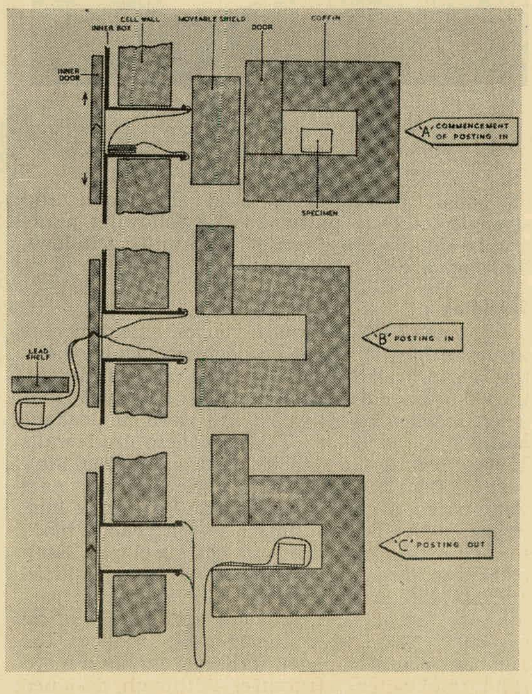

FIGURE 6:—Showing method of tranferring samples to alpha-beta-gamma box.

Care is necessary when carrying out these processes to ensure that the operators secure the maximum protection by working at the side of the coffin, and are not in line with the gap between the coffin and the wall when transfer of the specimen is being carried out.

DECONTAMINATION

The main difficulty with the lead-shielded cell is maintenance if any mechanical changes have to be made. Trouble has been experienced, for example, with bearings seizing, breakage of flexible drives, and faulty connections of couplings requiring dismantling of the lead wall.

In the event of equipment failure, which cannot be repaired with tongs, all active sources are passed out and the box decontaminated by swabbing with tongs until the beta-gamma radiation is reduced to a safe working level. A lead wall is removed after initial decontamination, the tong shafts and rotary drive couplings are withdrawn, and the alpha-box is removed from the lead shielding. Final decontamination and repair are carried out with gloves operating through the tong ports, or by removal of the box to a frog-suit area.

MANIPULATOR CELLS

The lead box, however, is undoubtedly satisfactory when routine operations have to be done. Where, however, experimental facilities are required in which the type of equipment is not known with certainty and may require very considerable modification during the research, the concrete cell with its over or through-the-wall manipulator would appear to afford a better prospect; the lead cell, too, suffers from the expense and delay in erection, commissioning, and major maintenance. The design of a concrete cell to handle alpha-beta-gamma-active materials is still under consideration, but the main features of the new Harwell beta-gamma facility are illustrated in Figures 7 and 8. These cells are intended to handle 100,000 curies. Each cell is 8ft. in internal width, 10ft. 6in. deep and 16ft. 6in. high. The walls are 5ft. 6in. concrete, not of special density. The partitions 2ft. thick between the cells are of steel, filled with steel shot concrete. Each partition forms a sliding door, weighing 30 tons, which can be driven through the near wall into the frogmen operating area behind the back wall. This arrangement allows the cells to be expanded up to five multiples of about 8ft. each.

Operations are viewed through a 5ft. by 3ft. shielded window in each cell. The window consists of seven lead glass laminations and a zinc bromide tank on the operators' side. These are mounted in a frame of reinforced epoxy resin, with a fibre glass laminate. Lighting of the cell

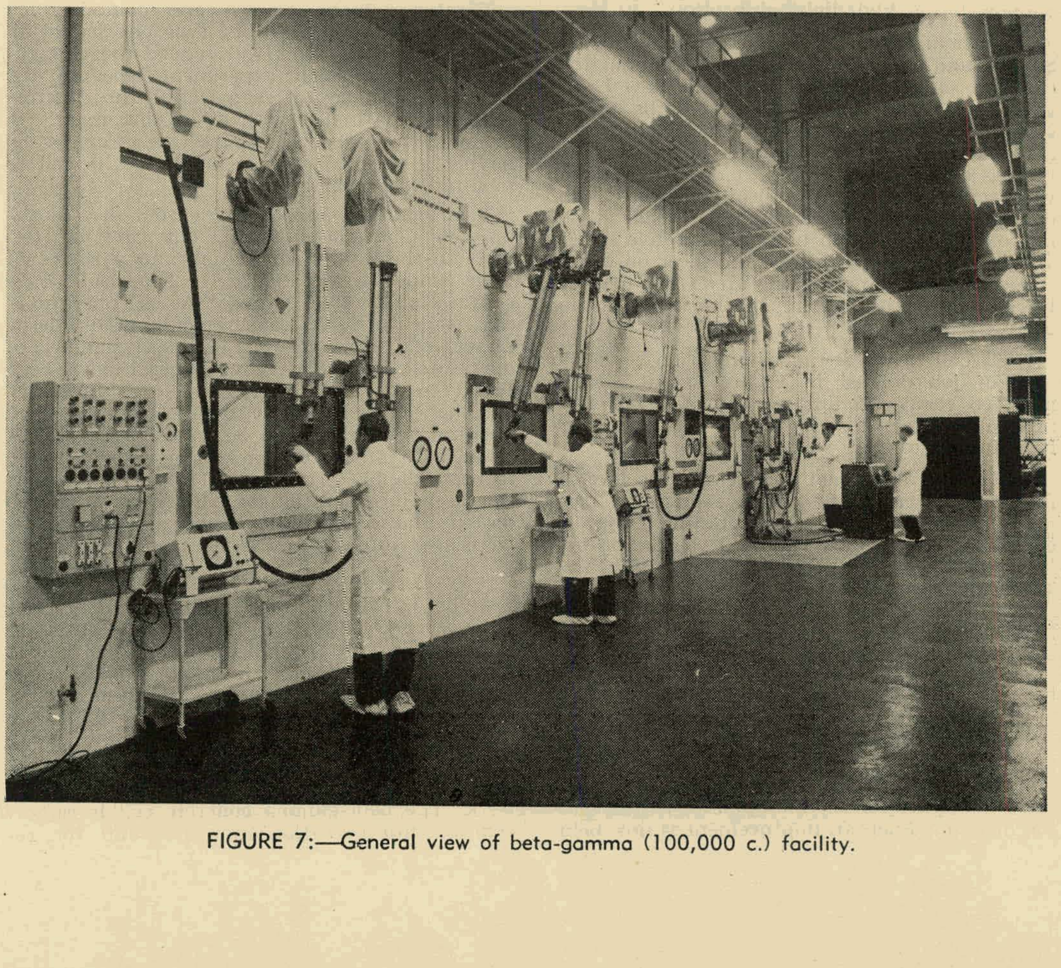

FIGURE 7:—General view of beta-gamma (100,000 c.) facility.

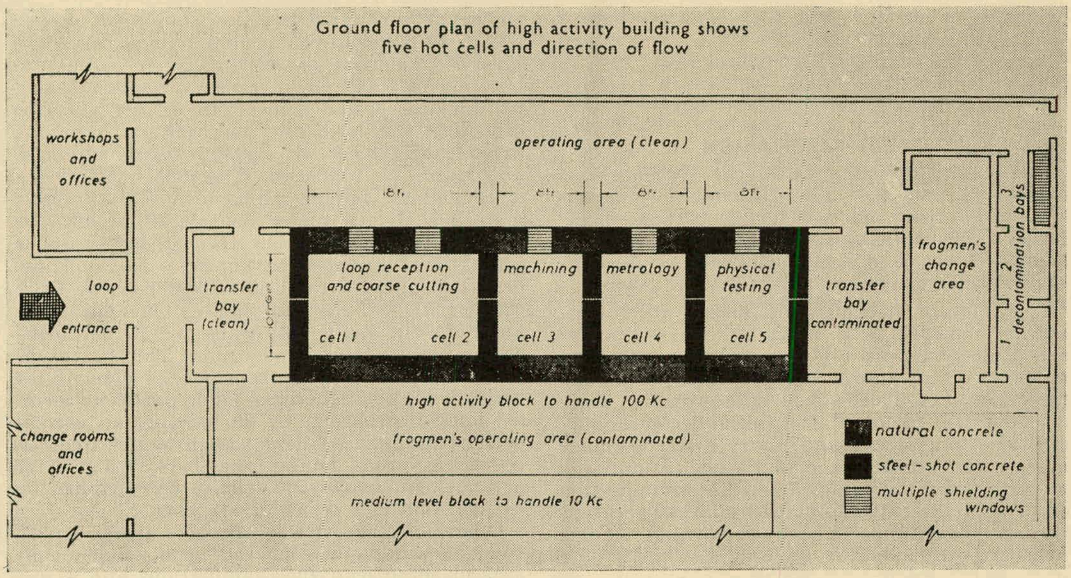

FIGURE 8:—Plan view of facility shown in Figure 7.

is by very bright sodium lights, because the window has a high light absorption.

Operations in the cell are carried out with Savage and Parsons master-slave manipulators. Running on rails over all five cells is a 1½-ton travelling crane and a General Mills power-assisted manipulator operated from outside the cells. To allow the crane and the manipulator to pass each cell, the top part of each partition wall consists of a hydraulically operated drop door. The General Mills manipulator has all the motions of the human hand. It has five speeds, and is controlled by a two-hand lever on a control console in the operating area. The manipulators carry gaiters in much the same fashion as in the lead box.

Each cell has been allocated to do a specific type of job defined in Figure 8, but can be modified easily. Equipment is posted out from any cell and can also be passed into the transfer bay for storage.

The cells are cleaned to a reasonable level by manipulators, and still further reduced by frogmen. The spread of contamination from the cells is prevented by a balanced ventilation system. Pressure of the cells is maintained at 0.7in. water gauge. The outgoing air is passed through filters capable of handling 10,000 cubic ft./minute. The air enters the cells from the ceiling, and is extracted through the floor.

FURTHER DEVELOPMENT IN CONCRETE CELLS FOR ALPHA-BETA-GAMMA HANDLING

The promise of the concrete cell for beta-gamma work suggests its further development for alpha-beta-gamma experimental tests using the p.v.c. gaiter for the manipulator as the sealing medium. Such an improvement is now being examined. In principle, the experimental apparatus will be erected and commissioned outside the cell in an alpha-type box seated on a wheeled table. This will reduce the change-over time for each experiment, because the apparatus can be got ready while the concrete cell proper is in use with other equipment. It is hoped to reduce cell waste time further by having a removable p.v.c. lining to the cell to facilitate decontamination.

This technique has been tried out in a mock-up cell made of plywood, but carrying two through-the-wall manipulators and a zinc bromide window (as in Figure 7). The results have shown considerable promise. Actual operations, e.g., lathe turning, measurements with micrometer, etc., have been tried by a number of operators. Manipulators in general have been found to be superior to tongs for handling, although there is a limit to the extent to which pressure or twisting can be done. Leakages followed with fluorescent powder have been found to be negligible. The main result of these tests has been incorporation of changes in final cell design and dimensions, which are now being translated into an actual working cell.

CONCLUSION

Alpha-active box practice and experience has now reached the stage at Harwell where it affords a trying complication, but no major handicap, to metallurgical work with such materials, and there has been no accident of a significant character to date.

Alpha-beta-gamma practice is still developing. Lead-shielded cells have the advantage at present, particularly for routine processes or tests. The beta-gamma concrete cell is rapidly coming into operation, however, and for research work of a non-routine type is attractive. Recent mock-up tests suggest that the use of such cells with alpha-active-type boxes may provide a facility of general applicability to the handling of any irradiated material in all circumstances.

ACKNOWLEDGEMENTS

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Gamma-ray Measurement of Defects in Poles and Trees

By B. J. J. McHugh and H. E. Booth*

Trees, poles and timbers often carry or develop defects in the form of cavities, gum veins, decayed wood, etc The non-destructive detection and measurements of such defects using gamma-rays from low intensity sources is described. Variations in gamma-ray intensity are measured directly to determine presence of defects. Eight possible techniques are discussed, and one, the "equal intensity" method, is analysed. The optimum type of gamma-ray source for various conditions is determined theoretically. The sources of error and the optimum conditions for accuracy are determined, and the results presented for a typical case. Absorption co-efficients for caesum 137 gamma-ray are given for four species of wood and various kinds of defective wood. The use of the "equal intensity" method in the field is described, and the preliminary results obtained on trees and poles are given.

INTRODUCTION

Mature forest trees in New South Wales are generally defective as a result of termite attack. This attack is usually confined to the heart wood, and results in a central hollowed out region of roughly cylindrical shape, which is referred to as a pipe. The non-destructive measurement of this pipe and other defects is economically desirable, and a program of research is being carried out by the Forestry Commission of New South Wales to this end.

X-radiography has been used with fair success on poles by various workers (e.g., Stoker, 1948), but requires fairly elaborate and bulky equipment and skilled diagnosis. Measurement of defects by means of gamma-rays requires much simpler equipment, and gives immediate results.

The absorption of gamma-rays by wood varies widely, and practicable source strengths and measuring times are severely restricted. Conditions for optimum accuracy must be investigated and various techniques considered in order to achieve the greatest efficiency consistent with the above limitations.

DEFECTS IN POLES AND TREES

Defects vary in size up to about half of the pole or tree diameter in any cross section, larger defects occurring rarely They may occur anywhere in the trunk, but are more often near ground level, particularly in poles.

The defects may take the form of a cavity, or may be filled with various types of matter. The absorption to gamma-rays will vary from the absorption of air to that of water, or even more in rare cases. Measurements of absorption coefficient for the 661 keV gamma-ray from Caesium 137 through various defect materials are given in Table 1.

TABLE 1:—ABSORPTION COEFFICIENTS OF SAMPLES OF TYPICAL DEFECT MATERIALS FOR THE CAESIUM 137 GAMMA RAY.

Defect Material	Collection Area	Absorption Coefficient ft^{-1}
Cavity	—	0.0
Termite nest	Windsor	0.3
Brown fibrous rot	Sydney	0.3
White fibrous rot	Sydney	0.5
Brown cubical rot	Thirlmere	0.6
Termite nest cap	Windsor	1.0
Termite carton material	Sydney	1.2
Brown rot and termite galleries	Wyong	1.5
Rot, mud and galleries	Wyong	1.9
Termite mud	Thirlmere	2.1
Water	—	2.2
Termite clay	Thirlmere	2.4

Precise measurement of these absorption coefficients and those of Table 2 was not warranted because of the variable nature of the materials encountered. The coefficient for a sample was calculated from a single measurement of the intensity of a roughly collimated beam of gamma-rays after penetration. The error in these measurements is of the order of 10 per cent., and the error in those of Table 3 about 5 per cent., as the gamma intensities were measured more accurately.

Under New South Wales conditions, pipe is the most prevalent type of defect in both poles and trees, although other defects, such as rot, gum veins, and shakes, are of wide occurrence. In certain areas, central pipes with low absorptions prevail. Some of the techniques considered take advantage of this.

Poles range from about six inches to two feet in diameter, and trees in New South Wales forests rarely exceed six feet in diameter, most being less than four feet. The absorption coefficient of the wood varies within a species, and between species. It also varies with moisture content. The absorption will not vary greatly

TABLE 2:—ABSORPTION COEFFICIENTS OF SAMPLES OF VARIOUS TIMBER SPECIES FOR THE CAESIUM 137 GAMMA RAY.

Species	Absorption Coefficient ft^{-1}	Moisture Content per cent. of dry weight	Density lb./cu. ft
Pinus Radiata	1.0	18	27
Auracaria cunninghamii	1.2	14	34
Tristania conferta	2.2	15	57
Eucalyptus crebra	2.4	15	71

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* Forestry Commission of New South Wales, Division of Wood Technology, Sydney. Manuscript received April 17, 1958.

TABLE 3. — ABSORPTION COEFFICIENTS OF SAMPLES TAKEN FROM A CROSS SECTION OF A GREEN LOG OF *Syncarpia Laurifolia* FOR THE CAESIUM 137 GAMMA RAY.

Absorption coefficient	Moisture Content per cent. of dry weight
ft^{-1}	
3.1	76
3.1	77
3.0	70
3.0	77
2.8	67
3.0	73
3.0	69
3.0	62

in one cross section of a particular pole or tree (see Table 3).

Measurements of absorption for samples of different species are given in Table 2. As an illustration of the effect of moisture content, a sample of **Syncarpia laurifolia** gave an absorption coefficient of 3.0 ft.$^{-1}$ at 62 per cent. moisture content, and 2.0 ft.$^{-1}$ at 0 per cent.

An error of 20 per cent. is permissible in assessment of defect size in forest assessment and pole inspection. Defects less than 4 per cent. of pole diameter can be ignored in most cases.

CHOICE OF EQUIPMENT

The gamma-source and gamma-detecting and counting equipment must be chosen to give the highest possible accuracy of defect measurement consistent with safe handling and short measurement time. High source strength gives greater accuracy but low source strength is required to simplify safe handling in forest conditions. It is also necessary to compromise between long counting times and speed of assessment.

Accuracy is lost through variations in average background, cosmic ray showers, coincidence losses, decrease of source activity in time of measurement, instrument noise and instability, geometric errors, and scattering effects. The greatest source of error, however, is the "statistical" fluctuation of gamma intensity due to the random nature of radioactive disintegration and of the background sources. The other sources of error can and should be kept small in relation to the statistical error, and will be ignored in the following.

If the error in measurement of the background count is ignored, the standard proportional error in a measurement of the source counting rate is

$$\frac{\sigma\ (S)}{S} = \frac{1}{\sqrt{T}} . \sqrt{\frac{1}{S} + \frac{B}{S^2}}$$

where S is the count rate due to the source
B is the background rate
T is the time of counting.

When the gamma rays from the source are passing through a material of absorption coefficient μ and thickness t,

$$S = S_0 \exp\ (-\mu t)$$

where S_0 is the source rate in the absence of absorber with the same separation of source and detector.

Thus it can be seen that the statistical error in source rate is a function of μt, S_0, B, and T. Its dependence on these parameters must be examined to minimise the resulting error in defect measurement.

Choice of gamma source

Gamma sources can be compared on the basis of the absorption in sound wood of the principal gamma ray. If the absorption coefficient is very low, there will be insufficient contrast between slightly different absorber thicknesses, and accuracy will be lost. If, on the other hand, the coefficient is very high, there will not be enough penetration to enable the gamma intensity to be measured accurately.

The absorption coefficient can be shown (see appendix) to be related to the standard proportional error in the direct measurement of a small defect by the equations

$$\left.\begin{array}{l} \dfrac{1}{\mu t} \sqrt{\dfrac{1}{S} + \dfrac{B}{S^2}} = \dfrac{\sigma\ (\delta t)}{\delta t} . \dfrac{\delta t}{t} \sqrt{\dfrac{T}{2}} \\ S = S_0 \exp\ (-\mu t) \end{array}\right\} \quad (1)$$

where δt is a small variation in wood thickness due to a defect

and the corresponding change in S is smaller than S.

In this equation, the error in the background measurement is ignored, as most techniques avoid the necessity for this measurement.

As μt increases, the function $\dfrac{1}{\mu t} \sqrt{\dfrac{1}{S} + \dfrac{B}{S^2}}$ decreases from infinity to a minimum value, and then increases to infinity. Consequently, there will be two values of μt corresponding to one set of values of $\dfrac{\sigma\ (\delta t)}{\delta t}$, $\dfrac{\delta t}{t}$, and T and the difference between these two values will be least when the right-hand side of the main equation (1) is smallest.

From these equations, it is possible to determine a range of values of μ for which the defect error will be less than 20 per cent. The critical range is that for which $\delta t/t$ is at its minimum of 0.04. The end values of this critical range will be the two values of μ satisfying.

$$\left.\begin{array}{l} \dfrac{1}{\mu t} \sqrt{\dfrac{1}{S} + \dfrac{B}{S^2}} = 5.66 \times 10^{-3} \sqrt{T} \\ S = S_0 \exp\ (-\mu t). \end{array}\right\} \quad (2)$$

For a 20 millicurie source and a typical Geiger counter, $S_0 = 10^5$ c.p.m. and $B = 10^2$ c.p.m. A counting time of 10 minutes can be tolerated, although a shorter time is desirable. With these values for S_0, B, and T, we find from equations (2) that the permissible range of μ is given by $0.19 < \mu t < 6.7$. For a six-inch pole, this gives 0.38 ft.$^{-1}$ $< \mu <$ 13 ft.$^{-1}$ and for a six-foot diameter tree it gives 0.032 ft.$^{-1}$ $< \mu <$ 1.1 ft.$^{-1}$.

This shows that with such equipment, caesium 137 would not be suitable for the larger trees (see Tables 2 and 3) which would require more penetrating radiation. With more sensitive equipment, cobalt 60 would fit all these requirements.

Special techniques may give less error than in the direct method on which this assessment is based. The critical range of μ and the optimum value for μ should be determined separately for each technique.

Choice of source strength and measuring equipment

It can be seen from the main equation of (1) that with δ, t, μ, and T fixed, $\frac{\sigma(\delta t)}{\delta t}$ is proportional to $\sqrt{\frac{1}{S} + \frac{B}{S^2}}$ and will be minimum when this is minimum. As a sufficient approximation, we require the other parameters to be such that $\left(\frac{1}{S_0} + \frac{B}{S_0^2}\right)$ is minimum. This quantity is reduced by increasing the source strength (which increases S_0), by increasing the sensitivity (which increases S_0), and by decreasing B.

The sensitivity and B are not usually independent, so that we require the greatest source strength consistent with safe handling and the sensitivity and B such that $\left(\frac{1}{S_0} + \frac{B}{S_0^2}\right)$ is minimum. The detector should not exceed two inches in diameter for geometrical reasons, and the equipment should be reasonably stable.

A 20 millicurie source and a sensitive scintillometer would be satisfactory.

Other equipment

Source handling gear and a frame to simplify manipulations during measurement are required. The caesium 137 source used was carried on the end of a three-foot rod in difficult conditions only, otherwise in a lead container.

TECHNIQUES

Generally speaking, detection of internal defects by means of gamma-rays depends on detecting any disparity between the apparent thickness and the thickness determined from the observed absorption of gamma-rays.

On the line of measurement (the line between the source and the detector), the wood will have an apparent thickness which we may call t_w, there may be a defect of thickness t_d with absorption coefficient μ_d, the wood absorption coefficient being μ_w. If the counting rate due to the source in the absence of absorber is S_o, the source count rate measured will be

$$S = S_0 \exp [\mu_w (t_w - t_d) - \mu_d t_d]$$

The same count rate will be observed for other defects of different absorption in wood of the same μ_w and t_w. In particular, it is useful to consider the "equivalent cavity," which is that cavity which in wood of the same μ_w and external thickness t_w would give the same count rate as observed with the actual defect. This concept has the advantage that it is possible to determine the equivalent cavity from a single measurement of S if μ_w and t_w are known, whereas it is not possible to determine the actual defect size. It can be shown that

$$\text{Equivalent cavity } C = \left(1 - \frac{\mu_d}{\mu_w}\right) t_d$$

Usually μ_d will not be known and t_d can not be found in this way so that the equivalent cavity is the best measure that can be obtained. It should be noted that this can be negative.

TECHNIQUES FOR MEASURING EQUIVALENT CAVITY

These techniques will be considered separately from those aimed at determining the actual defect size. They are mainly based on a comparison of readings through various parts of a pole. This is done to avoid the necessity for determining μ_w, and depends on the fact that a fair proportion of sound wood will be present. Usually the outer shell will be sound, so

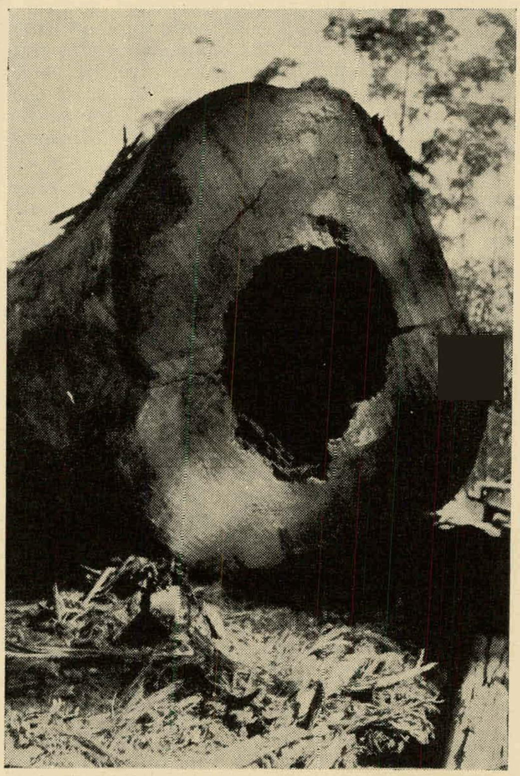

FIGURE 1:—Log-carrying pipe.

that readings on lines of measurement away from the centre can be assumed to be through sound wood. The sound wood absorption is assumed to be uniform (see Table 3).

These methods fail if the defect absorption is nearly equal to the sound wood absorption, for in this case C is zero, and is independent of the actual defect size.

VARIABLE SEPARATION METHOD

Readings are compared on different lines of measurement. The source to detector distance is adjusted so that the readings would be the same if the pole were sound, that is if

$$(\exp(-\mu_w t_w))/s^2$$

is constant where s is the source to detector distance. If the readings are not the same within the experimental error, a defect is present which can be estimated.

This method is only feasible if μ_w is known. As μ_w is not generally known, the technique does not appear to be suitable for practical application, except in very restricted cases.

DIRECT MEASUREMENT METHOD

The equivalent cavity on a line of measurement is estimated from a single measurement of counting rate from the formula

$$C = t_w + (\ln (S/S_o))/\mu_w .$$

Clearly, this can only be done when μ_w is known. However, μ_w can usually be roughly estimated, and C can be determined if it is fairly large. This fact is used in the equal intensity method for pipes, discussed later.

EQUAL THICKNESS METHOD

Two readings taken on different lines are compared. One line is taken obliquely through the sound outer part of the pole in such a way that the apparent wood thickness is equal to that in the other line. If the source to detector separation is kept constant, any difference in reading is due to a defect, and the equivalent cavity is given by

$$C = [\ln (S_2/S_1)]/\mu_w .$$

provided that the first reading S_1 is in fact through sound wood, and that μ_w is constant over the two lines of measurement.

This again requires knowledge of μ_w, but is a more satisfactory method than the previous one because it eliminates the measurement of t_w, and is capable of greater accuracy.

EQUAL INTENSITY METHOD

A measurement is taken along the principal line of interest (usually the diameter) and, if possible, another line is found which gives the same counting rate, but has a different apparent thickness. If the source to detector distance is kept constant, and if the second line is assumed to be through sound wood, the equivalent cavity is given by

$$C = t_{w_1} - t_{w_2}$$

where the subscripts 1 and 2 refer to the two different lines of measurement.

This method is satisfactory from a theoretical point of view, because it is independent of μ_w, and the equivalent cavity can be obtained by simple measurements. Also, no determination of background count is necessary, nor do systematic errors have any effect. The assumption that one line is through sound wood is not critical, but the practical problem of finding an equal intensity position is rather difficult in view of the fact that no such position exists if the wood is sound. The practical application of this technique to a special problem is considered later.

GRAPHICAL METHODS

A series of readings is taken, with constant source to detector separation, as the line of measurement is either rotated or translated through a plane section of the timber. These readings are graphed, and the graphs interpreted for defects.

This approach is necessary when the defects may be in any part of the tree. Parrish (1957) has applied this successfully in laboratory tests, and proposes to apply it in the field.

Various methods of assisting interpretation are possible. Sakamoto and Iizuka (1956) have used a graphical method, and compare the graph with an estimated graph using an estimate of absorption from the readings taken, and calculating the expected readings for the measured external thicknesses.

Calculations can be avoided in interpreting these measurements if the readings taken are plotted logarithmically against the apparent thicknesses. In sound wood of uniform absorption, these points will lie on a straight line, and any departure from linearity indicates the presence of a defect.

Once a defect is detected in this way, its size can be estimated by any of the methods described here.

Techniques for measuring actual defect size

It may be thought at first sight that this approach should take precedence over equivalent cavity methods. This is not the case, because measurement of actual defect size presents far greater technical difficulties.

FINE STRUCTURE METHOD

In X-raying poles for rot and similar defects, it has been found (Stoker, 1948) that best results are obtained by examining the radiographs for variations in wood structure. These variations will also occur when gamma-rays are used, but would be very difficult to detect by gamma-ray measurement. If they could be detected, the size of the defect could be readily found.

EDGE OF DEFECT METHOD

As the line of measurement passes through a position tangential to a defect, there will be a discontinuity in the observed counting rate. If the line of measurement is moved by a parallel translation through the tree and the discontinuities at either side of a defect are noted, the defect size will be indicated by the distance between lines of measurement at these discontinuities.

These discontinuities could possibly be picked up in a graph of readings, although counting rates would need to be measured to high accuracy. Surface discontinuities in the pole or tree would have to be allowed for.

MINIMUM INTENSITY METHOD

In the case of a cylindrical pole with a concentric cylindrical pipe, the minimum counting rate will occur when the line of measurement is tangential to the pipe, if the defect absorption is small enough, i.e., if

$$\mu d \leq \mu_w \ (1\text{-}P).$$

where P is the pipe diameter ratio. (See Figure 3.)

This fact can be used to determine the defect size in a similar way to the edge of defect method. In the cases to which it is applicable, it is a practicable method, although fairly accurate measurements of counting rate are necessary to locate the minimum point with any precision.

Comparison of methods

The variable separation, direct measurement, and fine structure methods do not seem to be suitable for practical application as they stand. Of the others, any comparison depends on the special features of the timber defects to which they are applied.

However, the principle of the equal intensity method seems most attractive for general application as it does not require any knowledge of μ_w, and can result in a considerable reduction in the number of readings taken in comparison with the graphical methods. A method yielding the actual defect size would appear to be preferable to the equal intensity method. This is not the case in general, because such a method

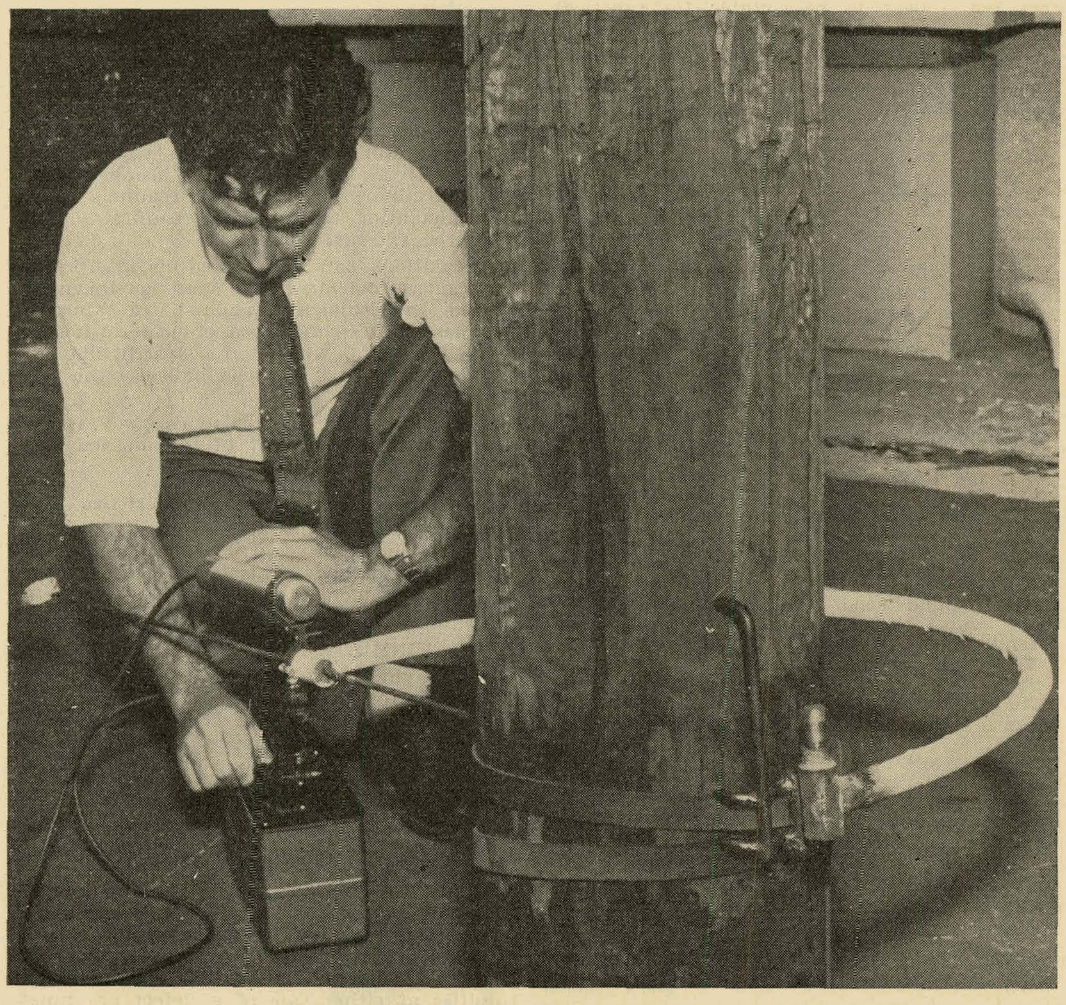

FIGURE 2:—Equipment in use on a pole.

would require much more elaborate and precise measuring equipment.

While the equal intensity method is preferred, the minimum reading method would be superior in the restricted circumstances to which it is applicable.

It should be mentioned that although the above descriptions apply to single defects, very little modification is required in applying these principles to cases where multiple defects may occur.

EQUAL INTENSITY METHOD FOR PIPE

This method is an adaptation of the equal intensity technique to cases which approach an ideal pipe in an ideal pole. Fundamentally, the method has been designed for a concentric cylindrical pipe in a cylindrical pole, the absorption coefficients of sound wood and pipe material being uniform. To this basic method, modifications have been added to cater for departures from the ideal case.

An outline of this technique is given below. A more detailed description will be published elsewhere.

The equipment used consists of a pivot which is attached to the side of the pole, a rigid arm which swings on this pivot in a plane perpendicular to the pole's axis, a source mounted on the arm over the pivot, a detector mounted on the other end of the arm, and counting equipment connected by a cable to the detector. This arrangement is designed to keep a constant separation between source and detector, and to allow the line of measurement to be rotated through a complete set of concurrent chords.

As the arm is rotated, the counting rate observed (S + B) will vary due to the varying thicknesses of wood and pipe encountered. This variation is essentially the same as the variation of S/S_0 which is a quantity independent of the equipment used. Variation of S/S_0 is shown in Figure 3 for typical cases of a sound pole and a pole carrying pipe.

In the case of a sound pole, the average observed counting rate will increase as the arm is moved away from the diameter position. It will remain constant at $S_0 + B$, when the line of measurement passes the position tangential to the pole.

When a pipe is present, the rate will decrease as the line of measurement moves from the diameter until it reaches the position tangential to the pipe. Beyond this point, it will increase as for a sound pole. As the diameter rate is lower than $S_0 + B$, it can be seen that two positions exist where the rate is equal to that at the diameter. This, of course, assumes that the defect absorption is small enough, that is

$$\mu_d \le \mu_W (1\text{-}P)$$

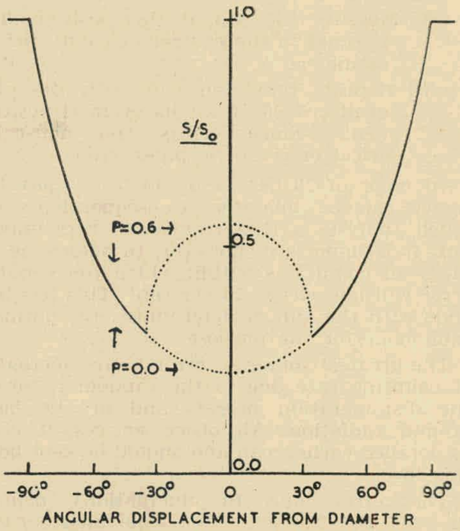

FIGURE 3:—Typical variation of source rate S after penetrating chords in a cross-section of a cylindrical pole. Curves are shown for two values of the pipe ratio P = pipe diameter/pole diameter, P = 0.0 corresponding to the case of a sound pole ($\mu_W d = 1.5$ and $\mu_D = 0$).

The measurement of the pipe depends on measuring the "gap" at the equal count position. The gap for a particular arm position is the difference between the pole diameter and the apparent wood thickness in the line of measurement. It is measured directly with the assistance of a marker on the arm which is set on the diameter. For a cylindrical pole, the gap increases as the arm is swung away from the diameter position. It can be shown that, in any position of the arm, if the average rate is less than, equal to or greater than the diameter rate, then the equivalent cavity is correspondingly greater, equal to or less than the gap.

These properties greatly assist the search for a position of the arm giving a rate equal to the diameter rate. If the rate is low, then the arm must be moved further away from the diameter and vice versa.

The procedure followed in applying this technique is as follows:—

(i) The apparatus is set in position.

(ii) A reading of counting rate is taken on the diameter.

(iii) A rough estimate of equivalent cavity is made. (This is done by the direct method, using some rough estimate of μ_W, an error of 100 per cent. being permissible. It is necessitated by the fact that the basic method becomes misleading when pipe ratios near unity are encountered.)

(iv) The arm is set so that the gap is equal to the smallest defect size which is to be detected, and a reading is taken. If this is greater than the diameter reading, the pole is assumed sound and steps (v) and (vi) are omitted, otherwise—

(v) Search for the equal intensity position which has been found to exist by step (iv).

(vi) Measure the gap at this position. This is equal to the equivalent cavity for the diameter.

(vii) Repeat from step (iv) with the pivot displaced 90° from its original position. (This reduces errors from non-cylindrical or eccentric pipes, etc.)

An error of 20 per cent. in the equivalent cavity can be tolerated. Consequently, a detailed analysis of the error is not very important. It is important, however, to determine the range of conditions within which the standard error will not exceed 20 per cent. This has been done with the aim of determining the optimum equipment for the purpose.

The greatest source of error is the fluctuation of counting rate due to the random nature of the disintegration process and of the background radiation. All other sources of error, as detailed earlier, can and should be kept below this level.

The error due to fluctuations depends on S_0 B, T, μ_w, the equivalent cavity C, and the pole diameter d. It decreases for decreasing $\frac{1}{S_0}\left(1 + \frac{B}{S_0}\right)$, increasing T, and decreasing d. The dependence on μ_w and C is more complicated and is shown in the form of error contours in Figure 4 for a typical case.

If the source to detector distance is adjusted for each pole to give maximum accuracy, S_0 is not constant and is proportional to $1/d^2$. The error contours of Figure 4 were calculated on this basis, and the optimum absorption coefficient for the typical conditions of this error calculation was found to be 0.3. This can be obtained, approximately, with cobalt 60 for the less dense dry poles, but is not attainable for the denser growing trees. This indicates that for best results, more sensitive equipment should be used than that used for this example. This again indicates the desirability of a scintillometer.

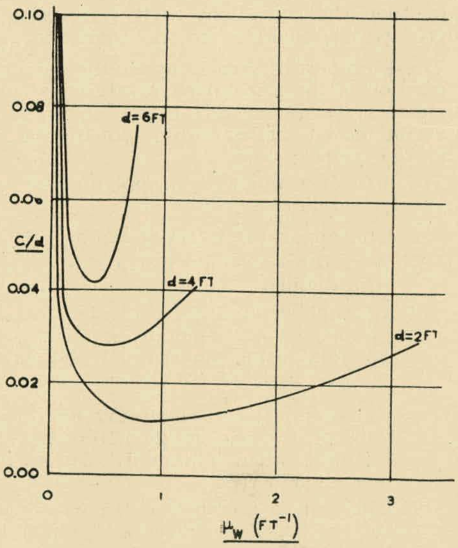

FIGURE 4:—Typical contours for 20 per cent. standard error in C/d and for constant pole diameters (d). C/d is the ratio of the equivalent cavity to the pole diameter; the values of C/d on the contours are the minimum values for which the error is less than 20 per cent. ($S_0d^2 = 2 \times 10^5$ c.p.m. ft^2, B = 10^2 c.p.m., T = 10 m.)

A preliminary field trial of this method was carried out with a 20 millicurie caesium 137 source and a portable Geiger ratemeter. The poles and trees were first measured for defect with the equipment, and then cut down and examined visually. Of the cases for which the defect absorption was found to be sufficiently low, 75 per cent. of the defect estimations were found to be sufficiently accurate. In all 70 cases, including those with high defect absorptions, 60 per cent. were measured successfully.

ACKNOWLEDGEMENTS

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APPENDIX

Equation (1) relating the wood absorption coefficient μ to the error in an estimation of a small defect of size δt and zero absorption in a piece of wood of thickness t is derived from the relation between δt and the corresponding increment in S, the count rate observed due to the source gamma rays after absorption.

$$\delta t \doteq \frac{dt}{ds} \quad \delta S$$

and as $S = S_0 \exp(-\mu t)$

we have $\delta t \doteq \frac{-1}{\mu} . \frac{\delta S}{S}$

The standard deviations (σ) are related in the usual way giving:—

$$\sigma(\delta t) \doteq \frac{-1}{\mu} . \frac{\delta S}{S} \left(\frac{\sigma^2(\delta S)}{(\delta S)^2} + \frac{\sigma^2(S)}{(S)^2} \right)^{\frac{1}{2}}$$

(S) and (S + δS) are measured to obtain δS so that $\sigma(\delta S) = (\sigma^2(S + \delta S) + \sigma^2(S))^{\frac{1}{2}}$.

We can assume that $\delta S << S$ so that

$$\frac{2\,\sigma^3(S)}{(\delta S)^2} >> \frac{\sigma^2 S}{(S)^2} \text{ and } \sigma(S + \delta S) = \sigma(S)$$

$$\text{so that } \sigma(\delta t) = \frac{-1}{\mu} \cdot \frac{\delta S}{S} \left(\frac{2\sigma^2(S)}{(\delta S)^2} + \frac{\sigma^2(S)}{(S)^2} \right)^{\frac{1}{2}}$$

$$\text{therefore } \sigma(\delta t) = \frac{\sqrt{2}}{\mu} \cdot \frac{\sigma(S)}{S}$$

If the error in measurement of the background B is ignored, the standard proportional error in S is given by the well known formula

$$\frac{\sigma(S)}{S} = \frac{+}{-} \frac{1}{\sqrt{T}} \sqrt{\frac{1}{S} + \frac{B}{S^2}}$$

The signs are irrelevant so that we have finally

$$\frac{1}{\mu t} \sqrt{\frac{1}{S} + \frac{B}{S^2}} = \sqrt{\frac{T}{2}} \cdot \frac{\delta t}{t} \cdot \frac{\sigma(\delta t)}{\delta t}$$

A General Purpose Wide-range Electrometer

By C. T. Murray*

A type of electrometer circuit is described wherein both anode and cathode of an input electrometer tube are made to follow closely the input signal. Very high input impedance is obtained, with extreme linearity over a large input voltage range. Typical values are: input resistance 3×10^{17} ohms, input capacitance 10^{-4} micro-micro-farads, maximum voltage range ±25 volts. Particular circuits may be designed to suit individual applications.

INTRODUCTION

The main difficulties associated with electrometer tubes are as follows:

(i) When operated under conditions such that grid current is fairly constant with charge of input signal, the plate characteristics can only be regarded as linear for very small excursions.

(ii) When operated in the floating grid condition to reduce grid current, the change in grid current with input signal is a maximum, and usually slideback type circuits are necessary.

(iii) Most methods of indication when measuring small currents (less than 10^{-14} amp) measure directly or indirectly the small change in anode current of the electrometer tube, and hence stability becomes a serious problem.

(iv) In usual circuits, when the grid resistor is of the order of 10^{12} ohms or greater, the frequency response is severely limited.

Many of these limitations are overcome by negative feedback circuits, such as that discussed by Plechowitch and Zaalberg van Zelst (1952).

In the type of circuit discussed in this paper, the aim has been to overcome most of these objections to electrometer tubes by using them as input elements in cathode-follower circuits in such a manner as to operate at all times in the floating grid condition. This allows a very large input signal to be accepted with the grid current remaining for most purposes negligible, a wider frequency response, and an input capacitance reduced by a factor proportional to the internal gain. These circuits have proved to be very satisfactory over the past four years, and have greatly simplified measurements in a variety of applications.

OPERATING PRINCIPLE

Essentially, the basic circuit consists of a cathode follower, to the input of which is connected an electrometer tube and its anode supply so that both anode and cathode follow the input signal (Figure 1).

The electrometer tube is operated as a cathode-loaded stage feeding the grid of the main cathode follower. In this configuration, the output from V_1, developed across R_1, will appear directly as a grid-cathode signal to V_2 and, therefore will be amplified by an amount $A_2 = \mu_2 \dfrac{R_2}{R_2 + r_{p2}}$ at the cathode of V_2. Also, if A_2 is high, both anode and cathode of V_1 undergo an excursion of the same magnitude as the input signal so that, if initially the operating conditions of V_1 and V_2 are such that the grid of V_1 is at earth potential in the floating grid condition, it will remain sensibly at this operating point when following a given input signal. Circuits have been described (Brewer, 1953; Krakauer, 1953) where R_1, the cathode resistor of V_1, is replaced by a constant current circuit, but the circuit behaviour as a whole is then limited by the characteristics of V_1. In Figure 1, the gain and linearity depend primarily on the behaviour of V_2. For this reason, and to eliminate the separate anode supply for V_1, the main cathode follower, V_2, is operated under constant current conditions.

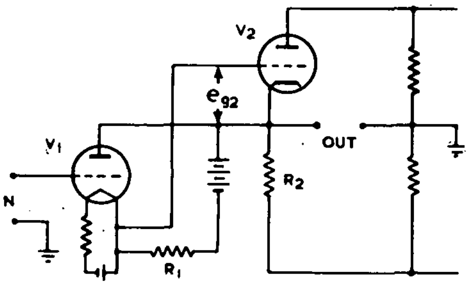

FIGURE 1:—Circuit showing principle of operation of electrometer.

In Figure 2 is shown the basic practical circuit. The pentode V_3 with its screen returned to cathode by the low voltage neon tube, NE2, provides the constant current load for V_2 to within about 0.3 per cent. (variation in anode current of V_1 being negligible). Therefore, the voltage drop across R_2 is constant and provides the anode supply for V_1. If the operating bias of V_2 is about 4 volts, the D.C. requirements of the circuit may be satisfied with no batteries other than the single cell for the filament of the electrometer tube, V_1.

ANALYSIS

Operation of the circuit may be analysed as follows:

With constant current cathode load, the gain of V_2 for a signal between grid and cathode e_{g2}

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*Department of Electrical Engineering, University of Sydney. Manuscript received April 3, 1958.

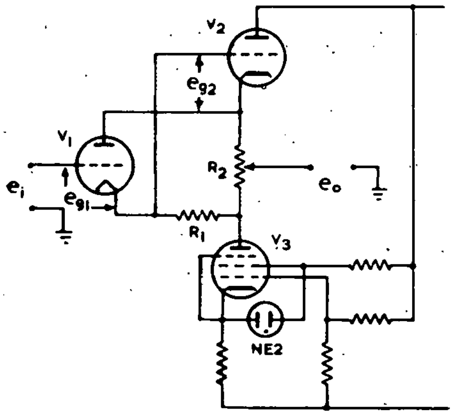

FIGURE 2:—Basic practical electrometer circuit.

is μ_2 (R_2 of Figure 1 approaches infinity). Now e_{g^2} is the voltage developed across R_1 (as (E_{r_2} is constant) and is equal to

$$\mu_1 e_{g^1} \cdot \frac{R_1}{R_1 + rp_1} = A_1 e_{g^1}$$

Therefore, output voltage $e_o = \mu_1 e_{g^2}$ (1)

and $e_{g^2} = \epsilon_{g^1} A_1$ (2)

and input voltage $e_1 = e_{g^1} + e_{g^2} + e_0$

or $e_o = e_i - e_{g^1} - e_{g^2}$ (3)

Substituting in (3) gives

$$e_o = e_i - \frac{e_o}{A_1\mu_2} - \frac{e_o}{\mu_2}$$

$$\text{i.e. } e_o \left\{ 1 + \frac{1}{\mu_2} (1 + \frac{1}{A_1}) \right\} = e_i$$

$$\text{Overall gain } A = \frac{e_c}{e_i} = \frac{1}{1 + \frac{1}{\mu_2}(1 + \frac{1}{A_1})}$$

For the average electrometer triode under these conditions,

$\mu = 1.5$ and $g_m = 80$ micro-amp/volt so that A_1 is approximately 1

$$\therefore A \doteq \frac{1}{1 + \frac{2}{\mu_2}} \text{ or } \frac{\mu_2}{\mu_2 + 2} \quad (4)$$

Also from (3) $e_{g^1} = e_i — e_{g^2} — e_0$

i.e., $e_{g^1} = e_i — e_{g^1} A_1 — e_{g^1} A_1 \mu_2$

$$\text{or } e_{g^1} = \frac{e_1}{1 + A_1 + A_1\mu_2}$$

$$\text{and if } A_1 \doteq 1,\ e_{g^1} \doteq \frac{e}{2 + \mu_2} \quad (5)$$

In (5), e_{g^1} then represents the deviation from the floating grid condition caused by the input signal e_i. For a modern subminiature electrometer tube such as the Mullard ME1403, or Raytheon CK5886 or CK5889 the slope of the grid current curve around the zero grid current point for approximately six volts plate to cathode is about 1.5 x 10^{-13} amp per volt change in grid bias.. If μ_2 has a typical value of 50, the actual input current drawn by the grid of V_1 becomes 3 x 10^{-15} amp per volt input swing, and the input excursion is limited only by the allowable input current or by the allowable voltage swing of the main cathode follower V_2 and V_3.

The input capacitance of the circuit as a whole will consist of two components due to grid to plate capacitance, C_{gp} and grid to cathode, C_{gk}.

From Figure 2, the magnitude of the grid to plate voltage, of V_1, e_{gp}, is given by $e_1 — e_0$:

$$\text{that is, } e_{gp} = e_i (1 — \frac{1}{1 + \frac{2}{\mu_2}})$$

$$= e_i \frac{\frac{2}{\mu_2}}{1 + \frac{2}{\mu_2}}$$

$$\text{or } e_{gp} = e_i \frac{2}{\mu_2 + 2}$$

Thus, effective grid-plate capacitance will be given by

$$C'_{gp} = C'gp \cdot \frac{2}{\mu_2 + 2}$$

For the grid-cathode component, we can see from equation (5) that the effective capacitance

$$C'_{gk} = C'_{gk} \frac{1}{\mu_2 + 2}$$

Therefore total input capacitance C_i will be given by,

$$C_i = \frac{1}{\mu_2 + 2} (2C_{gp} + C_{gk}) \quad (6)$$

Again, in Figure 2, the output e_0 is taken from a point along R_2 instead of from the cathode of V_2. If this is done, the quiescent value of e_0 may be adjusted to zero so that the absolute value of the output follows the input within the error, $\frac{2e_i}{\mu_2 + 2}$ and provides a convenient drive point for gard-rings to reduce the leakage and capacitance of the input system.

PRACTICAL CIRCUITS

From the expressions given in the previous section, it is seen that the practical behaviour of the circuit depends almost entirely on the gain of the main cathode follower. For a number of applications the simple circuit of Figure 2 may be adequate. For the more general case it is necessary to increase the effective amplification factor of V_2. The most obvious method of doing this is to substitute a pentode or pair of triodes in cascode for V_2, remembering the practical limitation that the operating bias of this new stage must still be about four to six volts to satisfy the D.C. requirements of the circuit. This limitation is removed and a significant improvement in gain is provided by the circuit of Figure 3.

In the high performance circuit of Figure 3, the main cathode follower consists of V_2, V_3, V_4 together with the subminiature neons, V_5, V_6 and the voltage reference V_7. Constant current cathode load is provided by V_4 and the two sections of V_3 are connected in cascode. The voltage reference V_7, being returned to the cathode of V_{3_B}, acts as a floating plate supply for V_2, which therefore becomes a differential input "boot-strap" amplifier for V_3, providing an in-phase gain of approximately 50, for a 12AX7. Current through V_6 and R_4 is very nearly constant, as is that through R_2. However, current variations do occur due to the connection of V_7 so that, although the voltage drop across R R_{V2}, R_5 is constant, V_3 does not operate under true constant current conditions and, using a 12AT7, has a grid to cathode gain of only 1000. Thus, in practice, μ_2 of the prevous equations is replaced by A_2, the internal gain of 50,000 from the grid of V_{2_A} to output. The grid of V_{2_B} is returned to the potentiometer R_{V1} and provides a convenient means of setting the D.C. levels of the circuit. Because no phase reversals are encountered in the signal path, frequency response is wider than usual, and there is no tendency to oscillation.

Inserting the value $\mu_2 = A_2 = 5 \times 10^4$ in equations (4), (5), and (6) gives the following results:

Overall gain A = 0.99996.
Input current = 3 x 10^{-18} amp per volt,
Input capacitance = 10^{-4} micro-micro farad.

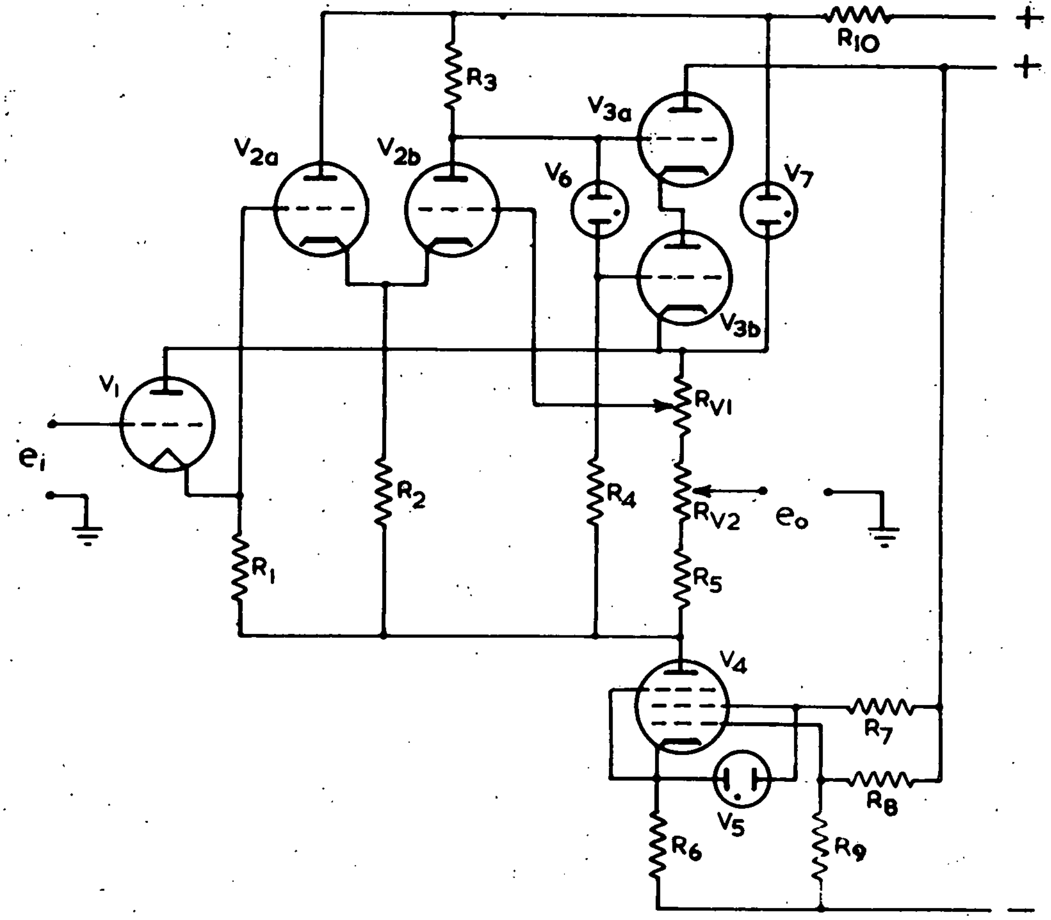

FIGURE 3:—High performance circuit using a "boot-strap" amplifier.

To the input current must also be added that due to leakage associated with the envelope of the electrometer tube and its mounting. If this can be confined to a guard assembly which is connected to the output point, the leakage then constitutes an internal current, the magnitude of which is negligible if e_0 is adjusted to within one or two millivolts of e_1, as it must be for correct floating grid operation. The external circuit will see only $\frac{2}{\mu_2}$ times the change in this current, which is usually then quite negligible.

In these circuits, if the output is taken as shown, from e_0, care must be exercised that only a small load current is drawn. Even though the output impedance may be a few thousand omhs, any load current that is comparable with the constant current of V_4 will cause deviations from the expected behaviour, and it has been the practice of the author to follow this type of circuit by some form of metering stage.

STABILITY

It is obvious that the type of circuit discussed here does not lend itself readily to differential configurations, other than a complete duplication, to reduce drift. Therefore, stability depends to a great extent on the stabilisation of both plate and heater supplies. However, many highly stable high tension supplies are available, and the heater supply used with these circuits has similar properties to a good lead-acid accumulator, so that even with the circuit of Figure 3 a stability of ±5 milivolts over several hours is usual with short term behaviour better than ±1 mV. Also, when high stability components are used throughout, the equipment has been operated for several months without the need for readjustment.

PERFORMANCE

The performance to be expected, of course, depends on the circuit used, and most experience has been gained with that of Figure 3. Table I gives overall values relating to the various types of main cathode follower circuit. In all cases, gain and frequency response are experimental values, the remainder being theoretical, except for the pentode, where all values are experimental.

TABLE 1.—A COMPARISON OF OVERALL PERFORMANCE FOR VARIOUS TYPES OF MAIN CATHODE FOLLOWER.

TYPE OF CIRCUIT	GAIN	INPUT CAPACITANCE μμF	INPUT CURRENT amp/volt	FREQUENCY RESPONSE Kc/s
TRIODE	0·95	10^{-1}	3×10^{-15}	200
CASCODE	0·998	5×10^{-3}	2×10^{-16}	150
PENTODE	0·999	3×10^{-3}	8×10^{-17}	300
BOOT-STRAP	0·99996	10^{-4}	3×10^{-18}	50

COMMENTS

The main use of these circuits, particularly Figure 3, has been in the measurement of low level ionisation in free air chambers. On occasion a gridded chamber has been used where the grid, or indeed the whole chamber, has been capacitively driven from the output to achieve large pulse response for alpha-particles while retaining a mean indication of the level of total ionisation. Also, using a range of about 10 volts full scale, integrating chambers have been used with good results on a number of occasions.

The general usefulness of these circuits is due, to a large extent, to the inherent linearity, and the fact that the gain may be regarded as unity Also, with a supply of 300 volts, an input signal range of ±50 volts is readily obtained with a maximum sensitivity, under most conditions, of ±10 millivolts. In general, no discernible difference results from a change of source impedance from zero to 10_{12} ohms, the highest value on hand. When using an input resistor, the electrometer tube, its filament cell (mercury or "Kalium") and input resistor are all mounted in a shield which is driven from the output to reduce shunting effects and maintain good frequency response.

As it is possible to easily achieve a large reduction in grid current, for many applications an indirectly-headed tube may be used for V_1 instead of an electrometer type.

The circuit of Figure 3 has been used with marked success by Dr. E. A. Johnson, of the Department of Pharmacology, University of Sydney, for the micro-electrode investigation of heart muscle action potentials. A greatly modified version is also in use with the Department of Physics, University of Western Australia, in a new mass spectrometer.

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Hazards Associated with the Use of Ionizing Radiations

By D. J. Stevens*

INTRODUCTION

The past 10 years have seen a rapid expansion in the use of sources of ionizing radiation in industry, research and medicine. Few people who have had an opportunity to study this expansion either at first hand or through the scientific and technological literature will disagree that the next decade or two will see a more widespread use of nuclear reactors as sources of power, and of radio-isotopes as sources of ionizing radiation and as tracers in research and investigations in industry, agriculture and medicine. The economic and technical advantages to be gained from the peaceful uses of atomic energy in all its phases have been discussed elsewhere at this conference.

Any program involving the use of ionizing radiation or of devices which give rise to radioactive materials which are a source of these radiations, potentially exposes humans to increased levels of radiation dose. To date attention has been devoted, in the main, to the radiation protection of the individual, and more particularly to the very limited group of persons exposed to radiation through their particular occupations.

It is appropriate that as we in Australia embark on a second decade in the atomic era, we should take stock of the situation. As a population, we have to learn to live with ionizing radiation if we are to reap the benefits, economic, technical and social, which must surely come with expansion in this field. On many occasions in the past men have stood at the doorway of great technological advancement (perhaps not so great as ours today), without thought of evaluating the debit side (if such an evaluation were within their capabilities) of the new development. There is no doubt in my mind that the pioneers in the development of the motor car had no thought that in the present decade, some 200 persons per year per million of population in Australia would die as a result of accidents arising from the use of this technical development which means so much to our present way of life.

So that radiation protection facilities in the expanding application of atomic energy in industry, research, agriculture and medicine may be appropriate, it is proposed that consideration should be given to the establishment of a maximum permissible radiation dose for the population genetically at risk. A tentative suggestion of one authority is analysed into the contributions which the various sources of ionizing radiation may make, in the future, to the population dose.

Should we not pause before we plunge into the atomic future to think whether the criticism could be levelled at us by generations to come that we thought only of the credit and not of the debit side of the ledger in our enthusiasm to reap the benefits which appear to be so great? There can be no doubts that we must go forward, but we should plan our developments so that the debit side of the ledger has the minimum of entries.

Let us look and see how the whole population will be involved in the future. While it does not cover the whole picture, it is proposed for brevity to consider in this paper only the genetic effects of radiation. In such a context it is appropriate to consider only that portion of the population which is genetically at risk —that is, the population which receives radiation exposure to the gonads before and during the years of reproductive activity. On the basis of experiments conducted with relatively high radiation on lower forms of life than man, there appears to be justification for the hypothesis of a strictly cumulative effect of radiation as regards genetic effects. Thus, radiation exposure of the population which is genetically at risk gives rise to a population pool of mutations which may be transmitted to future generations. These may appear either as major abnormalities in a relatively small number, or as minor (and not easily discernible) aberrations, such as reduced life span and intelligence, in a greater number.

SOURCES OF RADIATION

The dose which may be of genetical significance will arise as a result of the exposure of the population which is genetically at risk, to the following sources of radiation:—

(i) Natural background.
(ii) Medical sources, such as X-rays and radioactive isotopes, used in diagnosis and treatment.
(iii) Occupational exposure in a "controlled" area.
(iv) Exposure outside a "controlled" area.
(v) Environmental contamination.
(vi) Miscellaneous sources.

A brief comment on each of these sources is appropriate.

Natural background

The gonads of each member of the population, in successive generations, are exposed from conception to radiation from natural sources at a dose-rate which is fairly uniform over the continent. The dose is contributed by such external sources as cosmic rays and gamma-rays from naturally-occurring radioactive materials in the soil, the atmosphere, and the masonry of buildings in which we live and work, and by such internal sources as the naturally-occurring potassium 40, and radium 226, which

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* Commonwealth X-Ray and Radium Laboratory, Melbourne. Manuscript received April 21, 1958.

with its daughter products, occur in our body tissues. The contribution of each of these tends to vary with locality and living habits, but on the average the gonad dose of the individual from natural background is of the order of 100 mrem per year. We are not sure what debit we reap from this radiation exposure, but if our working hypothesis for genetic effects is correct, then we have incurred some undesirable genetic effects from the exposure of our forebears to this radiation and we will, in turn, contribute to the pool of mutant genes for the future as a result of natural background exposure.

Medical sources

When we are thinking in this context of radiation exposure to the patient in medical practice as a result of using X-ray equipment or radioactive isotopes, we should recall

(i) that only those patients in a specified group are genetically at risk,
(ii) that these exposures are received as a part of a medical investigation in the case of a diagnostic procedure, and on a doctor's prescription in the case of treatment,
(iii) that only the dose received by the gonads has genetical significance,
(iv) that many X-ray diagnostic examinations involve only very small doses to the gonads of the individual, and
(v) that the techniques which make a major contribution to the gonad dose of the individual are those which include the gonads in the direct radiation beam.

It is worth repeating that when we are considering the impact on the whole population, the dose of interest is not that which an individual may receive, but the contribution this makes to the dose averaged over the population which is genetically at risk.

Occupational exposure

A very small percentage of the population genetically at risk is at present exposed to radiation through their professional and technical employment, and accordingly the contribution made to the population genetic dose is very small. Many of these occupationally exposed persons are using radiation sources which, because of their nature and the radiation they emit, make no contribution to the dose to the gonads.

It can be anticipated, of course, that as more extensive use is made of radioactive isotopes in industry, research, agriculture and medicine, the number of occupationally exposed persons will increase with a possible rise in the contribution from this group to the dose to the gene material of the population.

It is envisaged that persons occupationally exposed to radiation will use ionizing radiations in "controlled" areas, that is, areas with which there are associated facilities to monitor the radiation dose in the working area and to the individual.

Exposure outside "controlled" areas

The increased use of ionizing radiation in its various fields of application will increase the number of people who, although not "occupationally" exposed, will receive radiation in the course of their occupation. It will be appreciated, for example, that while a large unit intended to sterilize, say, pharmaceutical products by gamma radiation, can be designed to keep the radiation levels in the "controlled" areas below those recommended for the occupationally exposed group, others in neighbouring "uncontrolled" areas will be exposed to certain levels of radiation. These levels can, by design, be made as low as desired, but with gamma-ray sources they will never be reduced to zero.

It is difficult to predict, at this stage, the number of the population genetically at risk which will be exposed in this way, and accordingly contribute to the genetically significant dose to the whole population. It is a group to whom consideration must now be given in the planning of future expansions in the peaceful uses of atomic energy.

Environmental contamination

Evidence at present available indicates that the genetically-significant dose to the population, arising from the environment contaminated with radioactive material by man, is very low, amounting to less than one per cent. of the natural background dose. Potentially, man could contaminate his environment significantly by lack of care in the disposal of radioactive wastes, particularly those from nuclear reactors, and by lack of care in the release of fission products either deliberately or accidentally from these reactors. High levels of dose to the gene material of the population are not inevitable from these sources. On the contrary, appropriate authorities are taking elaborate precautions in regard to them, but it should be realised that as a result of the more widespread use of nuclear reactors, there may be some increase in the level of environmental contamination.

Miscellaneous sources

Technological advancement in a particular direction quite frequently brings with it a crop of applications of the technique which are remote from the original intention. So it is with radiation. Some of these applications are useful and contribute to our welfare and technical advancement, but others are of doubtful value. As miscellaneous sources I include, without any attempt to classify them into degrees of usefulness, radioactive luminous paint used on watch and instrument dials, X-ray shoe-fitting devices, television sets, and radioactive "gadgets." Individually, some of these devices contribute a very small gonad dose, but unless care is taken, the widespread use of them may contribute, in the sum, a significant dose to the gonads of the population. We are fortunate, in Australia, that arrangements for the import of radioactive materials are such as to exclude the entry of "gadgets" which contain radioactive material and which cannot be used usefully and safely. From time to time one hears of such "gadgets" which appear to have very doubtful value but which, if subjected to widespread use, could raise the level of radiation dose to which the population is exposed.

PERMISSIBLE DOSE

Many will be familiar with the concept of maximum permissible dose for those occupationally exposed to radiation. The actual value of this dose is recommended from time to time by the International Commission on Radiological Protection, and by various national protection authorities. You will recall that this permissible dose is defined as "a dose of ionizing radiation that, in light of present knowledge, is not expected to cause appreciable bodily injury to a person at any time during his lifetime. As used here, "appreciable bodily injury" means any bodily injury or effect that a person would regard as being objectionable and/or competent medical authorities would regard as being deleterious to the health and well-being of the individual. As the use of ionizing radiation is to expand it appears appropriate to define a maximum permissible radiation dose for the whole population. It would be logical to set an upper limit for this dose which should not be exceeded (on the average) by that portion of the population genetically at risk, and it would be reasonable to include in this level all radiation doses to the gonads of this population, except that from natural background. If the hypothesis of cumulative effect of radiation as regards genetic effects is justified, any quantity of radiation, however small, will ultimately have some consequences. I suggest that an upper limit for the population at genetic risk should be set so that a balance can be made of the advantages to the community from the use of ionizing radiation against any future detriment to the human race. Perhaps, at this stage, the most difficult problem would be to set an actual value because of the lack of real knowledge of the effects of radiation at low dose and dose rate levels on the hereditary material of man. As pointed out before, we cannot set the value at zero dose above natural background if we wish to have the benefits of X-rays in medical diagnosis and treatment, of nuclear power reactors, and of radioactive materials in industry, agriculture, research and medicine. Any maximum permissible radiation dose for the whole population must be realistic. The setting of such a value could have real advantages.

I do not intend to take on the responsibility of the geneticists and arrive at a value which I would recommend. I propose, however, as an exercise, to examine what would be the practicability and value of accepting the guarded statement of the Committee appointed by the Medical Research Council to Report on the Hazards to Man of Nuclear and Allied Radiations (Medical Research Council, 1956). The statement is as follows:

> "In the meantime, we feel bound to state our opinion that it is unlikely that an authoritative recommendation will name a figure for permissible radiation dose to the whole population, additional to that received from the natural background, which is more than twice that of the general value for natural background radiation."

For convenience of comparison it is intended to express radiation doses of the population genetically at risk as that for the 30-year period from conception.

Accepting an average value of dose to the gonads from natural background as 100 mrem per year (i.e., 3 rem in 30 years), the extract quoted above infers a value for a permissible radiation dose to the whole population of 6 rem in 30 years, in addition to natural background. If this value were accepted it would be of interest to attempt to apportion it to the various sources of man-made radiation. The following appears to be realistic:

A dose of 3 rem might be allocated for "Medical Sources." This is somewhat lower than the actual level to which the whole population is now, on the average, exposed from this source. Means are available for the reduction to this level of exposure to the gonads from the necessary medical use of ionizing radiation. Detailed examination of these procedures are not here appropriate. The allocation of the 3 rem for this source, and any consequent effort to achieve it could result, in fact, in a reduction of the average dose to the population at genetic risk to a somewhat lower level. The setting of such a value could lead to a re-evaluation by the medical profession of the medical advantage of certain procedures which involve radiation exposure to the gonads. This might be particularly true of techniques involving the use of radio-isotopes in diagnosis and treatment—some of which might, on re-evaluation, prove to have little or no advantage over those which do not involve radiation exposure. It should not be inferred from this that radio-isotopes at present used in medicine make a major contribution to the dose to the genes of the population, but rather that their use might be evaluated not only on the grounds of clinical or technical advantage but on the impact on the hereditary material of the population.

The contribution arising from the occupationally exposed group in a "controlled" area might be set at 1 rem. It is difficult to assess the number which might, in the future, constitute this work-force. I have postulated that 1.5 per cent. of the population at genetic risk might, in the future, be exposed occupationally to radiation in a "controlled" area. This percentage appears to be an over-estimate of the situation in the foreseeable future. As regards levels of radiation exposure for this group, I have taken a value of 5 rem per year as suggested by the International Commission on Radiological Protection (1958) and inferred by the United States National Committee on Radiation Protection (1957). Assuming a period of employment of 12 years (from age 18 years to 30 years) of all these 1.5 per cent. in the "significant" population, the contribution to the genetically-significant dose of the population would be slightly less than 1 rem. Although the effort may be expensive, this contribution could be reduced to a still lower level by the design of extensive protection facilities. However, the value arrived at is, I believe, an overestimate, because the number of the population exposed in this group is likely to be smaller and many of those occupationally exposed will, in practice, receive less than an annual average dose of 5 rem over the period under consideration.

Thus far 4 rem of the hypothetical maximum permissible radiation dose for the population has been allocated. The majority of the remainder, 2 rem, is at present not being used. However, establishing this reserve at this stage will enable some planning of future facilities for radiation sources not yet taken into account.

As mentioned before, the environment which man has contaminated with radioactive materials contributes, when averaged over the population, somewhat less than one per cent. of the natural background dose (say, 0.02 rem in 30 years). Future environmental contamination from radioactive waste disposal, and the release of radioactive material either deliberately or accidentally from nuclear reactors yet to be built, cannot be estimated. The environmental contamination from these sources can, within limits, be kept to whatever level is pre-determined. However, this is possible only with special care in design of facilities, the construction of which is inevitably expensive.

The contribution to population dose of the group which is exposed to radiation through their employment and yet not classified as "occupationally" exposed is very difficult to estimate. The number involved will grow as radiation is more widely used. If we accept the statement of the International Commission on Radiological Protection (1958) as a basis, the radiation exposure of these persons in an "uncontrolled" area should be limited to 10 per cent. of the level for a person "occupationally" exposed in a "controlled" area. Accordingly, the group exposed through their employment could, as individuals, receive 0.5 rem per year. If we assume that 10 per cent. of the population at genetic risk is exposed in this manner in the future, the average dose to the population genetically at risk in a working period of 12 years (from age 18 years to 30 years) would be 0.6 rem. A greater percentage of the population so employed would raise this dose, while, on the other hand, more extensive protection facilities could reduce the radiation contribution significantly.

The contribution from the miscellaneous sources referred to previously is largely unknown. Potentially it is a large one but, in the main, the radiation levels have been kept within bounds. For example, the levels of ionizing radiation from household television sets are extremely low, but any developments which require the application of very high voltages to the image tube could raise problems if the equipment is used extensively without thought of radiation protection. The use of radioactive luminous paint on the dials of wrist watches, in this age when quite young children wear them, could expose the gonads of the population at genetic risk to radiation levels much greater than the present environmental contamination.

If it is practical to look on the major portion of this 2 rem as a reserve of radiation exposure, it seems important to stress that its expenditure in the future should be carefully weighed. Very great expansion in the use of nuclear reactors and radio-isotopes will not be possible without careful consideration of radiation protection problems beyond the person occupationally exposed. In some cases, the provision of the necessary radiation protection facilities will only be achieved after large expenditure, but in others careful planning of procedures and siting of sources of radiation will achieve large reductions in radiation doses with minimal costs. Each situation calls for consideration, not only of those occupationally exposed, but of those employed outside the "controlled" area.

I would not like to create the impression that radiation protection necessarily involves great financial outlay. Certainly, part of the problem can be solved by having adequately trained technologists available to use the various sources of ionizing radiations under carefully designed codes of safe practice. Planning for the expanding use of ionizing radiation must be carried out now. If we should adopt at this stage of development criteria which in light of future biological knowledge prove to have been inadequate, future generations may incur the penalty. As long as the criteria are not so restrictive as to impede progress and deny us benefits which can be justified, it is safer to err on the side of caution from which it will be easier and indeed cheaper to retract, if increased biological knowledge warrants such action.

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Radio-iodine Studies of the Equilibrium Turnover of Thyroxine in the Thyroid Gland

By T. H. Oddie and I. D. Thomas*

Tracer measurements with iodine 131 in human subjects can give reasonably accurate measurements of the rates of clearance of iodide from the plasma by the thyroid gland and the kidneys. If it is assumed that metabolic equilibrium exists between the various iodine compartments, the rate of secretion of hormonal (thyroxine) iodine by the thyroid gland may be deduced in terms of the two clearance rates and the dietary turnover of chemical iodine 127. Determination of these variables in a series of volunteer subjects has allowed the variation of hormonal secretion to be studied when the subjects are given exogenous thyroxine at several dose levels. An approximate relation is found to exist between the rates of flow of endogenous and exogenous thyroxine. This relation is used to develop a method for estimating the endogenous secretion without the need for a microchemical assay of dietury iodine 127 in a given subject.

INTRODUCTION

Methods used (Riggs 1952) for the quantitative determination of hormonal flow from the thyroid gland have included:

(i) inference from the amount of exogenous thyroid extract (or analogous derivative) necessary either to correct a complete hypothyroid state (Plummer and Boothby 1921; Thompson, McLellan, Thompson, and Dickie 1933; Means 1948) on the one hand or to suppress almost completely the thyroidal uptake of iodine in euthyroid subjects (Greer 1951; Starr and Liebhold-Schueck 1953; Perlmutter and Slater 1955) on the other;

(ii) derivation from the specific activity of the urine together with estimation of the thyroidal uptake of radio-iodine (Stanley 1949; Stanbury, Brownell, Riggs, Perinetti, del Castillo, and Itoiz 1952); and

(iii) derivation from urinary measurements of stable iodide, with estimation of the proportion of the iodide pool accumulated by the thyroid gland (Stanbury et al. 1952), it being assumed that equilibrium obtains in the body turnover of iodide at the time of observation.

The validity of these general methods of study is open to major criticism in each case, and it is pertinent to note that, by the different methods, the values hitherto derived for normal hormone flow (expressed as micrograms hormonal iodine for 24 hours) have ranged from 45 to 240 micrograms.

The present study is based on method (iii), recognising fully the lack of validity of the assumption of equilibrium of iodine turnover. A precise method (Oddie, Meschan and Wortham 1955) has been used for the determination of thyroidal and renal fractional rates of uptake from the iodide pool, and the thyroidal hormonal flow has been calculated in the same subject while receiving known varying additions to his usual food intake of thyroxine or potassium iodide.

In this paper, preliminary results are described for seven subjects receiving thyroxine, without any additional potassium iodide.

EXPERIMENTAL METHODS

Seven volunteer subjects—senior medical students, resident medical officers and research workers—aged between 17 and 46, were selected. For the purposes of this study, no attempt was made to define their thyroidal status. For the duration of the experiment (some four to five months) they continued to eat their usual calorically - adequate, well - balanced diet, avoiding foods known to be unusually rich in iodine, such as sea foods, offal, agar and so on.

Radioiodine uptake studies were performed after an initial two-week period, during which no thyroid-affecting medication was administered, and then at three-week intervals, during which time the subjects received l-thyroxine sodium tablets administered orally in divided daily doses. The dose was increased for a subject after each three-week period until either the radioiodine uptake was reduced nearly to zero, or loss of weight or an increase in the resting pulse rate suggested an increase in metabolic rate. When either point was reached, the administration of thyroxine was stopped and the radioiodine uptake measurements were continued at intervals until the initial level was reached for each subject. At this time a 24-hour specimen of urine was collected for assay of inorganic iodine content.*

MATHEMATICAL BASIS FOR DETERMINATION OF THYROID HORMONE FLOW

With certain minor modifications, the following equations derive from Riggs' analysis (Riggs 1952).

Let us represent the major iodine compartments in the human body as in Figure 1, and

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* These assays were made by Dr. W. Zimmermann, C.S.I.R.O., Micro-analytical Laboratories, Melbourne.

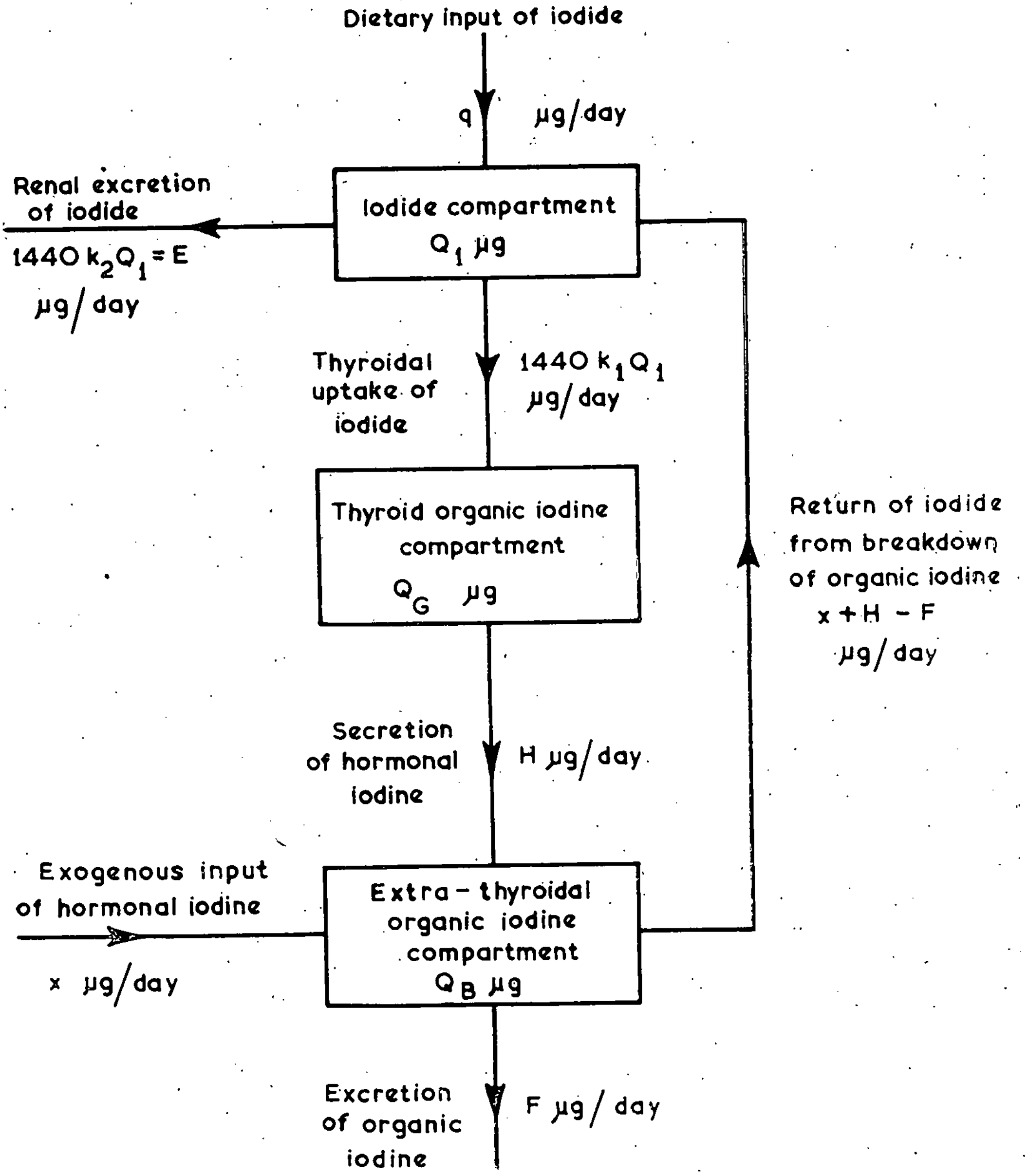

FIGURE 1: Metabolism of iodine.

consider the flow of iodine, in micrograms per day in and out of each compartment, using the following symbols:

Q_I μg = the total quantity of iodine in the iodide compartment.

Q_G μg = the total quantity of organic iodine in the thyroid gland.

Q_B μg = the total quantity of extra-thyroidal organic iodine.

q μg/day = the total dietary input of iodine as iodide, including medicinal doses.

x μg/day = the input of exogenous hormonal organic iodine.

k_1 per minute = the iodide uptake rate factor (Oddie et al. 1955) of the thyroid gland.

k_2 per minute = the renal iodide excretion rate factor (Oddie et al. 1955).

H μg/day = the secretion of hormonal iodine from the thyroid gland.

F μg/day = the non-renal excretion (chiefly faecal) of hormonal iodine.

E μg/day = urinary excretion of inorganic iodide.

If equilibrium conditions can be assumed—that is, Q_I, Q_G, and Q_B have reached steady values at the time of observation—the flow into each compartment must equal the flow out.

For the whole system

$$q + x = F + 1440\, k_2Q_I = F + E \quad \text{(1)}$$

and for the thyroid gland

$$H = 1440\, k_1Q_I \quad \text{(2)}$$

In the usual case when x = 0, Riggs estimated that F is about 6/70 of H and suggested that this fraction is approximately independent of the thyroidal status.

It seems reasonable to extend this to the more general case where $x \neq 0$, giving an estimate for the relatively small excretion of hormone

$$F = 0.086\,(H + x) \quad \text{(3)}$$

Eliminating F and Q_I from the above three equations gives

$$H + x = \frac{k_1q + x\,(k_1 + k_2)}{0.086\, k_1 + k_2} \quad \text{(4)}$$

Let $H = H_0$, $q = q_0$ and $k_1 = k_0$ under the initial conditions when neither medicinal iodide nor hormonal iodine is given.

For any given patient, k_2 has been shown by numerous tests to remain substantially unaffected by large changes in the other parameters.

So equation (4), for the initial status is

$$H_0 = \frac{k_0\; q_0}{0.086\, k_0 + k_2} \quad \text{(5)}$$

Since $E = 1440\, k_2Q_I$, equation (2) gives

$$E/H_0 = k_2/k_0 \quad \text{(6)}$$

Combining (5) and (6)

$$q_0 = E\,(1 + .086\, k_0/k_2) \quad \text{(7)}$$

The daily excretion of iodide E may be estimated by microchemical assay of urine collected over a 24-hour period; k_1 and k_2 are measurable by counting techniques with tracer doses of radioiodine; and $q - q_0$ and x, the daily medicinal doses of iodine, are known.

Thus equations (7) and (4) together allow the endogenous flow of hormone, H, to be estimated while the subject is under various conditions of thyroidal medication.

RESULTS

Table I gives the experimental results on all seven volunteer subjects, including q_0 calculated from equation (7) with the value of E from the chemical assays, and the total hormone flow, H + x, found from equation (4).

DISCUSSION

The hormone flow found in six of the volunteers lies within the range 140 to 451 micrograms per day—values rather higher than those reported by other workers for euthyroid subjects. Volunteer No. 6 shows a much higher value of about 2,000 micrograms per day. As stated above, no definition of the thyroidal status was made in this preliminary study, but further studies will attempt to correlate hormonal flow with an assessment of thyroid function.

The administration of exogenous thyroxine results in a lowering of the thyroidal rate of iodine uptake, an effect achieved by partial inhibition of the pituitary secretion of thyrotropic hormone, it is postulated by Greer (1951). As a consequence of this homeostatic mechanism, the increment in total hormone flow is less than the added amount of exogenous thyroxine.

This effect is demonstrated by the figures in Table I, where, on the average, H + x does not increase nearly as rapidly as x. Within the

TABLE I.—RESULTS WITH SEVEN VOLUNTEER SUBJECTS GIVEN THYROXINE IN GRADED DOSES

Quantity	x	Subject						
	μg/day	1	2	3	4	5	6	7
Mean renal rate 10^3k_2		3.15	2.82	2.55	3.06	2.86	3.36	2.58
E μg/day	0	332	152	336	279	385	1336	531
q_0 μg/day	0	361	183	372	308	424	1518	543
Thyroidal rate 10^3k_0	0	3.13	6.67	3.17	3.62	3.34	5.33	0.69
10^3k_1	32	3.24	3.25	2.66	3.86	4.26	4.42	0.52
10^3k_1	95	2.92	2.16	1.66	3.88	2.41	5.47	0.12
10^3k_1	159							0
10^3k_1	254	1.22	1.04	1.64	3.51	0.70	3.84	
10^3k_1	445	0	0.41	0	0.46	0.30	3.08	
10^3k_1	635						3.80	
Total hormone flow	0	331	360	419	330	451	2120	141
H + x μg/day	32	400	254	416	415	631	1860	145
	95	480	289	380	547	498	2390	124
	159							159
	254	477	403	623	818	412	2080	
	445	445	530	445	551	531	2080	
	635						2800	

limits of accuracy for these experiments, the rise in flow may be represented by a linear relation.

$$H + x = H_0 + Ax \quad (8)$$

where A should be less than 1.

Values of A, found by least squares, are

Subject	1	2	3	4	5	6	7
A	.18	.52	.65	1.41	—.06	.85	.07

Neglecting the value for Subject 4, whose experimental curve for k_1 shows marked inconsistencies, the mean A = 0.37.

Combining equations (4), (5) and (8) gives, for $q = q_0$ because no iodide medication was given in these cases,

$$\frac{k_1 q_0 + x\,(k_1 + k_3)}{0.086\,k_1 + k_2} = 0.37\,x + \frac{k_0\,q_0}{0.086\,k_0 + k_2} \quad (9)$$

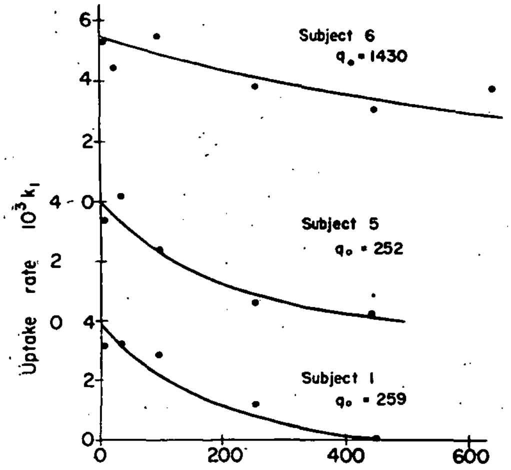

FIGURE 2.

Providing the same value of A is sufficiently accurate for all cases, equation (9) gives a way to find q_0, and thus H_0, from observations of k_1, k_2, and x without the chemical assay of iodine in the urine.

To test this possible method, k_1 was plotted against x for each volunteer, so that k_0 and k_1 could be taken from the smooth curve. Calculated in this way q_0 has the values shown below, compared with the corresponding results by chemical methods.

Subject	1	2	3	4	5	6	7
q_0 from equation (9)	259	246	237	302	252	1430	356
q_0 by chemical assay	361	183	372	308	424	1520	543

Finally, the way equation (9) fits the observed k_1 curve is illustrated in Figure 2. This is tolerable agreement, and suggests that investigation with a larger number of volunteer subjects will be worthwhile.

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Transistor Decade Scaling Circuits for Use in Radiation Counting

By R. E. Aitchison*

The use of transistor decade scaling circuits in radiation counters is discussed. Circuits which use a minimum number of junction transistors are considered, and specific designs given for operation at counting rates up to 1 Mc/second. The application of such circuits is illustrated with a counter giving a nine-figure count with a pulse resolution time of less than 1 microsecond. The unit incorporates five transistor decade units, a transistor-regulated power supply, input pulse shaping amplifier, and transitor-driven mechanical counter.

INTRODUCTION

Transistor circuits offer obvious advantages in many applications over the equivalent unit using vacuum tubes. In particular, in scaling circuits for radiation counting the reduced power consumption and increased reliability are important features.

For the cost of transistor scaling circuits to be comparable to those using other counting methods, strict control over the number of transistors per decade must be achieved, because this determines to a large extent the finished cost.

Owing to frequency limitations of junction transistors, the greatest difficulty in the design of scaling circuits is associated with operation at high speeds. In the following sections, designs are presented for operation at various speeds, using simple bistable circuits so as to keep to a minimum the number of junction transistors per decade.

BASIC BISTABLE CIRCUIT

The basic bistable multivibrator circuit (Shea 1957) is shown in Figure 1.

Although this circuit may be modified in various ways (Prugh 1955; Linwill 1955; Bradley 1955) to improve high frequency operation, such modifications give designs which in general either cannot be cascaded, or in which the number of transistors used is excessive (i.e., increased from two to from four to ten transistors). This basic circuit has been analysed elsewhere (Papian 1957). The particular form of triggering shown is by positive pulses to the base of T_1 and T_2, via capacitors C_1 and steering diodes D_1. The capacitors C_1 are returned to appropriate collectors through resistances R_4. This method of triggering is used because at high frequencies the use of steering diodes gives reliable operation.

The switching speeds of transistors have been analysed (Elers 1954). In bistable circuits the actual speed is a compromise. The capacitances C_1 and C_2 enable fast switching of T_1 and T_2, but with associated resistances introduce time constants which limit the speed of operation.

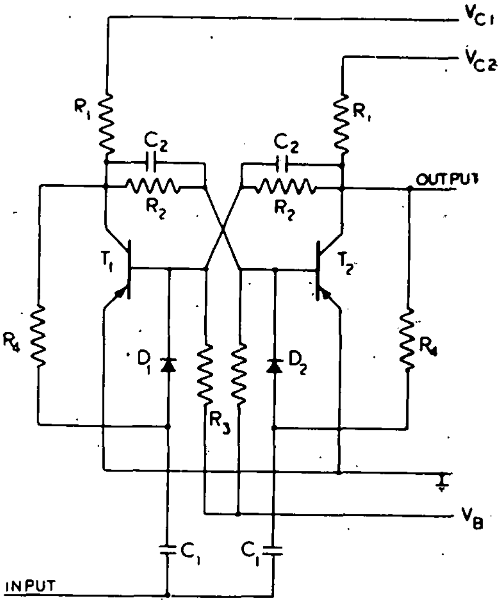

FIGURE 1:—Basic bistable multivibrator.

In Table 1 are given a number of designs of this basic circuit. Using the same circuit, these give increasing speed of operation by reducing the impedance levels in the circuit and using higher speed junction transistors. For high speeds $>$ 1 Mc/sec) the diodes D_1 and D_2 become the limiting factor in determining the operating speed.

The numerical results in Table 1 for maximum operating speed (f max.) are for a cascaded section, i.e., the bistable circuit is driven from a source equivalent to its own output circuit impedance, and loaded by two similar circuits on the output. Under these conditions the maximum speed is reached when the input pulse to drive the circuit is greater in amplitude than the output pulse.

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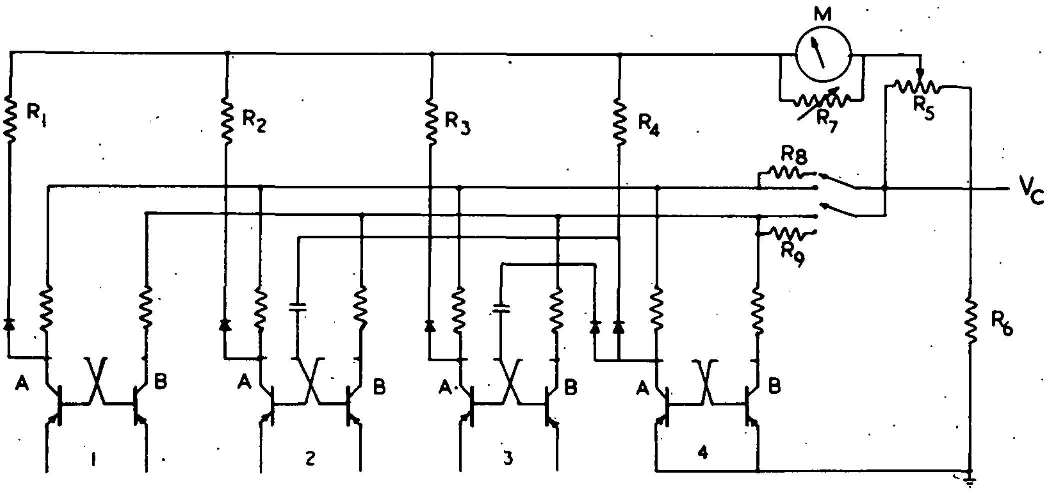
TABLE 1

	1	2	3	4	5	6
R_1 (ohms)	2.2K	2.2K	2.2K	1K	1.5K	560
R_2 (ohms)	10K	10K	10K	10K	2.2K	820
C_2	.002μF	220pF	220pF	470pF	220pF	220pF
R_3 (ohms)	8.2K	8.2K	8.2K	10K	15K	15K
T_1,T_2	OC71	OC45	OC44	2T71	SB100	SB100
C_1	.002μF	220pF	220pF	470pF	220pF	220pF
R_4 (ohms)	10K	10K	10K	10K	3.3K	1.5K
V_C (Volts)	—9	—8	—8	+10V	—4.5V	—4.5V
V_B (Volts)	+3.3	+3.3	+3.3	—3.3	+3.3	+3.3
f max	33kc/sec	250kc/sec	330kc/sec	220kc/sec	1Mc/sec	3.5Mc/sec
Pulse height (Volts)	7	6	6	4	2.5	2.0



METER INDICATION OF COUNT

FIGURE 2:—Basic feedback decode and the use of a moving coil meter to give indication of the count.

	STAGE 1		STAGE 2		STAGE 3		STAGE 4	
	A	B	A	B	A	B	A	B
1	●	○	○	●	○	●	○	●
2	○	●	●	○	○	●	○	●
3	●	○	●	○	○	●	○	●
4	○	●	○	●	●	○	○	●
5	●	○	○	●	●	○	○	●
6	○	●	●	○	●	○	○	●
7	●	○	●	○	●	○	○	●
8	○	●	●	○	●	○	●	○
9	●	○	●	○	●	○	●	○
0	○	●	○	●	○	●	○	●

● = ON   ○ = OFF

FIGURE 3:—ON or OFF states of the transistor collectors of Figure 2 for counts from one to nine.

Because the cost of a bistable unit is largely dependent upon the cost of the junction transistors used, it is not economic to use the high-speed designs for operation at low speeds where the less costly audio transistors are adequate. Similarly, at the lower impedance levels necessary for high speed operation, the power supply drain is much higher than is necessary for operation at low speeds.

DECADE CIRCUIT

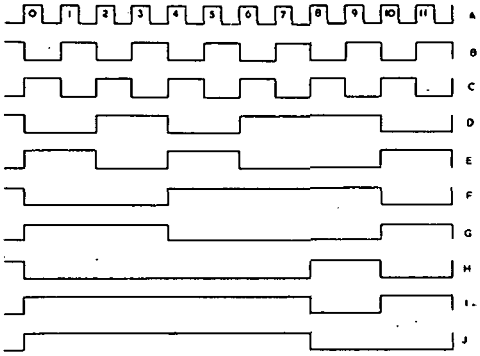
A number of methods can be used to arrange bistable circuits to give decade operation. For example:

(i) Ring counter. [10 transistors, ~ 90 diodes.]

(ii) Count of 2 count of 5 ring. [7 transistors, ~ 30 diodes.]

(iii) Count of 16 with feedback. [8 transistors, 10 diodes.]

The count of 16 with feedback to give decade counting is chosen for simplicity and ease of metering the count. [For very high speeds the count of 2 followed by a count of 5 ring is capable of similar performance.] The basic arrangement is shown in Figure 2. A feedback



WAVEFORM AT COLLECTORS IN DECADE COUNTER

FIGURE 4:—Waveforms at the collectors of each stage of the decade.

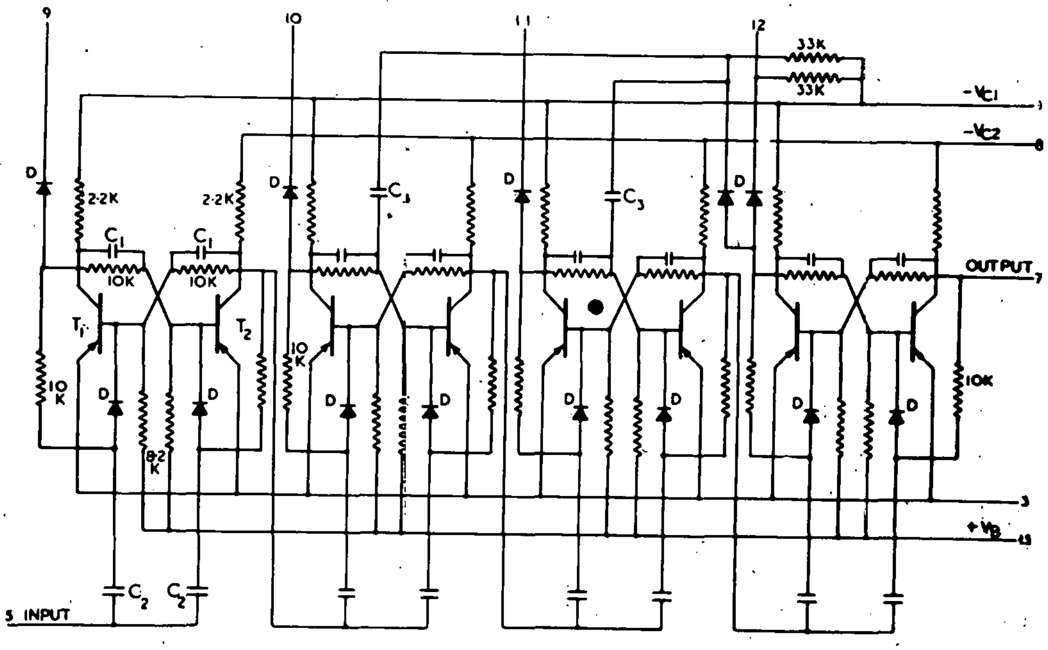


FIGURE 5:—Complete decade counter (metering not shown).

pulse from collector A of stage 4 is fed back to the base B of stages 2 and 3 to give decade operation. This is illustrated in Figure 3, which shows the states of the collectors of each stage for a series of pulses.

A simple form of indicating the count can be achieved using a moving coil meter. The resistors R_1 to R_4 in Figure 2 from the ON collectors of each stage contribute currents to the meter M, which are weighted to give counting indication. With $R_1 = 2R_2$, $R_2 = 2R_3$, $R_4 = R_2$ the meter M can give a progressive indication of the total number of input pulses. R_5 is used to set the zero count, by inserting R_6 in one set of collectors and hence ensuring that this set is OFF. Similarly R_8 and R_7 are used to set the 9 level. Where the meter indication for any digit is incorrect, the table of Figure 3 enables the appropriate resistance in the series R_1 to R_4 to be adjusted.

The resulting waveforms at the collectors of each transistor are shown in Figure 4 (A to I).

At high operating speeds, the feedback pulse from stage 4 to stage 2 is delayed slightly, and hence the change of state which is normally suppressed appears as a pulse through the system. This causes incorrect operation of the decade, usually appearing as a count of 12 operation instead of decade counting, the output waveform appearing as in 4 (J).

The circuit of a complete decade is shown in Figure 5. The method of coupling between stages and applying the feedback pulse is shown. It is evident that stage 4, collector A, is loaded by two input base circuits. Collector A of the other stages is loaded by two base circuits. The DC loading by the metering system does not affect operation provided the meter current is small compared with the ON collector current (i.e., 0.1 and 5 mA, respectively).

Alternative methods of indicating the counting position involve the use of visual indicators operated by the signal from the A or B collectors of each stage of the decade. Either miniature fluorescent vacuum triodes (i.e., Philips DM160) or high voltage transistors and neon indicator lamps can be used. Both methods have the disadvantage that the count must be interpreted from binary notation back to decimal.

FIVE DECADE COUNTER

For a complete counter, it is necessary to vary the design of the various decades to suit the pulse resolution time which is required for that decade. For example, in a five decade counter with a minimum pulse resolution time of the microsecond, the first stage only need operate at this speed. For an adequate margin of safety, the bistable circuit No. 6 of Table I can be made the basis of the decade. For operation at this speed it is preferable to use an extra three transistors per decade as emitter followers (see Figure 6). These are used as coupling elements between the first and second bistable stage, and in the two feedback paths.

For the second decade, the minimum spacing between pulses can, under the worst possible conditions, be of the order of 10 microseconds, and a design using radio frequency alloy transistors is adequate (i.e., Table I, No. 3). A pulse amplifier is necessary between these two stages to convert the 2-volt pulse from the first decade to the 6-volt level necessary to trigger the second decade.

For the remaining decade, the spacing between pulses will be greater than 100 microseconds, and the decade design using inexpensive audio frequency alloy transistors is adequate (Table I, No. 1).

The relative cost of the three basic decades, including interstage amplifiers, is then approximately in the ratio 3: 1.5: 1. It is possible to vary the design of the bistable circuits used in each section of a decade. However, this complicates the metering system design and the mechanical design of the decades, and these factors outweigh benefits achieved in power supply drain and economy of components.

INPUT CIRCUIT

The maximum reliability and maximum speed

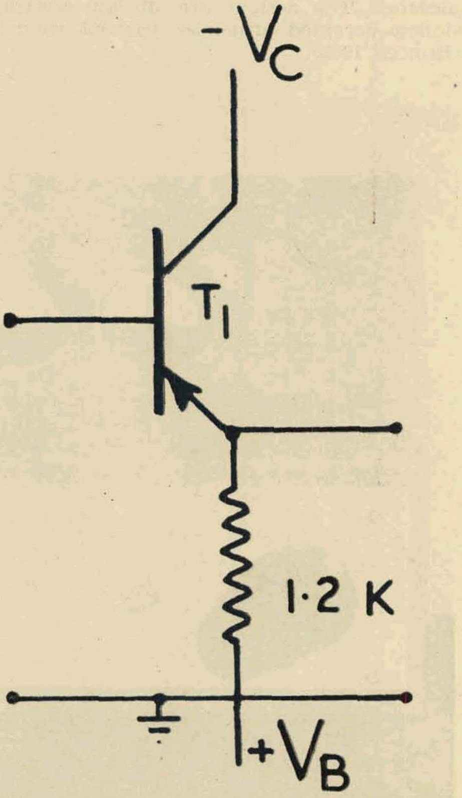


FIGURE 6:—Emitter follower coupling stage for high frequency decodes.

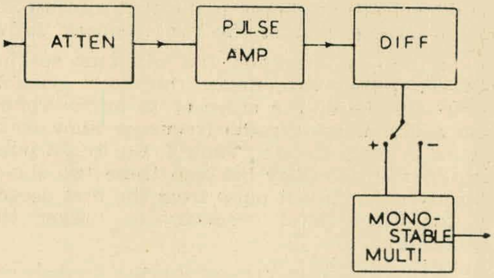


FIGURE 7:—Block diagram of the input system

of operation of the first decade are only obtained with an input pulse of fixed amplitude and optimum width. Hence, an input system is essential to shape the input pulses, and cope with slow and fast input pulses of various amplitudes. The block diagram of a suitable input system is shown in Figure 7.

An input attenuator and pulse limiting amplifier are followed by a differentiating unit and monostable multivibrator. Surface barrier transistors are used throughout to ensure operation at speeds up to the limit set by the first decade. The designs are straightforward, and follow accepted principles (Schenkerman 1954; Hunter 1956).

COUNTING REGISTER

Where long counting periods are used, an economic method of extending the counting capacity is obtained by the use of a counting relay. A monostable multivibrator (Aitchison [in press]) is used to drive the counter, giving a pulse of accurately determined width. This enables the mechanical counter to be used at speeds up to its maximum counting speed (20 counts per second).

POWER SUPPLY

For reliability, it is necessary to regulate the supply voltages V_C and V_B, because any variation or transients in these supplies will give incorrect counting. Separate transistor regulated supplies are used for V_C stages 2 to 5 (—9 volts) and stage 1 (—5 volts). A Zener diode is used to regulate V_B.

COMPLETE COUNTER

A complete 9-figure counter is shown in Figure 8. This is based on the circuits above, and has a pulse resolution time of less than 1 microsecond. Provision is made for positive or negative input pulses, of amplitude from 1 to 100 volts peak.

ACKNOWLEDGEMENTS

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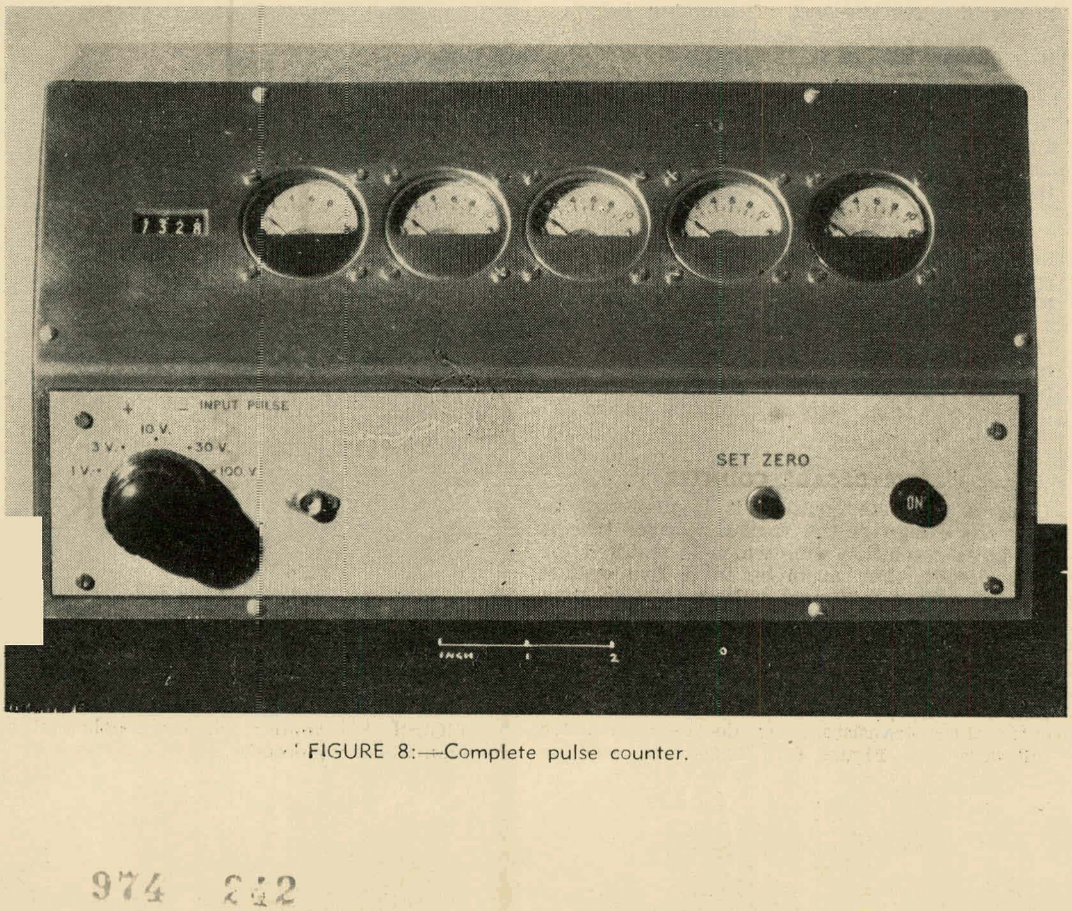

FIGURE 8:—Complete pulse counter.

speed counters, and to K. Murray and C. C. Bennett for the mechanical construction of the unit illustrated in Figure 7.

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