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"RADIOISOTOPES - ATOMIC POWER TOOLS"

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FOLDER Radioisotopes - Atomic Power Tools

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RADIOISOTOPES - ATOMIC POWER TOOLS

For more than fifteen hundred years, man tried to change less valuable elements into gold. Then one day in August 1945, the atomic age burst upon us violently, proclaiming to every man the fact that a new science had been developed for both war and peace. Man had discovered how to make gold, more important, man had created atomic power tools, radioisotopes, to assist scientists unlock new technology that will lead to fascinating adventures in the future.

The story of isotopic transmutation, is a fascinating tale of human quest for knowledge, a story of scientific adventure and success.

"A Half Century of Progress"

Fifty years ago, Medelyeev's "Periodic Law" had already been developed. The atom was then a tiny heavy nucleus surrounded by a solar system of electrons and was considered the smallest indivisible unit. Alpha, Beta, Gamma and X-ray radioactivity experiments were in progress. X-rays were being used in medicine. Albert Einstein at the age of 26 had already written what was to become the most famous equation in science.

By 1911, nearly forty species of elements having different radioactive properties were known, but the species within each of the groups appeared to be chemically identical. In 1913, Soddy proposed the name ISOTOPES for elements that are chemically indistinguishable although their radioactive properties and even their atomic weights are different. During the same period, Soddy, Fleck and others were also studying

Fig.

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changes resulting from the decay of natural radioisotopes and developed the GROUP DISPLACEMENT LAW that explained natural transmutations, i.e., "when an alpha particle is emitted in a radioactive change the product occupies a position two places to the left of its parent in the periodic system, while the emission of a beta particle results in a shift of one place to the right."

In 1919, Rutherford showed that fast natural alpha particles from the radioactive decay of radium caused the conversion of nitrogen into oxygen with the emission of an energetic proton. This first artificial transmutation of one element into a different one also afforded evidence that the proton was a constituent of the atom.

$$_{2}He^{4} + {}_{7}N^{14} \longrightarrow {}_{8}O^{17} + {}_{1}H^{1} + \text{energy}$$ Fig.

Up to this point, scientists had been chipping pieces off the atom. The real attack on the nucleus began in 1920 when there appeared indications from several widely separated sources that an entirely new particle might be an important unit in the structure of the atom. This hypothetical particle, given the name neutron early in the roaring twenties, later identified by Chadwick during the depression of 1932, changed the fate of nations in World War II.

During the roaring twenties, physicists began to consider the possibility of transmuting elements by bombarding them with laboratory accelerated particles. It was evident from earlier transmutation experiments with natural particles that the bombarding particles would have to be a stream of particles of very high energy.

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Some of the first experimenters thought to harness lightning, or the high potentials existing on storm clouds. Van der Graaf developed a method for utilizing the potential acquired by the accumulation of charge on an insulated body in which a large sphere is insulated from the earth and is fed a continuous stream of charge along an insulated belt. Another method for producing high voltages was a cascade or series of transformers, each capable of stepping the voltage up to around two hundred thousand volts.

In 1932, Cockcroft and Walton succeeded in producing nuclear disintegrations by artificially accelerated particles. They used an ingenious multiplying circuit of transformers, rectifiers and condensers capable of producing accelerating voltages up to 710,000 volts. With this device, they accelerated charged atoms of hydrogen up to velocities of about 7250 miles per second.

During this same period, E. O. Lawrence and others developed cyclotrons and linear accelerators for accelerating particles to even higher energies. While others were seeking methods of obtaining very high voltages, Lawrence worked on an idea of speeding a particle up to very high energies by giving it successive accelerations. This idea led to the invention of the cyclotron and by 1932 transmutations were achieved with protons having over one million electron volts.

In 1934, Irene Joliot-Curie and her husband, F. Joliot bombarded aluminum, boron and magnesium with alpha particles from polonium. They produced the first man made radioisotope, phosphorus 30, by (a,n)

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transmutation reaction:

$$_{13}Al^{27} + {}_{2}He^{4} \longrightarrow {}_{15}P^{30} + {}_{0}N^{1}$$ Fig.

During this period, the neutron was also used to pry further into the secrets of the atomic nucleus. Having no electrical charge, the neutron is not effected by the negative electrons in the atomic shell and is not affected by the strong charge of the heavy nuclei. First to attack the atom with neutrons were Enrico Fermi and his co-workers.

In 1935, a group of researchers working under Otto Hahn entered this field and concentrated on uranium. In 1938, Hahn and his co-worker Fritz Strassmann experienced a sensational event. They split the uranium atom in two. The uranium nucleus, on swallowing a neutron, immediately split, then because both fragments contain dozens of protons they repel each other with tremendous force and a terrific recoil which generates the heat of atomic fission. When the nucleus splits, it also gives off extremely powerful gamma rays and one, two or three single neutrons.

The possibility of a controlled nuclear chain reaction accompanying the fission of uranium created a lot of interest in late 1939 and early 1940. By December 1942, a self-sustaining uranium chain reaction was demonstrated in the first nuclear reactor at the University of Chicago. However, the prospects of exploiting atomic energy for war were already recognised and consequently research was directed toward the immediate and urgent problems of the military application of atomic energy.

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In the war years of 1940, 1941, and 1942, several physical research groups emerged in the United States whose aim was centered on the exploration of techniques for releasing power from the atomic nucleus. There were three main groups - one at the University of California in Berkeley, one at Columbia University in New York, and one deliberately misnamed the "Metallurgical Laboratory" at the University of Chicago. Plutonium in kilogram quantities was the objective of the moment. However, it was unsafe to operate a high-powered nuclear chain reactor in the midst of Chicago, with little shielding, limited knowledge, and recognition that the reaction had in it the power to go out of control and cause a disaster. Therefore, the first nuclear FIG. reactor, the X-10 air cooled Graphite Reactor, to be run at power to gain experience in the behavior of reactors was built at what is now known as the Oak Ridge National Laboratory.

In addition to production of plutonium, the uranium reactor produced fission products and other radioisotopes. These radioisotopes were used by biologists in their studies of radiation hazards, chemical researchers used them to develop separation processes and radiochemical analytical techniques, and physicists studied the characteristics of each radioactive species and their yields. Research on radioisotope production processes was so successful that, by the end of the War, some radioisotopes were in routine production.

During the early postwar period, the United States declared, via the Atomic Energy Act of 1946, a policy of the people that, "Subject at all times to the paramount objective of assuring the common defense

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and security, the development and utilization of atomic energy shall, so far as practicable, be directed toward improving the public welfare, increasing the standard of living, strengthening free competition in public enterprise and promoting world peace".

In 1946, the United States, recognizing the many benefits and contributions that could be made to science through the use of radioisotopes, initiated a Radioisotope Production and Distribution Program, thus making radioisotopes available to users outside the atomic energy project in accordance with the objectives of the Atomic Energy Act.

The forging of radioisotopes into practical, economical atomic power tools is still one of mankind's most exciting adventures.

Production of Radioisotopes via Proton, Deuteron and Alpha Bombardment in a Cyclotron

With the development of modern ideas pertaining to the structure of the atom, the possibility of transmutation, previously regarded as fantastic, was given serious consideration by reputable scientists both here and abroad. Many scientists became interested in the possibility of developing methods for building up high potentials whereby protons, i.e., charged hydrogen atoms, could be given sufficient energy to penetrate atomic nuclei. Their purposes were many fold. Some were interested in the way nuclei react, some were interested in nuclear structure and nuclear forces, and others were interested in the production and identification of previously unknown isotopes of naturally occurring elements and their radioactive properties.

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Throughout the period 1930-1945, most radioisotopes used were produced in cyclotrons. Today, the cyclotron largely complements rather than competes with the nuclear reactor for producing radioisotopes. Cyclotron-produced isotopes usually lie on the neutron deficient side of the nuclear stability line. They decay principally by electron capture and positron emission, although some undergo beta decay. In contrast, nuclei formed by neutron capture or as fission products usually have a neutron excess and decay by electron emission, i.e.,

neutron ----------> proton + electron + neutrino Fig.

Cyclotron produced radioisotopes, in general, have a distinct advantage of high specific activity since they ordinarily are a different chemical element than the target and hence can be frequently separated carrier-free. On the other hand, reactor produced radioisotopes, with few exceptions, are usually mixtures containing rather large amounts of the stable isotopes of the same element. However, the reactor can produce much larger quantities of certain radioisotopes more economically than can be done with a cyclotron. Thus, the cyclotron complements the reactor by making additional isotopes available at reasonable costs. Fig.

The standard cyclotron consists of a source of charged particles and a pair of accelerating electrodes placed in a high vaccuum chamber between the poles of a powerful electro-magnet. The electrodes are

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"D-shaped" with their edges closely adjacent to form a narrow gap. The source of ions is an electrically heated filament near the center of the gap which causes ionization of the gas, hydrogen, deuterium or helium contained in the system. Positive ions, namely protons, deutrons, or alpha particles, respectively are projected by the electric field into the interior of the dees. The alternating electric field is provided by a radio-frequency voltage applied to the two electrodes, or dees. The ions travel along a circular spiral path within the dees, crossing the gap twice at each revolution and gaining energy at each crossing. The ultimate energy of an ion at the outer reaches of its spiral path is equal to that gained in a single traversal of the gap multiplied by the number of traversals. In a typical cyclotron, the ions may make between 25 and 100 revolutions. A target may be placed inside the dees to intercept the outer reaches of the spiral path or the ions may be deflected by electrostatic or magnetic means to an external target. This relatively simple principle is basic to the operation of more than 70 cyclotrons which have been built throughout the world.

Transmutation by Protons

When hydrogen is introduced into the vaccuum chamber, positive ions, protons, are accelerated by the electric field. With protons as the incident particles, radioactive capture process of the (p,) and (p,n) type reaction occurs in lighter elements provided the proton has

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sufficient energy to penetrate the energy barrier of the large nucleus. Reactions of the (p,2n) and (p,pn) types also can occur when high-energy protons are used. In fact, protons can initiate reactions in which 3, 4, or even more particles are emitted. However, nuclear processes of the (p,) and (p,d) types are comparatively uncommon because of the small probability of penetrating the relatively high energy barriers. Typical examples of the nuclear process are: Cr-50 (p,) Mn-51, Rb-85 (p,n) Sr-85, Sr-88 (p,2n) Y-85, Mn-55 (p,pn) Mn-54, N-14 (p,)C^{14} and Be-9 (p,d) Be-8. Fig.

Transmutations by Deuterons

When deuterium is used in a cyclotron, deuterons are accelerated by the electric field. Nuclear reactions of the (d,p) type are quite common and have been observed with nearly all elements. Competing with the (d,p) reaction are reactions of the (d,n) type, especially with target elements of low atomic weight. When the incident deuterons have high energies, 10 Mev, reactions of the (d,2n) type occur with target elements of moderately high mass numbers. At still higher energies, reactions of (d,3n) type and others even more complex have been observed. Reactions of the (d,) type and (d,t) type are observed only with deuterons of high energy and elements of fairly low atomic number. Examples of the above reactions are: Cd-114 (d,p) CD-115, Bi-209 (d,n) Po-210, Ag-109 (d,2n) Cd-109, Fe-56 (d,3n) Co-55, Rb-87 (d,) K-85, and Be-9 (d,t) Be-8, Fig.

Transmutation by Alpha Particles

You will recall that Rutherford's first observed transmutation was

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an (α,p) type reaction, which is common with elements of low atomic number. However, as the atomic number increases, the potential barriers preventing the entry of the alpha particle and the emission of the proton become higher and the probability of the (α,p) process decreases, although with the availability of artificially accelerated alpha particles (helium ions), the (α,p) reaction has been detected for nuclei with high atomic numbers. When the energy of the alpha particle is large enough for it to penetrate a highly charged nucleus, other reactions such as (α,2n), (α,np), (α,3n), (α,4n) and (α,3np) can also take place.

Thus we see that the cyclotron is a very versatile tool for producing a variety of useful radioisotopes that cannot be made in a reactor.

Of the dozen or so cyclotrons available in the United States for the production of radioactive isotopes and including the machines in Great Britain at Birmingham and Hammersmith, the Oak Ridge National Laboratory 86 cyclotron has remained unequaled. Fig.

The Oak Ridge National Laboratory 86" cyclotron is novel in that it is a verticle cyclotron, the dees hanging suspended from above. In other respects it is a rather conventional fixed frequency machine which produces 23-Mev protons for nuclear reaction studies and isotope production. Beams of 1000 to 2000 ua are routinely employed for the production of neutron-deficient radioisotopes by proton transmutations. Beams are available from 1 ma (used to simulate radiation damage studies pertaining to the Van Allen belt) to 2600 ua (used in the production of cadmium 109 by the irradiation of a solid silver target).

Means are provided for utilizing the internal beam for purposes

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of high yield isotope production. This involves a system for inserting water cooled target assemblies into the region near the periphery of the dees. A grazing incidence type target (approximately 6" x 10") Fig. is used for cyclotron bombardment when the target material can be applied as surface coatings. The element to be bombarded is coated on the surface in 0.010 - 0.020 inch thickness. Coating is done in several ways; by casting in a shallow recess in the target surface and then machining to uniform thickness; by electroplating; by braising a foil to the target surface; and by using a metallizing gun. The beam whose radial width is 3/16" is spread along the target placed at a small angle to the beam path. Cooling water flowing through the target assembly at 30 gallons per minute dissipates as much as 66 kilowatts of heat to keep the surface temperatures below the target material melting or vaporization point.

Some elements are not amenable to these techniques; others even though they may be applied, may not allow heat transfer rates high enough Fig. to dissipate the large beam power, even when large grazing incidence type targets are used. Non metals, such as the halogens, and other elements not readily available in metallic form must be bombarded as chemical compounds, generally in a thin walled water cooled capsule target. This later technique also may be necessary for metals with very low melting points such as potassium and rubidium. Tubing walls and water layer reduce proton energy to about 15 Mev.

The capsule type target is a tubelar capsule, 200 mil outside diameter surrounded by a jacket, both made of 10 mil seamless aluminum

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tubing. Cooling water at 140 pounds per square inch flows in a 15 mil annulus between the capsule and jacket. In general, somewhat lower bombardment efficiency results and operations at currents less than 1 ma are necessary because of the heat dissipation problems.

Over 250 stable isotopes are available for proton bombardment and many reaction types, e.g., (p,n), (p,2n), (p,2p), (p,d), (p,γ), and (p, alpha) are possible. Facilities are available for the production and subsequently removal from the cyclotron of very short half life material as well as aluminum 26m, which has a half life on the order of 800,000 years. Not only can curie quantities of radioisotopes be produced in a single run (for example, cobalt 57 and iron 55) but quite often specific activities may be obtained which far surpassed neutron produced isotopes. For example, of the two materials mentioned above, cobalt 57 may be produced with a purity such that for every 10 radioactive cobalt atoms present, there is only one stable cobalt atom present. Iron 55 is produced with a specific activity between 3 and 10 times that available under present reactor techniques.

The Oak Ridge National Laboratory 86" cyclotron is available for the production of isotopes other than those routinely sold by the laboratory. Examples of radioisotopes produced by service radiation on the Oak Ridge National Laboratory 86" cyclotron include:

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Be-7	Co-57	Rh-102	Ce-139	Eu-149	Fig.
Na-22	Co-61	Pd-103	Pm-145	Yb-171	
V-48	As-74	Cd-109	Pm-148	Au-195	
Mn-52	Sr-85	I-124,125,126	Pm-150	Tl-202	
Fe-55	Y-87,88	I-125	Eu-146	Bi-207	
Co-56	Tc-95	Ba-133	Eu-147	Pa-231	

Customers for radioisotopes service irradiations using the 86" cyclotron include several universities, hospitals, U. S. government agencies, the Oak Ridge National Laboratory itself, foreign organizations, and two U. S. commercial processors. The commercial processors maintain a service for the production, processing and distribution of carrier free accelerator produced radionuclei on a continuing and easily available basis. In addition to maintaining stocks of commonly used radioisotopes, they make available upon special request any radionuclei which can be produced with a cyclotron.

Although the cyclotron can be used to make practically every radioisotope, the nuclear reactor can make certain radioisotopes cheaper and in larger quantities.

Neutron Transmutations in a Reactor

The nuclear reactor has two unique characteristics, the production of a fission chain reaction and the production of a large amount of neutrons. These features make the reactor a useful productive unit for making radioisotopes. The radioactive isotopes are created by two processes: (1) fission of U-225 or other fissionable nuclei and (2) neutron absorption by non-fissionable nuclei placed in the pile.

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The fission process generally results in two radioactive fragments with a total combined mass less than 235 by the mass of the 1 to 3 Fig. neutrons emitted during the fission event. The fission fragments are very unstable and range from element 30, zinc, to element 64, gadolinium, but a greater percentage of them have mass numbers ranging between 90 to 100 and 135 to 145. Each of these fission fragments decay by a series of or chain of beta-ray emissions and the intermediate decay products are the source of fission products. The fission products, because they differ chemically from the original uranium and each other, can be isolated and purified by chemical processing. The separated fission products are, however, mixtures of stable and radioactive nuclei of the same species rather than pure specific isotopes.

Production of radioisotopes by neutron absorption yields a much wider variety of radioelements than can be obtained through fission. About 275 stable nuclei are available, at various enrichments, for neutron bombardment to produce a variety of radioisotopes. Radioisotopes

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of most elements may be made by simple neutron capture in which (1) Fig. only a gamma ray photon is emitted and the product formed is an isotope of the target element and (2) a proton or alpha particle is emitted and the product formed is an isotope of an element different than the target.

Radiative Capture (n,)

Capture of neutrons by a nucleus is a process which takes place easily, since no electrostatic barrier has to be overcome. In the radiative capture of neutrons, the final nucleus is an isotope of the target nucleus and, if it is radioactive, it usually disintegrates with the emission of electrons. Absorption of a neutron into the nucleus of an atom does not change its chemical nature, it merely produces another isotope of the same chemical element. Neutrons of relatively low energy, are particularly effective for the radiative capture reaction. The best example of the radiative capture process is the production of cobalt 60.

$$_{27}Co^{59} + {}_{0}N^{1} \text{ ---------- } {}_{27}Co^{60} + \gamma$$ Fig.

Splitting or Ejection of Charged Particles; (n,p), (n,)

Processes of the (n,p), (n,d), and (n,) types can also take place when the neutron supplies sufficient energy to overcome the force binding the charged particle in the compound nucleus and to also permit it to escape. This in general, requires fast neutrons with increasing energy as the atomic number of the target material increases. Processes of the (n,) type are not common with target materials of atomic numbers

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exceeding 40. As a general rule, when the neutron has sufficient energy to make the (n,) reaction possible with an element of high atomic number, other processes, such as (n,2n) take place at about 10 Mev in preference to the (n,) reaction. At incident neutron energies above 30 Mev the compound nucleus may eject three neutrons or even two neutrons and a proton, i.e., (n,3n) or (n,2np).

The ejection of charged particles results in an element different than the target element and it therefore can be isolated as a high specific activity isotope. Examples of radioisotopes produced by these processes are:

$_{7}N^{14}$ (n,p) $_{6}C^{14}$ and $_{16}S^{32}$ (n,p) $_{15}P^{32}$

$_{3}Li^{6}$ (n,), H^{3} and $_{17}Cl^{35}$ (n,γ) $_{15}P^{32}$

$_{6}C^{12}$ (n,2n) $_{6}C^{11}$

<u>Production of Radioisotopes</u>

Other than the nuclear reactor itself, the main parts of the radioisotope production process are target preparation, facilities for inserting the target into the reactor and removal of the irradiated target, chemical processing facilities, storing and packaging facilities and decontamination and waste disposal facilities. Without going into excessive detail, we might discuss some of the important considerations concerning techniques of radioisotope production.

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Target Preparation

Target materials are generally encapsulated in aluminum containers before insertion in the reactor. In some instances, it may be desirable to first seal the target in a quartz ampoule prior to enclosure in the aluminum container. Glass is not used because of high neutron absorption. Solid metallic elements are generally used as targets when they can be obtained with high purity. Stable oxides and carbonates are also used but consideration must be given to possible multiple production from compound targets such as from compound elements containing chlorine which yields S-35, P-32, and Cl-36. Further, targets should not volatize, decompose appreciably or produce materials that will attack the container. For example, salts such as nitrates decompose and organic compounds may char, often producing pressure which might rupture the target capsule and contaminate the reactor cooling system. Consideration must also be given to self-absorption of neutrons by relatively large prices of target material and the effect on activation because of localized flux depression.

Fig.

Production Rates

The production of radioisotopes by neutron transmutation can be estimated by the following equation:

$$A_{(t)} = \frac{W N \sigma f}{M\,(3.7 \times 10^{10})} \left(1 - e^{-\lambda t}\right)$$

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Where: $A_{(t)}$ is curies of radioisotope produced. Fig.

W is grams of target element
N is Avogadro's number, 6.023×10^{23} atoms per gram atomic weight
ϕ is neutron flux in target area, neutrons/ (cm^2) (sec)
σ is activation cross section in Cm^2
f is isotopic abundance of the target isotope in the target
M is atomic weight of the target isotope
λ is the disentegration constant, 0.693/ half life of the product isotope, and
t is the irradiation time.

The above equation assumes that the amount of target material remains essentially unchanged throughout the irradiation. However, in high fluxes or targets having very large cross sections, allowances must be made for target burn-up. Similarly allowances must be made for burn-up of the radioisotope produced if it has a large neutron capture cross section as well as the fact that the desired radioisotope is sometimes produced by the decay of a primary radioisotope.

In general, the activity of the short-lived isotope accumulating in the target depends primarily on the strength of the neutron flux when its half-life is less than the irradiation time. The activity of a long-lived isotope accumulating in the target is governed by the product of neutron flux and irradiation time.

Long-lived radioisotopes can be produced from targets encapsulated in aluminum containers placed within the reactor core. The targets are inserted and removed during reactor shutdowns. Short-lived radioisotopes are produced using facilities designed to insert and remove the targets Fig.

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while the reactor is operating. For example, the Oak Ridge Research Fig.
reactor has a hydraulic facility which carries targets, by water pressure, into and out of the reactor while the unit is operating. Upon removal of the irradiated target from the reactor it may be necessary to process the radioisotope immediately to obtain the optimum yield of short-life isotopes. On the other hand, it may be desirable to defer processing of the target to permit decay of undesired isotopes Fig.
and/or build up of the desired radioisotope.

Radiochemistry

The operation of a reactor results in a built-up of fission products to such a high concentration that they ultimately interfer with the operation of the reactor. Consequently, it becomes necessary to remove the uranium fuel from the reactor and reclaim the valuable fuels by removing the contaminants. Thus from the very beginning of the chain reaction, research in chemistry was primarily concerned with isotope separation and purification processes. The English Scientist, A. Smithells, aptly described radiochemistry in 1908 when he called it "The chemistry of phantoms".

Since the Oak Ridge National Laboratory Graphite Reactor was the first capable of producing large amounts of fission products, plutonium and radioisotopes by neutron transmutation, chemical studies of these new substances occupied the attention of many chemists at what is now the Oak Ridge National Laboratory. These chemists pioneered methods of radiochemical analysis, tracer techniques, labeled compounds, develop-

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ment of ion-exchange processes for the separation of pure rare earths Fig.
and are credited with the discovery of the element promethium. Research and development along these lines was very successful, and it supplied information which enables us to predict the rate of formation of most fission products and neutron transmutation in the reactor.

The National Laboratory has continued to develop methods for the Fig.
production and chemical processing of these isotopes. Perhaps the most outstanding developments in chemical process development are ion-exchange and solvent extraction processes for separation and purification of various elements from gross mixtures of fission products. Each radioisotope, however, is a separate production problem, involving its own combination of requirements for target material, irradiation Fig.
time, chemical purification process and safety precautions.

The United States pioneered radioisotope production and it still maintains world leadership in the production and distribution of radioisotopes. We have come a long way since the first shipment of 1 millicurie of carbon 14 on August 2, 1946. Therefore, rather than elaborating on details pertaining to process, equipment, facility and instruments development during the past 15 years, let us review some of the exciting things we are doing today. However, before we do, it might help to first review growth and trends in the AEC Radioisotope Production and Distribution Program.

Distribution and Availability of Radioisotopes

To describe the growth and trends of radioisotope production and distribution, we must first establish a reference of comparison.

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Unfortunately, most people are impressed by large things. For example, the tremendous devastation following an atomic bomb explosion, huge atomic power reactors, atomic powered submarines and rockets exploring the vastness of outer space. We are talking about atoms.

The accomplishments from the use of radioactive atoms will probably be of greater ultimate benefit to mankind. Radioisotopes have been proclaimed by scientists as "the most useful research tool since the invention of the microscope". Yet, if all the radioactive atoms produced and distributed by the Oak Ridge National Laboratory under the Commission's Radioisotope Distribution program during the past 15 years were combined into a single mass, it would weigh only seven (7) pounds.

At first radioisotopes were only allotted for research and medical use. By late 1947, the increased supply of radioisotopes permitted liberal allocations for research in all fields. However, the demand seemed to keep just a little ahead of production capacity, and by early 1948 it was apparent that production facilities would soon be strained to their limit. The rapid increase and demand for radioisotopes made it essential for Oak Ridge National Laboratory to expand its production Fig. facilities. A new radioisotope processing area was provided to meet the special requirements arising from the handling and processing of radioactive materials.

In a comparatively short time, isotope distribution became a well established and growing U. S. program. The Oak Ridge National Laboratory, although primarily a research organization, developed as a primary supplier of radioisotopes. Later other Commission laboratories began

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producing radioisotopes to meet various specific needs as they arose. In addition to Oak Ridge, isotopes are now produced at Brookhaven National Laboratory, Argonne National Laboratory, the National Reactor Testing Station, Savannah River, Hanford, and Mound Laboratory. In short, the entire resources available to the Commission are called upon as necessary to assist in meeting the needs for radioisotopes. However, the Oak Ridge National Laboratory is the main Commission center for distribution of radioisotopes throughout the world. The number of radio- Fig. isotope products offered for sale has grown from only two: Carbon 14 and iodine 131 to more than 100 at present. During this time, the techniques of manufacturing, handling, and shipping large amounts of highly purified radioisotopes have been developed to the point where most of the operations are now routine and similar in many respects to those of other industries.

A primary objective of the radioisotope development program is to devise production techniques for all useful radioisotopes with half-lives of sufficient length to permit distribution. This objective has been largely accomplished, although new products are still being added to the catalog of radioisotopes available. Continuing efforts are aimed at increased radiochemical purity, lower costs, and a higher specific activity for all radioisotopes distributed.

Among the milestones of this program can be listed a great number of achievements. First is the extremely rapid development of isotope technology during the past 15 year period. Second, of course, has been the great advance in availability of isotopes not only in number and quality but equally as important, in dependable supply and reduced

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costs. Third, is the pioneering and product leadership provided by Commission facilities in developing isotopes, isotope compounds, radiochemical services, special types of sources, instrumentation and safe handling devices.

Another milestone in the program was the completion of the Fission Product Development Laboratory for the separation and purification of kilocurie quantities of important long-lived fission products. Research and development of radiochemical processes on a pilot plant scale is one of the important functions of the Oak Ridge National Laboratory. Initially, the Fission Product Development Laboratory planned to produce individual fission products intermittently during the year as the needs arose. Today, it is apparent that the demand for these products is larger than initially anticipated and additional equipment has been installed in the Fission Product Development Laboratory to increase the availability of all principle fission products. The Fission Product Development Laboratory currently has the capability for separating the purifying up to 500,000 curies per year of cesium 137 as well as separation and purification of 250,000 curies of strontium 90, 4,000,000 of cerium 144, and 500,000 of promethium 147 plus gram quantities of technetium 99. Fig. Fig. Fig. Fig. Fig.

Another milestone in the radioisotope production distribution program is the Radioisotope Process Development Laboratory which will be completed late this calendar year. It will be used to develop new processes and equipment necessary to improve the availability and purities of the more than 100 radioisotopes routinely available for research and industrial applications. It will also be used to develop Fig.

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larger and improved sources for industrial and research applications.

Today, the business associated with radioisotope usage is growing in sizeable proportions. Commission facilities are becoming producer wholesalers of radioisotopes with commercial firms purchasing bulk quantities for redistribution and reprocessing of the isotopes into labeled compounds, certified drugs for direct medical use, and other manufactured radiation and energy devices. The continuing growth of the radioisotope utilization is illustrated by Tables I and II in Apendix A.

The growth in isotope utilization through 1958 was accomplished with little advertisement or promotional effort. However, now that commercial advertising and promotional efforts have increased the public awareness of advantages and benefits of radiation applications, sales of radioisotopes in domestic and foreign markets has accelerated. Fig.

The existance of several institutions and commercial firms both here and abroad devoted to the task of producing and distributing radioisotopes and related services means that a certain rivalry inevitably has grown up. We feel that competition is good since it serves to stimulate us and hastens the day when mankind can enjoy the full benefits of successful nuclear energy. The Commission, however, is not trying to compete with the several institutions and many industrial firms both here and abroad that are providing radioisotopes and related services. In fact, we do very little advertising, we provide no field services, we price radioisotopes such that industry is encouraged to enter the business, and we go so far as to provide technology and withdraw from some of our most profitable and productive services in order

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to foster private participation. In fact, no organization will do so much or go so far as the Commission does to assist its competitors.

The Commission follows a policy of furnishing materials and services only when they are not reasonably available commercially. As the private industry has grown, the Commission has recognized the development of commercial sources of supply by gradually restricting its own role as a supplier. Industry is now in a position to furnish a number of materials and services formerly only available from the Commission. Prior to 1950 nearly all tagged compounds were prepared in Commission laboratories. In 1949- to 1950 the Commission gave financial assistance to the development necessary for the synthesis of selected labeled compounds by private groups. Once private groups obtained the appropriate techniques and experience in preparing these materials the Commission in 1955 withdrew from providing them. Today, hundreds of different tagged compounds are stocked, and made routinely available by dozens of companies.

In 1958 the Commission withdrew from providing cobalt 60 encapsulation services in favor of more than 10 companies who notified the Commission that they were prepared to encapsulate large cobalt 60 sources. In 1961 the Commission withdrew from providing cobalt 60 reasonably available from private reactors in keeping with its policy to withdraw from providing materials and services reasonably available commercially. The Commission also discontinued providing gamma radiation services to private groups. Now more than 120 academic and industrial organizations have their own radiation facilities and many of these are willing to perform irradiation services for others on a commercial basis.

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curie or less in quantities greater than 100,000 curies.

In 1949, cesium 137 was available at a price of $50,000 per curie. New facilities and processes make it possible to sell cesium 137 at prices as low as $1.00 per curie in large quantities. Similar examples of price reductions can be cited for other radioisotopes and substantial price reductions are still possible as sales volumes increase in the future.

The future trend in radioisotope prices, however, is somewhat uncertain. Although substantial price reductions are possible if the Commission were to remain the sole supplier of radioisotopes, private producers and foreign suppliers are competing for radioisotope markets that were formerly supplied by the Commission. The Commission could, of course, enter into active completion and because of its unique facilities and capabilities, remain the sole domestic supplier of radioisotopes. But such action would be contrary to Commission policy and Section 81 of the Atomic Energy Act. Thus we appear to be approaching a point wherein prices for radioisotopes may have to be held high enough to encourage private production and possibly increased to recover costs of production and distribution as the Commission withdraws from providing those radioisotopes available from private producers.

The major problem we face is that industry is only taking over those radioisotopes providing the bulk of revenues which enable the Commission to supply a complete service at reasonable costs to radioisotope users, leaving more than 95% of the radioisotopes which are not large volume sales items. The net effect may be a need to increase the prices of the remaining radioisotopes supplied by the Commission to recover some of

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Pioneering New Frontiers

Development to date in applying radioisotopes and radiation to increasing human well being and to economic gains through progress in industry represent only the beginnings of what can be accomplished. The contribution of this new atomic power tool to scientific work is broad and continuous. The Commission through its research program regularly supports agricultural, biological, medical, physical and chemical projects in applying radioisotopes and radiation. These and other projects require the development of improved radioisotopic products and processes to keep pace with the changing needs of science.

Higher specific activity radioisotope preparations are often needed. Additionally, development of new applications and techniques utilizing radioisotopes also continues to require improvement in the safety, quality, quantity and cost of available radioisotope products. For example, to study a biological system using radioisotopes, it usually is necessary to employ very high specific activity preparations to avoid undesirable side effects because biological systems in particular are extremely sensitive to "poisoning" by inert materials. More wide spread use of radioisotopes in tracer applications requires development of new shorter half-life isotopes. Potential applications using radiation and heat energy, on the other hand, requires development of new and improved processes capable of economically recovering large quantities of long-life fission products.

In addition to new and expanded uses of relatively small amounts of radioisotopes for such operations as tracing and gauging, direct use of the ionizing properties of high intensity radiation in production

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processes is under development. The irradiation of a chemical system produces a variety of changes, depending both on the system and the time of irradiation. Radiation has already demonstrated special application in improvement of polymers, sterilization of drugs, pharmaceuticals and preservation of foods. Ionizing radiation also represents a new and potentially an important source for process energy. Since radiation directly produces a variety of ions, molecular fragments and excited molecules, it provides a new tool to supplement pressure, temperature and catalyst in chemical systems.

Both the U. S. Army Quartermaster Corp. and the Atomic Energy Commission are pioneering technology for radiation preservation of foods. Relatively low doses of radiation can inhibit the growth of microbes for days or weeks during the usual conditions of distribution and storage of perishable foods. Low dose preservation would in particular reduce the necessity for freezing perishable foods that are to be sold within a short time, and extend shelf life. The Army's sterilization program, designed to improve logistics and acceptability of military rations, may ultimately have civilian applications. These programs although in an early state of development, require research irradiators with as much as 100,000 curies of cobalt 60 and up to 200,000 curies of cesium 137 for experimental studies on the effects of radiation and its ability to preserve foods. The U. S. Army Quartermaster Corp. is also completing construction of a facility which will contain a 1,000,000 curie cobalt 60 source.

In addition to the fascinating programs underway to develop the radiation potentials of radioisotopes there are even more fascinating

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programs underway that show considerable interest in the heat generated by fission products currently stored in underground waste tanks. Isotopic power and heat generators have particular glamor and practical application in remote regions where logistics of maintainence and service are costly considerations. Isotopic power devices offer long-life reliability, unattended operation, compact size and the sole means for providing power in certain environments. Potential applications for isotopic power devices include; auxiliary power in space, remote navigation and weather monitoring stations, deep sea oceanographic probes, navigation bouys, and under-sea power supplies. Radioisotopic heat sources also have interesting potential for supplying heat to Arctic installations and equipment, for the desalination of water in isolated regions, and for subterranean heating applications such as the recovery of viscous oils from oil bearing sands.

Each of the above new frontiers utilizing atomic power tools is an exciting program but the development of processes and products to meet the needs of these new applications is truly the big challenge and greater scientific adventure.

Process Development

Most people are impressed by large things. Therefore, before we discuss the challenges of separating megacurie quantities of the fission products or the fabrication of massive radiation sources, I would like to first discuss process and production developments that are making available new radioisotopes for research and medicine.

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Cyclotron Products

Cyclotron produced radioisotopes have much potential in tracing applications. Improvement in the production and availability of these products is one frontier that has gone unchallenged for too long a period. Recently the Commission completed the Oak Ridge Isochronous Cyclotron which will be used solely for research to study nuclear phenomena in the largely unexplored energy region between 20 and 100 Mev. The 86" Oak Ridge cyclotron has been turned over to the Radioisotope Production Program and essentially all of its time is now available for the production of radioisotopes. Although the Commission withdrew from providing processed radioisotopes produced by cyclotron irradiations, the Commission continued to irradiate targets for industrial suppliers and thus remained the primary source of cyclotron produced radioisotopes within the United States because private industry has not provided a reliable source of supply for these radioisotopes, independent of Commission facilities. Consequently, the Commission, in order to support this facility through revenue from sales of cyclotron produced radioisotopes, will now have to increase the utilization of the 86" cyclotron via increase sales of these radioisotopes. It is anticipated that with some additional work on target development and promotion of these radioisotopes, that the availability and cost of cyclotron radioisotopes can be significantly improved.

Reactor Products

In addition to development of cyclotron production, technology for improved production in reactors is being pursued on several frontiers. Isotopic enrichment of stable and radioactive nuclei, higher flux

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reactors and continuous production methods have potential for new, high specific activity radioisotope products.

Isotopic enrichment of radio-nuclides by electromagnetic separation, thermal diffusion, electromigration, or burn-out of unwanted nuclides in high neutron fluxes introduce new possibilities for the use of extremely high specific activity radioisotopes in all scientific fields. Isotopic enrichment of stable nuclei and irradiation in high flux reactors also enhances the availability of high specific activity radioisotopes. Isotopes produced by (n,γ) reactions such as cobalt 60, or fission products such as krypton 85 are associated with large quantities of stable isotopes. Kilocurie quantities of cobalt 60 could be enriched to specific activities of about 1,000 curies per gram and cesium 137 could be separated from stable cesium isotopes with a three fold increase in specific activity. The greater specific activity obtained by enrichment of the radio-nuclide can find improved usefulness in such applications as teletherapy, radiography, and isotopic power devices.

Another interesting potential technique for reactor production of enriched radioisotopes involves fixation of the stable nuclei onto an inorganic resin and elution of the radioactive nuclei from the resin after irradiation. This technique postulates that the energy absorbed during radionuclei formation will break the bond fixing the original stable nuclei onto the resin. Thus greater enrichment of the radionuclei may be obtained because a large percentage of the stable nuclei will remain fixed on the resin during elution of the product radionuclei.

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Isotopic enrichment of stable nuclei has also resulted in considerable improvement in the availability and quality of many radioisotopes. For more than ten years there was an urgent demand for the 4.5 day calcium 47 radioisotope for use in diagnosis and therapy relevant to bone cancer. Electromagnetic enrichment of calcium 46 and neutron bombardment at high flux to produce calcium 47 by a Ca-46 (n, γ) Ca-47 reaction, now provides a relatively high specific activity calcium 47 on a routine basis.

Included in the new frontiers relevant to reactor usage are studies of burn-up in ultra-high flux reactors and investigation of the production of radioisotopes not now available by the use of (n,p), (n,γ), (n,2n) and triton reactions. Continuous "radioisotope factories", such as in-pile loops to circulate target materials through the reactor are also under study for possible production of higher specific activity carbon 14 and iodine 125. For example, iodine 125 is produced by irradiation of xenon 124, either natural or enriched, by the following reaction:

$$\text{Xe-124} \xrightarrow{(n,\gamma)} \text{Xe-125} \xrightarrow[18h]{\text{E. C.}} \text{I-125}$$

The use of a loop to circulate xenon provides a means of removing the xenon 125 from the reactor, thus preventing the loss of iodine 125 by burn-up (σ 1200 b) to iodine 126. In addition to producing a higher specific activity product, the in-pile loop permits recirculation of the target materials and thus has potential for lowering production costs.

Fission Products

In addition to reactors and accelerators as sources for radioisotopes,

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the streams from processed spent reactor fuel are reservoirs of large quantities of fission product radioisotopes. The tens of megacuries of cesium 137, cerium 144, promethium 147 and strontium 90 being produced annually have long been recognized as a potential valuable source of radiation and heat energy. The possibility of recovering these valuable radioisotopes has been enhanced through process development studies conducted in the Fission Product Development Laboratory. This facility has also provided large kilocurie quantities of the major fission products for source and application development studies. However, if large scale utilization of fission products are to be developed, newer and less expensive processes have to be developed.

Higher specific power sources are being demanded for auxiliary power devices. Promethium 147 must be free of promethium 148 and also free of rare earths. Strontium 90 must be low in strontium 89 content and free of cerium 144, barium and calcium to meet requirements for minimum weight shields and maximum power density. The weight of barium produced in fission is about equal to that of the strontium. The weight of calcium induced during the processing of reactor fuels may be 20 to 40 times the weight of the strontium. However, the chemistry of calcium, strontium and barium is quite similar. Thus, batch precipitation processes for producing ultra pure strontium 90 or separation of pure promethium 147 from rare earths are impractical. Here then is the challenge. First, to get rid of undesirable radioisotopes, the product must be stored a sufficient time to permit the undesirable nuclei to decay. Second, new and more economical processes must be developed

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to achieve the higher purity products now desired. Work on development of ion-exchange technology and solvent extraction technology shows considerable promise and potential to meet changing needs for higher purity fission products.

Processes have already been developed to meet current and short range future needs for fission products. Crudd fractions of cerium 144, promethium 147 and strontium 90 are currently being isolated from the Purex waste stream in large quantities and cesium 137 is being recovered from the waste supernates stored in underground tanks at Hanford. These crude fission product fractions, of course, must be further purified to obtain the desired high purity fission product radioisotopes.

Cerium 144

Cerium 144 is recovered from the Purex raffinate stream by precipitation of cerium and rare earths as a cerium-rare earth double sulfate at a PH of #0.4 using sodium sulfate and tarteric acid. The tarteric acid is used to complex iron and prevent its precipitation. If further initial separation of cerium from rare earths is desired, the above cake can be dissolved and the cerium separated from the rare earths by a peroxy-acitate precipitation. Either product may then be shipped to Oak Ridge for further purification. Cerium is purified by batch extraction with di(2-ethylhexyl) phospheric acid solvent to remove yttrium and rare earths. The cerium is then precipitated as an exalate and calcined to cerium oxide at 700°C.

Promethium 147

Following removal of the rare earth fraction from cerium, promethium

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is purified by ion exchange separations. The rare-earth fractions are absorbed on Dowex 1 resin and eluted with citric acid. Extremely good separation of promethium 147 is obtained by this method. Ion exchange systems using ethylene diamine tetraacetic acid (EDTA) and di-ethylene-triamine penta acitic acid (DTPA) have also been developed for separation and purification of rare earths. Solvent extraction systems using tri butyl phosphoric acid (TBP) and di(2-ethylhexyl) phosphoric acid (D2EHPA) for purification of promethium are being developed at the Oak Ridge National Laboratory.

Strontium 90

The purex raffinate stream is processed through a lead carrier sulfate precipitation step and a carbonate-hydroxide metathesis step to recover a concentrate mix of strontium, cerium and rare earths. Strontium is separated from the cerium and rare earths by an oxalate precipitation. The semi-pure strontium fraction is then sent to interim storage to allow the strontium 89 to decay. The aged strontium is then purified by either ion exchange or solvent extraction processes, the latter being the newer and better process.

Large kilocurie quantities of strontium 90 were purified using ion-exchange equipment in the Hanford High-Level Radiochemistry Facility. In the first run, a mixed copper-hydrogen cycle was used to obtain a sharp separation of strontium from cerium and calcium. However, the cerium and yttrium that collected behind the copper caused an intense, localized radiation that reduced some of the cupric ion to metallic copper and also caused severe gassing, channeling and partial plugging

of the column. This was an unexpected occurance and illustrates the kinds of unusual things that make hot chemistry so much different from cold or tracer level chemistry. To eliminate the above difficulties, calcium was used as the restraining ion in the second run and it proved very satisfactory. The strontium 90 product is essentially free of cerium, calcium and barium.

The Oak Ridge National Laboratory developed a solvent extraction process using di(2-ethylhexyl) phosphoric acid (D2EHPA) as an extractant. This process was modified and adopted by Hanford for use in the Hot Semi Works pulse-columns. The aged crude strontium fraction is treated with 50% excess ethylene diamine tetraacetic acid (EDTA) to complex iron and lead and then the feed stream is adjusted to a pH of about 4.2. The feed stream is countercurrently contacted with 0.4M di(2-ethylhexyl) phosphoric acid plus 0.2M tri butyl phosphoric acid in Shell E-2342 diluent. Under these conditions, all the strontium, calcium and about 10% of the cerium present in extracted by the solvent. Sodium and other undesirable metal ions are stripped from the solvent by a 0.6M citric acid scrub stream, pH 2.6 to 3.0, introduced at the top of the first column. Strontium is removed by counter current contact with 1M citric acid in the stripping column. The calcium, cerium and other rare earths remain in the solvent. A two cycle solvent extraction system produces an ultra-pure strontium 90 product.

The di(2-ethylhexyl) phosphoric acid solvent extraction system is being studied at Oak Ridge National Laboratory. This system shows excellent potential for economical direct recovery of ultra pure products of strontium, cerium, promethium and selected rare earths from Purex waste streams.

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Cesium 137

Cesium 137 with a very low cesium 134 content is currently being recovered from aged waste supernates presently stored in underground waste tanks at Hanford. The waste supernates contain cesium 137. The strontium 90 and rare earth fission products are mostly in the sludge at the bottom of these tanks. The supernate is centrifuged to remove particulate matter, and passed through an inorganic ion exchanger wherein the cesium 137 is absorbed and concentrated on the ion exchange bed. This cesium recovery operation is a relatively simple process and cesium absorbed on the ion exchange bed can be transported in a safe manner. In fact, this is exactly what we are doing today.

Four hundred gallons of Decalso, an alumino-silicate compound costing about $700 per ton, is contained inside a Shielded Transfer Tank. Supernate waste is passed through the Decalso until 50% of the cesium breaks through. At this point, the 400 gallons of Decalso contain approximately 30,000 curies of cesium 137 that are ready for shipment to Oak Ridge.

The cesium 137 is eluted from Decalso at Oak Ridge with 8 to 10 volumes of 1 M ammonium sulfate, concentrated by ammonium alum co-crystallization, purified by a cesium chloroplatinate precipitation step, and converted to the cesium chloride powder.

The above, of course, just briefly skimmed over the area of fission product process development. In doing so, we have discussed accomplishments and only hinted at what is still uncomplished. High level radiochemical processing is still an infant. Oxidation reagents decompose, so do ion exchange resins and solvents, foreign substances are formed by

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radiation, and the hot atoms sometimes won't precipitate as readily as cold atoms. All these problems in addition to the usual problems associated with remote hot cell operation illustrate that no radiochemical process is a proven process until demonstrated in a full scale test.

Much additional research and development must be done at both Oak Ridge and Hanford before economical processes will be developed for the isolation and purification of megacurie quantities of fission products on a routine production basis. The recovery of fission products also affects reactor fuel reprocessing and waste disposal systems and these factors must be explored to optimize the economics of the whole system. Further, in addition to development of new processes, new equipment must be developed to meet the requirements of new process technology and product handling.

The high radiation levels and associated heat generated by large quantities of concentrated fission products truly represents pioneering of new technological frontiers. Development of high yield, low cost solvent extraction systems have potential for isolation of long life fission products which can be either safety stored for indefinite periods or made available for radiation and heat applications. Preliminary analysis indicates that it may be more economical and safer to isolate specific long-life fission products for storage as pure or semi-pure solids than it would be to concentrate and calcine the gross mixture of fission products contained in the high level waste streams.

Product and Source Development

Increased interest in radioisotopic sources for thermoelectric generators, thermoionic generators, regenerative fuel cells, heat and

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radiation devices requires an accelerated program to develop radioisotopic products that will meet the ridged source safety requirements for these new applications. Currently, the interest in large scale fission product sources is related to the use of strontium 90, cesium 137, cerium 144, and promethium 147, depending upon the life and specific criteria of the intended application.

The increased interest in large heat and radiation source applications will require a better definition of the physical, chemical and nuclear characteristics of large fission product sources to insure safe application. For example, up until a few months ago, the actual heat output and shielding requirements for large fission product sources had not been measured. In fact, radiation sources and heat sources are still being designed based upon decay scheme data, calculated power outputs and shielding estimates rather than measured values. Additional research is needed to obtain data and actual measurements to better define such source characteristics as; specific activities, bremsstrahlung emission, practical power density, thermal conductivity, radiation stability, thermal stability and chemical stability.

Safe insoluble radioisotope source compounds and improved encapsulation techniques must also be developed to achieve a high degree of utilization efficiency and safety. Not only must the initial product compound have chemical, thermal and radiation stability, but so must the products of radioisotope decay. For example, cerium 144 decays by:

$$4Ce\text{-}144\ O_2 \xrightarrow[285\ da]{\beta' \nearrow} 2Pr_2^{144}\ O_3 \xrightarrow[17.5\ min.]{\beta' \nearrow} 2Nd_2^{144}\ O_3 + O_2 \uparrow$$

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Consequently, very large sources of cerium oxide must provide for the release of oxygen resulting from decay to neodymium. Similarily, changes in chemical and physical product compositions must be considered in the design and construction of other large radioisotopic sources. This includes, of course, source safety testing whereby designs of source containers and source compounds are proved by actual fabrication testing.

Current costs of source encapsulation are rather high. Improved materials and techniques of encapsulation, such as electroplating, gas phase deposition, metal and ceramic flame spray coatings, and welding must be developed to expand the variety of source configurations and extend the range of radioisotope source applications.

In summary, the Commission is continuing to pioneer new frontiers of technology to develop new, more economical processes, products and sources to meet the changing and advancing needs of science. Currently, the Commission has the capability and technology to provide a few megacuries of the various fission products at reasonable costs for development of radiation and isotopic power applications. However, considerably more research and development will be required before multimegacurie quantities of fission product radioisotopes can be economically produced on a routine production basis. Ultimately, it may be possible to produce strontium 90 for as little as 30 cents per curie, cesium 137 for less than 20 cents per curie, and promethium 147 and cerium 144 for only a few cents per curie. Final fabrication of the radioisotope source, encapsulation and transportation costs would be, of course, additional expenses and depending upon the final form and specific design of the source.

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Conclusion

In the United States, the Atomic Energy Commission, because of its specialized interests in the fields of isotopes and radiation development and in fission product utilization, has undertaken to pioneer and develop radioisotopic power and isotope technology to the point where further commercial acceptance and application is feasible. The Commission has also accepted a public responsibility for providing radioisotopic materials and services to the extent that they are not reasonably available from sources of supply independent of the Commission. Private industry, has also responded to their public responsibilities through development and promotion of radioisotope applications.

The Commission is uniquely endowed by the availability of excellent facilities and personnel to contribute substantially toward the advancement of both radioisotope process and source development and utilization development programs. Private industry also has unique skills and technology not readily available within the Commission's complex. A blending of these human and capital resources via the Isotopes Development Program permits a strong, diversified effort toward the accelerated development of new applications and uses for radioisotopes. Thus, private industry and the Commission by combining their critical skills and resources in a program full of scientific adventure have produced some fascinating developments. In fact, the Isotopes Development Program is such a fascinating, dynamic adventure, so filled with exciting potential developments that it can never be anything short of a success. Radioisotopes are truly modern new Atomic Power Tools.

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FISSION PRODUCT HEAT & RADIATION SOURCES

Fission Product	Half Life	Radiation Energy β	Radiation Energy γ	Curies per Thermal Watt	Product Compound	Specific Activity	Practical Power Density
	(years)	(Mev)	(Mev)	(c/w)		(c/g)	(watts/cc)
Sr-90, Y-90	27.7	0.61 2.2	/1	159	$SrTiO_3$	40	1.24
Cs-137,Ba-137m	26.6	0.51 1.17	0.662	213	Cs Polyglass Cs Cl	12 27	0.23 0.50
Ce-144,Pr-144	0.78	0.17 0.30 2.98	0.08 0.134 0.70 1.5 2.2 Others	133	CeO_2	400	19.50
Pm-147	2.6	0.223	--	2500	Pm_2O_3	600	1.55

/1 Bremsstrahlung from Sr-90TiO_3 is equivalent to a one tenth value of 1.60 inches of lead. The observed one-tenth value of Cs-137 is 0.84 inches of lead.

TABLE III

Number and Types of Radioisotope Licensees /1
(or Groups Using Radioisotopes in U. S.)
(1946 - 1960 Calendar Years)

	1946	1947	1948	1949	1950	1951	1952	1953	1954	1955	1956	[illegible]	[illegible]	195
Medical institutions and physicians	38	86	117	144	206	307	445	651	870	1,149	1,533	[illegible]	[illegible]	2,28
Colleges and universities	22	48	69	89	123	154	178	198	212	225	248	[illegible]	[illegible]	28
Industrial Firms	18	35	56	103	206	371	583	793	1,020	1,246	1,493	[illegible]	[illegible]	1,62
Federal and State Laboratories	4	9	17	32	49	117	157	201	237	257	284	[illegible]	690	1,17
Foundations and Institutions	4	11	16	19	29	34	43	47	53	57	63	41	49	[illegible]
Others	0	0	0	0	1	3	5	15	24	34	44	34	69	[illegible]
Totals	86	189	275	387	614	986	1,411	1,905	2,416	2,968	3,665	3,392 /2	4,357	5,5

/1 Cumulative

/2 In November 1957 the AEC adjusted the number of licensees to include only those organizations having a currently valid licen years the number of new users had been added to old users to give a summary total. Therefore, the apparent number of organi isotopes dropped for the following seasons: (1) During nearly 12 years of radioisotope utilization, some organizations have radioisotopes; (2) numberous large institutions at multiple locations have now been issued a single basic license; (3) gen quantities of radioisotopes were increased. Some users now purchase radioisotopes under a general license and do not receiv thus, they are not included here.

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TABLE III

Number and Types of Radioisotope Licensees /1
(or Groups Using Radioisotopes in U. S.)
(1946 - 1960 Calendar Years)

	1946	1947	1948	1949	1950	1951	1952	1953	1954	1955	1956	1957 2/	1958	1959	1960	1961 Sept.
ıs	38	86	117	144	206	307	445	651	870	1,149	1,533	1,598	1,900	2,286	2,568	2,749
·sities	22	48	69	89	123	154	178	198	212	225	248	229	251	283	297	331
	18	35	56	103	206	371	583	763	1,020	1,246	1,493	1,221	1,398	1,623	1,817	1,935
	4	9	17	32	49	117	157	201	237	257	284	269	690	1,179	1,153	1,271
;titutions	4	11	16	19	29	34	43	47	53	57	63	41	49	52	55	58
	0	0	0	0	1	3	5	15	24	34	44	34	69	82	50	58
	86	189	275	387	614	986	1,411	1,905	2,416	2,968	3,665	3,392 2/	4,357	5,505	5,940	6,402

,57 the AEC adjusted the number of licensees to include only those organizations having a currently valid license. During previous
ɔer of new users had been added to old users to give a summary total. Therefore, the apparent number of organizations using radio-
ɔed for the following seasons: (1) During nearly 12 years of radioisotope utilization, some organizations have discontinued using
; (2) numberous large institutions at multiple locations have now been issued a single basic license; (3) generally licensed
radioisotopes were increased. Some users now purchase radioisotopes under a general license and do not receive a specific license;
e not included here.

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RADIOISOTOPES DISTRIBUTED BY OAK RIDGE NATIONAL LABORATORY **

(Includes Project Transfers) 1946-1961 Calendar Years (in curies)

.47	1948	1949	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959	1960	1961 Sept.	1961 Sept.
	1	3	4	9	6	6	5	6	9	10	10	16	24	35	144
	(*)	1	22	1	5	21	327	2,032	2,862	6,076	5,854	76,037	37,720	26,232	157,191
0	0	85	534	3,991	9,634	6,181	25,493	50,532	93,618	146,505	173,235	168,817	143,747	235,298	1,057,610
23	55	177	311	468	490	595	557	657	735	891	1,074	649	527	156	7,273
28	45	74	93	102	119	147	153	158	165	179	230	163	151	112	1,919
----	----	151	----	----	60	135	168	587	5,848	5,280	39,769	40,902	8,050	36,768	137,718
----	----	6	----	4	0	0	3	66	339	1,145	1,978	3,107	3,439	3,146	13,221
----	----	6	----	9	20	61	117	129	23	18	56	122	338	98,260	99,159
0	0	0	0	(*)	(*)	(*)	(*)	6	14	19	663	567	2,407	839	4,516
----	----	24	----	(*)	0	12	1,200	3,524	5,721	6,190	5,525	8,666	9,097	4,558	44,517
6	8	13	146	81	90	82	41	75	166	317	320	319	174	828	2,544
58	109	353	1,111	4,665	10,415	7,150	28,063	57,772	109,500	166,630	228,714	299,365	205,674	406,232	1,525,812

t include several thousand curies of radioisotopes produced and distributed directly from other AEC facilities as Mound khaven National Laboratory nor from reactors owned by private industrial firms and universities. Radioisotopes made in d imported into United States are also not included.

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RADIOISOTOPES DISTRIBUTED BY OAK RIDGE NATIONAL LABORATORY **

(Includes Project Transfers) 1946-1961 Calendar Years (in curies)

	1946-47	1948	1949	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959	1960
Carbon 14	(*)	1	3	4	9	6	6	5	6	9	10	10	16	24
Cesium 137	(*)	(*)	1	22	1	5	21	327	2,032	2,862	6,076	5,854	76,037	37,720
Cobalt 60	0	0	85	534	3,991	9,634	6,181	25,493	50,532	93,618	146,505	173,235	168,817	143,747
Iodine 131	23	55	177	311	468	490	505	557	657	735	891	1,074	649	527
Phosphorus 32	28	45	74	93	102	119	147	153	158	165	179	230	163	151
Hydrogen 3	------	------	151	------	------	60	135	168	587	5,848	5,280	39,769	40,902	8,050
Krypton 85	------	------	6	------	4	0	0	3	66	339	1,145	1,978	3,107	3,439
Strontium 90	------	------	6	------	9	20	61	117	129	23	18	56	122	338
Promethium 147	0	0	0	0	(*)	(*)	(*)	(*)	6	14	19	663	567	2,407
Iridium 192	------	------	24	------	(*)	0	12	1,200	3,524	5,721	6,190	5,525	8,666	9,097
All Other	6	8	13	146	81	80	82	41	75	166	317	320	319	174
Total	58	109	353	1,111	4,665	10,415	7,150	28,063	57,772	109,500	166,630	228,714	299,365	205,674

* Less than 1 curie

** This table does not include several thousand curies of radioisotopes produced and distributed directly from other AEC facilities Laboratory or Brookhaven National Laboratory nor from reactors owned by private industrial firms and universities. Radioisotopes other countries and imported into United States are also not included.

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TABLE II

OAK RIDGE NATIONAL LABORATORY RADIOISOTOPE SALES /1
(calendar years)

Thousands of Dollars

6-48	1949	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959	1960	1946-60
35	156	293	307	760	863	1040	1325	1751	1873	1549	1432	1369	12,902
28	31	17	34	17	51	53	76	112	225	468	378	258	1,751
13	187	310	341	777	914	1093	1401	1863	2099	2017	1910	1627	14,653
45	535	509	7465	560	419	502	280	347	500	421	512	478	6,162
58	722	819	1086	1345	1333	1595	1681	2210	2599	2438	2322	2105	20,815

es not include sales from other AEC facilities or sales by commercial firms.

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TABLE II

OAK RIDGE NATIONAL LABORATORY RADIOISOTOPE SALES /1
(calendar years)

Thousands of Dollars

Customers	1946-48	1949	1950	1951	1952	1953	1954	1955	1956	1957	1958	1959	1
Domestic	185	156	293	307	760	863	1040	1325	1751	1873	1549	1432	1
Foreign	28	31	17	34	17	51	53	76	112	225	468	378	2
Total Commercial	213	187	310	341	777	914	1093	1401	1863	2099	2017	1910	1
Within AEC	345	535	509	745	568	419	502	280	347	500	421	512	1
Total	558	722	819	1086	1345	1333	1595	1681	2210	2599	2438	2322	2

/1 This table does not include sales from other AEC facilities or sales by commercial firms.

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TABLE IV

RADIOISOTOPE EXPORTS AND IMPORTS
(thousands of dollars)

Calendar Year	Exports	Imports
1952	$ 239	$ 86
1953	522	170
1954	536	150
1955	1288	189
1956	806	514
1957	1367	823
1958	1534	908
1959	1283	1145
1960	1286	1394

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TABLE V

MAJOR 1960 EXPORTS AND IMPORTS
(thousands of dollars)

	Exported to	Imported From
Canada	$ 132	$1,054
United Kingdom	224	205
Belgium	122	107
France	112	21
Japan	140	--
West Germany	61	--
Argentina	57	--
Switzerland	54	--
Italy	47	--
Others	337	7
Total	$1,286	$1,394

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TABLE VI

AEC RADIOISOTOPE PRICES /1

Radioisotope	1951	1961
Carbon 14	$36.00/mc	$9.50/mc.
Cesium 137	12,000/c	1.00/c
Cobalt 60 (<30 c/g)	1,650/c	1.00/c
Tritium	100/c	2.00/c
Iodine 131	0.75/mc	0.30/mc

/1 In large quantities, i.e., lowest price per unit.

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