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AEC RESEARCH AND DEVELOPMENT REPORT

QUARTERLY PROGRESS REPORT

RESEARCH AND DEVELOPMENT ACTIVITIES IN THE FIELD OF RADIOLOGICAL SCIENCES

APRIL - JUNE, 1960

AUGUST 5, 1960

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UC-41, Health and Safety
(TID-4500, 15th Ed.)
C.36

QUARTERLY PROGRESS REPORT
RESEARCH AND DEVELOPMENT ACTIVITIES
IN THE FIELD OF RADIOLOGICAL SCIENCES
APRIL - JUNE, 1960

By
Members of the Staff
of the
Hanford Laboratories Operation

August 5, 1960

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1) - 1350 between
the Atomic Energy Commission and General Electric Company

Printed by/for the U. S. Atomic Energy Commission

Printed in USA. Price $1.00 Available from the
Office of Technical Services
U. S. Department of Commerce
Washington, D. C.

UNCLASSIFIED

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IN THE FIELD OF RADIOLOGICAL SCIENCES
APRIL - JUNE, 1960

I. INTRODUCTION

This report is the 43rd and last in this series* presenting the research and development work in the field of Radiological Sciences. Some items charged to control and programs other than those of the Division of Biology and Medicine are included for general interest. Such programs are identified in the headings.

The previous report in this series was numbered HW-64945, dated April 27, 1960.

II. PERSONNEL

	Biology Operation			Chemical Research and Development Operation			Physics and Instrument Resea and Development Operation		
	3-31 -60	6-30 -60	Net Increase	3-31 -60	6-30 -60	Net Increase	3-31 -60	6-30 -60	Net Incr
Supervisors	8	8	0	2	2	0	3	3	
Scientific and Technical	44	49	+5	14	11	-3	12	17	+
Non-Technical	15	12	-3	7	6	-1	7	8	-
Clerical	8	8	0	1	1	0	2	2	
Total	75	77	+2	24	20	-4	24	30	+

* This report will be superseded by a monthly summary and an annual re

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III. LABORATORY FACILITIES

The scoping for the Biology Facility improvements was completed and has been forwarded to the AEC. A revised project proposal incorporating a $40,000 addition is currently under preparation.

The walls, floor and roof slab of the shielded animal monitoring station have been poured and the cylindrical shell plate is in place.

IV. RESEARCH AND DEVELOPMENT PROGRESS - BIOLOGY

(H. A. Kornberg, Manager)

A. Aquatic Biology (R. F. Foster)

Effect of Reactor Effluent on Aquatic Organisms

(P. A. Olson)

(Not charged to research)

Migrant size chinook salmon fingerlings were exposed to effluent from one reactor at three different concentration levels: (1) a concentration typical of those which exist in the Columbia River at this time of year, (2) a slightly higher concentration , and (3) a concentration twice that predicted for the future. This test was designed to simulate the conditions which might be encountered in this section of the Columbia River by young salmon originating upstream from the Hanford Plant during their migration to the ocean. In each case the concentration of the effluent was adjusted to compensate for changes in river flow typical of this period of the year. The fish were exposed to the effluent for 12 weeks which is considerably longer than the time ordinarily required for their passage through the region. Final results are summarized in Table I.

No adverse effect on mortality or growth was evident except in the lot exposed to "twice future concentration" effluent. In this case the results were biased by "gas-bubble" disease produced by a supersaturation of gases in the experimental water which resulted from the higher water temperature associated with addition of the effluent.

TABLE I

EFFECT OF REACTOR EFFLUENT ON MIGRANT-SIZE CHINOOK SALMON AFTER 12 WEEKS' EXPOSURE

Concentration of Effluent	Per Cent Mortality	Avg. Weight (g)
Control	5.3	15
Control	2.7	17
Present	2.7	17
Present	4.0	18
Future	2.7	18
Future	5.3	16
Twice Future	8.0	15
Twice Future	6.7	13

Effects of Radioactive Elements on Aquatic Life

(R. E. Nakatani, P. A. Olson)

Oral administration of Sr^{90}-Y^{90} to trout was continued through the 17th week. Duplicate lots of 10 fish each are being force fed five days a week at levels of 5×10^{-3}, 5×10^{-2}, and 5×10^{-1} $\mu c/g$ of body weight. Average mortality and growth data at the end of the quarter are shown in Table II.

TABLE II

EFFECT OF ORALLY ADMINISTERED Sr^{90}-Y^{90} ON TROUT

$\mu c/g$ of Fish/Day		Initial Avg. Wt. (g)	Avg. Wt. (g) at 17 Weeks	Per Cent Mortality
0	A	144	346	20
(Control)	B	133	369	0
5×10^{-3}	A	133	328	20
(Low)	B	131	381	50
5×10^{-2}	A	139	322	20
(Medium)	B	129	379	20
5×10^{-1}	A	137	293	50
(High)	B	132	313	40

Survival of the control fish is slightly better than that of the fish receiving the isotope but a clear dose-effect relationship is not yet established. Growth curves strongly indicate radiation damage in the high level group although the difference is not yet statistically significant.

Exposure of cichlids to water which contained 1×10^{-4}, 5×10^{-4}, or 2×10^{-3} μc P^{32}/ml was discontinued after approximately eight months. The test was designed to determine the body burden of P^{32} which affected spawning and egg fertility. Consistent deposition of the isotope in the fish was not achieved, however, and spawning was erratic. Cursory observations did not reveal radiation damage in the exposed fish, some of which contained more than 0.1 μc P^{32}/g.

B. Experimental Animal Farm (L. K. Bustad)

Biological Effects of I^{131}

(V. G. Horstman, R. L. Persing, P. L. Hackett, S. Marks and L. K. Bustad)

The relative thyroidal uptake of I^{131} by lambs from *in vivo* or *in vitro* I^{131}-labeled milk was found to be approximately the same in a preliminary study. Further investigation is being made.

Strontium Metabolism and Toxicity

(W. J. Clarke, J. R. McKenney, R. O. McClellan, P. L. Hackett, V. G. Horstman and L. K. Bustad)

The blood picture of the adult miniature swine fed 1, 5, and 25 μc/day of Sr^{90} for over one year and that of their three-month-old offspring is indistinguishable from the blood picture of the control animals. There is, however, a continued suggestion of damage in the offspring of swine fed 25 μc/day, as indicated by weight and general appearance. It is estimated that these swine received an average skeletal dose of 80-100 rads during fetal life and the suckling period. The radiation dosage to tissues other than those adjacent to bone may be considered negligible since the concentration of the Sr^{90} in soft tissues was less than 0.5 per cent of that in bone.

The concentrations of Sr^{90} in the skeletons of fetuses from sows fed daily amounts of Sr^{90} are nearly comparable to those in newborn swine. Weaned pigs (six weeks old) contain approximately 2.5 times as much Sr^{90} per gram of skeletal ash as the newborn. On the basis of only three of the offspring, it appears that the concentration of Sr^{90} per gram of ash in the teeth is indicative of the average concentration in the skeletons.

Comparative Toxicity of Sr^{90}, Ra^{226} and Pu^{239}

(W. J. Clarke, J. R. McKenney, R. O. McClellan
P. L. Hackett, V. G. Horstman and L. K. Bustad)

Miniature swine were given intravenous injections of Sr^{90} (64 $\mu c/kg$), Ra^{226} (6.4 $\mu c/kg$) and Pu^{239} (1.3 $\mu c/kg$). Doses chosen were estimated to cause similar biological effects. There was a suggestion of an initial depression in the neutrophilic components of the blood in all age groups (six weeks, six months, one year). There was also some evidence of bone pathology in the X-ray plates of the swine. Because of the small number of animals in each age group, however, proper evaluation of any radiation damage will require observation over a longer period of time.

Plutonium in Skin

(R. L. Persing, V. G. Horstman and L. K. Bustad)

Twenty-four intradermal injections of plutonium nitrate (0.2 N) in doses varying from 0.0016 to 5 μc per site were made in a white miniature pig. The intensity of the response was related to dose with erythema, swelling and foci of necrosis being evident within twenty-four hours. Scabbing occurred within a week. Control areas injected with 0.2 N nitric acid showed a similar response to that of sites injected with the lower level plutonium nitrate.

Tissue Transplantation and Radiation Therapy

(W. J. Clarke)

Six miniature swine exposed to 950-1000 r whole-body X-irradiation four of which received 11-day fetal hematopoietic cells at 72 hours after exposure, died within two weeks.

One animal, given 550 r whole-body X-irradiation and a transfusion ıdult marrow two years ago, was sacrificed. Histologic evidence of nage was restricted to a diffuse fibrosis of the spleen and lymph nodes. sticular tissue and bone marrow appeared normal.

Metabolism (R. C. Thompson)

Plutonium Absorption and Metabolism
(J. E. Ballou)

Complete data are now available on three pigs which were injected th plutonium followed by treatment with a single nine gram dose of DTPA. ıe treated pigs were sacrificed after six days and retained less than 0.5 r cent of the administered dose in liver and less than 6 per cent of the iminister dose in bone. This compares with the retention of 14 per cent liver and 56 per cent in bone of untreated control pigs. The treated animals creted 88 per cent of the administered dose during a six-day period fol- wing treatment, as compared with an excretion of less than 3 per cent by e control animals during the same period.

Studies on the lethal effects of combined plutonium administration us X-irradiation were continued with groups of rats being administered 2 μc Pu/kg followed at intervals of 30 minutes, 14 days, or 28 days, by 00 r X-irradiation. Preliminary indications of survival following the -ray dose suggests that there is little effect attributable to the delay period. This is in contrast to earlier results obtained at lower plutonium and X-ray evels.

Preliminary results have been obtained from experiments studying he effect of EDTA applied to plutonium contaminated wounds. Results are presently available from only six animals and there is considerable variation between animals. There is a consistent indication, however, that the decon- amination of wounds with a 10 per cent EDTA - "Tide" solution, results in an enhanced initial deposition of plutonium in liver and bone by perhaps a factor of two as compared to the case of water-washed wounds. Excretion of

plutonium in the urine was enhanced by a factor of five during the two days following treatment. The EDTA "Tide" treatment appeared to be only slightly more effective than water in effecting decontamination of the wound. These conclusions are based upon very limited data and additional experiments are in progress to more clearly define the situation.

Fission Product Absorption and Metabolism

(R. F. Palmer and R. C. Thompson)

Preliminary results are available from the experiment studying the effect of simultaneous variation of phosphorus and calcium levels in the diet on the distribution and retention of orally administered Ca^{45} and Sr^{90}. Data thus far obtained indicate a considerably more complex picture than has been described by others. Final conclusions must await the completion of additional analyses.

Gastrointestinal Radiation Injury

(M. F. Sullivan)

A number of rats which survived various types of irradiation of the intestine have been followed for the appearance of intestinal tumors. Tumors of the small intestine developed in 5 of 18 animals whose exteriorized intestine was exposed to doses ranging from 1500 to 1900 r. Tumors of the small intestine developed in 2 of 12 animals whose intestine was exposed *in situ* to radiation in this same dosage range. None of 25 control animals or of 21 animals exposed in the dosage range 900-1300 r developed intestinal tumors. No tumors of the large intestine were observed in any of the animals.

A number of procedures have been studied in an attempt to influence the increased fecal excretion of polyvinylpyrolidone (PVP) which follows irradiation of the intestine. Shielding segments of the intestine had no effect on the radiation induced increase. Administration of drugs known to decrease capillary fragility and permeability had no effect. Ligation of the bile duct decreased PVP loss following a 1000 r X-ray by a factor of two or more. The

data suggest that a major portion of the fecal flow in radiation diarrhea is composed of bile and that this fecal flow enhances the loss of protein from the irradiated intestine. Studies are continuing in an attempt to determine the mechanism of these effects.

D. Pharmacology Operation (W. J. Bair)

Deposition and Pharmacodynamics of Radioactive Particles
(W. J. Bair, R. A. Hennacy, D. H. Willard, and H. E. Dziuk)

In preliminary experiments with rats exposed via intubation to non-radioactive aerosols it appeared that the amount deposited in lungs is proportional to the positive pressure of the air containing the particles during filling of the lungs. These studies are being extended to radioactive aerosols to study various factors influencing deposition of particles in the respiratory tract.

In fourteen dogs killed immediately after exposure to plutonium dioxide, the mean content of the lungs was 60 per cent of the total plutonium deposited. The median was 58 per cent. Two weeks after exposure twelve other dogs were killed. The mean content of lungs was 35 per cent of the total plutonium deposited, and the median was 29 per cent. In earlier experiments the mean plutonium content of lungs in mice was 33 per cent of the total deposited in those killed immediately after exposure and four per cent in one group of 15 mice, and 10 per cent in another group killed about one week after exposure. This marked discrepancy in the deposition and retention of plutonium in dogs and mice emphasizes the need for comparative studies on several species before extrapolation to man.

Toxicity of Radioactive Particles
(D. H. Willard, J. E. West)

To date 30 beagle dogs have died following inhalation of plutonium dioxide. The time of death was never less than two months after exposure even when the initial lung burden exceeded 200 μc. Assuming uniform distribution of plutonium in lung tissue the mean total dose to lung of the

dogs which died was about 20,000 rad. Although death was preceded by a period of accelerated respiratory rate, decreased numbers of blood lymphocytes and weight loss, there was no evidence of pain or discomfort.

E. Plant Nutrition and Microbiology (F. P. Hungate)

Microbiological Studies

(R. T. O'Brien)

X-irradiation renders yeast cells deficient in catalase more sensitive to H_2O_2 than if not irradiated. Presumably this increased sensitivity results from greater penetration of H_2O_2 into irradiated than into unirradiated cells.

Heavily irradiated yeast cells bind less mercury to their cell surface than do unirradiated cells. Furthermore the mercury bound to irradiated cells is more easily lost to a suspending medium than is mercury bound to unirradiated cells. The data are consistent with a concept of SH group destruction by both radiation and mercury.

Columbia River Contamination

(M. P. Fujihara and P. A. Olson - Aquatic Biology)

(Not charged to research)

The degree of resistance to lethal effects of C. columnaris appears to be dependent on fish strain. Local Columbia River stock is killed off more readily than fish obtained from the White Salmon hatchery and the most resistant salmon so far tested were from the University of Washington. It is possible that differences in sensitivity may be due in part to size differences between the fish tested from the various hatcheries. Increased age and size cause increased resistance to columnaris in both salmon and trout.

Uptake of Radioactive Substances by Plants

(R. L. Uhler, J. F. Cline and J. D. Stewart)

Rate studies with bean plants have shown that individual plants differ significantly in their uptake of Sr^{85} and Ca^{45}. This means that previous data

using groups of plants may have failed to identify minor differences in ion uptake. In some plants it appears that there is a distinctly preferential uptake of Sr^{85} and in others studied at the same time, it appears that Ca^{45} is preferred. There is at present no data suggesting why individual plants behave differently and no psyche is implied for plants.

With extremely low concentrations of calcium in the nutrient substrate, addition of strontium causes increased chlorophyll content in beans. Apparently strontium is partially satisfying a calcium requirement of the plant.

When the sum of potassium and cesium ions were held constant, but the proportion of the two varied, there was only a modest effect on the discrimination factor (DF). The change in DF in roots was opposite that in other plant parts suggesting that ion selection occurred during penetration of the ions into and through the roots. Cesium toxicity was noted in leaves when the cesium concentration in the substrate exceeded the potassium.

Experiments relating soil moisture to ion uptake by plants showed that Cs^{137} was more readily taken up by plants when soil moisture was at a minimum. With low moisture the DF for Cs^{137} was clearly higher than at higher moisture levels. In these experiments carrier cesium was added to the soil along with the Cs^{137} and potassium.

F. Radioecology (J. J. Davis)

Project Chariot

(J. J. Davis, W. C. Hanson, D. G. Watson)

Samples of plants and animals collected at the site of Project Chariot, Cape Thompson, Alaska, during 1959, were found to have pronounced differences among species in content of radioelements from worldwide fallout. Highest levels among plants were in lichens, sphagnum moss and sedge and lowest levels in emergent plants from ponds. Among birds, highest levels occurred in Lapland longspurs which feed upon insects and seeds; lowest

-urred in piscivorous marine birds. Field work for limnology, terrestrial invertebrates ecology and distribution elements in the biota was started at the test site during May.

Effects of Radioactivity on Population

(H. E. Erdman)

The longevity of Habrobracon wasps was reduced by being maintained on food containing 0.1 μc of Pu^{239}/ ml of media. Plutonium occurred in eggs from these insects.

Population Dynamics

(W. C. Hanson)

Two hundred fifty-five nests of Canada geese were found on river islands within the Hanford reservation.

Biological Monitoring

(D. G. Watson, W. C. Hanson)

Concentrations of I^{131} in jack rabbit thyroids decreased slightly during the quarter. The average was about 29 per cent of the previous quarter average and equal to that of one year ago. Amounts of total beta emitters in rabbit tissues also were comparable to one year ago.

Concentrations of beta emitters in small fish at Hanford increased during the quarter. At the end of the quarter they were about six times higher than for the same period of the previous year.

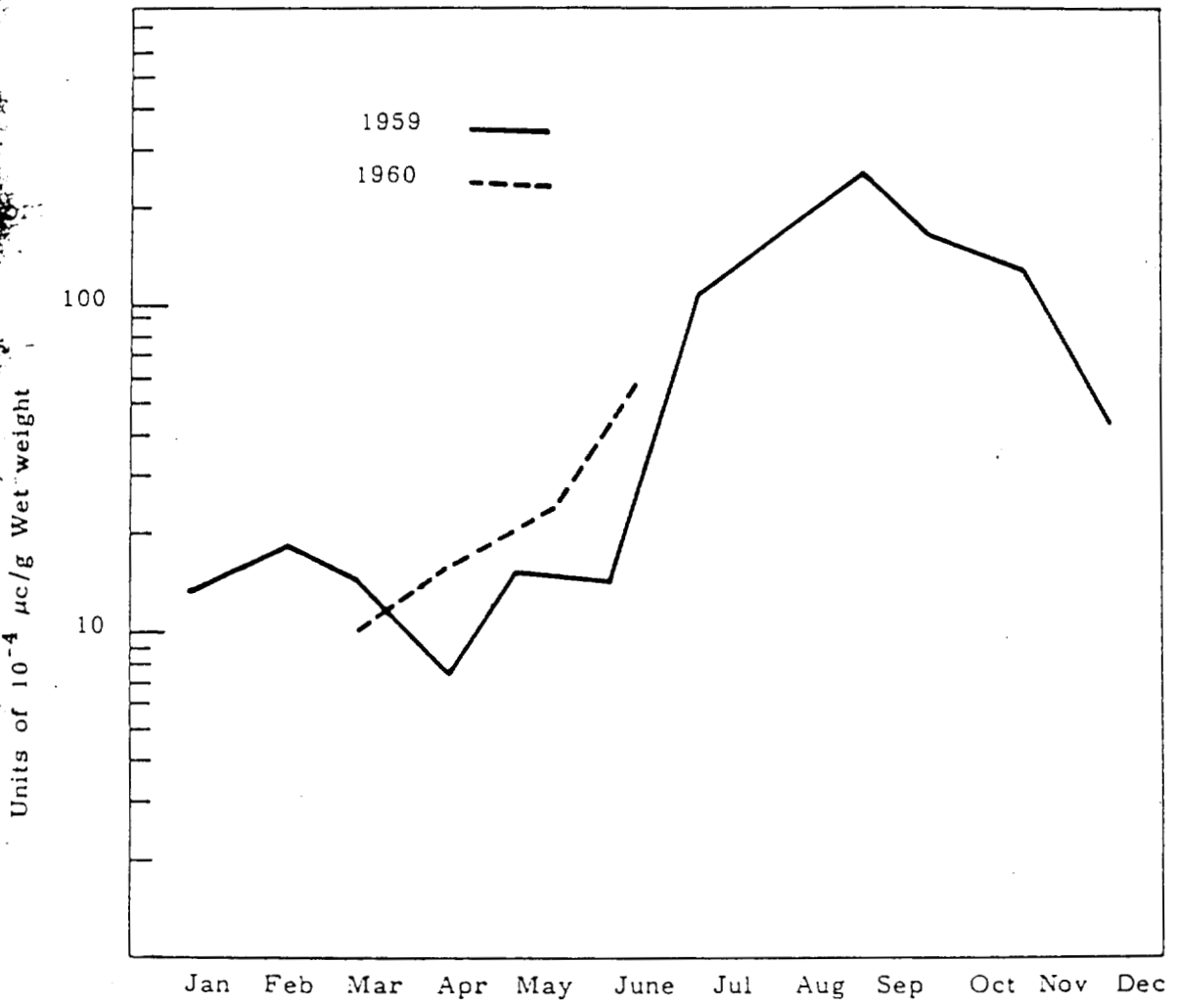

FIGURE 1

Concentration of Beta Emitters in Small Fish at Hanford

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V. RESEARCH AND DEVELOPMENT PROGRESS - PHYSICS AND INSTRUMENTATION (P. F. Gast, Manager)

A. Meteorology (J. J. Fuquay)

Diffusion and Transport Studies

(J. J. Fuquay, C. L. Simpson, P. W. Nickola, M. F. Scoggins, R. J. Engelmann, C. E. Elderkin)

The major research and development effort within the Atmospheric Physics Operation was devoted to development and calibration of assaying techniques for determining the mass of ZnS tracer material on membrane filters. Intensive effort in this direction was prompted by two major difficulties encountered during the summer 1959 transport and dispersion experiments; namely: (1) prolonged high volume sampling at 8 and 16 miles distance from the release point caused an undesirable collection of natural air-borne dusts, which tended to obscure the fluorescent tracer particles, and (2) the calibration of the automatic counters by visual assay of exposed filters irradiated with ultraviolet light exceeded the imposed uncertainty limits. The uncertainty in the visual counting was due largely to complexities introduced by the geometry of the filter backing material. However disconcerting these difficulties have been, in the final analysis, it may have proved to be a rather fortunate circumstance, for it motivated experimentation with data reduction techniques that could distinguish between the dust and the tracer particles. Further data analysis was deferred until the calibration and dust problems could be adequately resolved.

When the filters are relatively free of impurities, they are assayed with the "Rankin" counters. In this equipment, the zinc sulfide particles are activated on the surface of the filter by a fixed-strength source of plutonium alpha particles. The resulting scintillations are counted by means of a photomultiplier and associated amplifying and counting equipment. The observed counting rate is almost directly proportional to the mass of zinc sulfide on the filter.

For dusty filters and for calibrating the "Rankin" counters according to mass, a new method developed by the Radiological Chemistry group is used. The membrane filters are placed in a small vial and dissolved. The sample is then exposed to a fluorescent light source, and the resulting phosphorescence is measured in a "Tri-Carb" automatic liquid scintillation counter. A correction factor for the effect of impurities in the sample is determined from a turbidity measurement with a colorimeter.

In relating Tri-Carb counts to mass of pigment, known amounts of pigment were suspended in sampling vials and several repeat counts were extracted and the counting repeated. The residual variances were then determined by using a stratified analysis of variance. The results were that the errors introduced in making the sample were of the approximate magnitude of the counting error inherent in the Tri-Carb counter. A mass determination of a single sample could thus be in error 20-25 per cent at the 2σ limit. Best estimates of the machine counts were obtained for each mass level by employing a non-orthogonal analysis of variance and a quadratic relation between counts and mass obtained by standard weighting procedures.

The effects of dust were examined, using paired readings of Tri-Carb counts of sample filters and those of the same filters after known amounts of dust had been added to the solutions. The dust was measured by a colorimeter which was capable of reproducing readings within one per cent. From these data, regression yielded a linear relation between the ratio of the true mass to the apparent mass ($\frac{M}{M*}$) and the dust loading as measured by the colorimeter reading K. In analysis, this ratio is the factor that is multiplied by the mass obtained from a dusty filter to obtain the mass with the dust removed.

The Rankin-mass relationships were similarly determined through regression with paired readings of Rankin counts and mass determinations of Tri-Carb counts. Because the counting variability of the Rankin equipment is very small, greater confidence can be placed in the resulting mass determination.

Mass and exposure determinations for all samples of the Green Glow program can now be made. The dust loading problem encountered at the remote distances has been solved.

B. Radiation Dosimetry (W. C. Roesch)

Gamma-Ray Dosimetry

(W. C. Roesch, I. T. Myers)

A new control system was put on the water bath of the gamma-ray calorimeter. It operates like damping in a mechanical system. Without it there was a 0.001 C overshoot in the bath; now the temperature cycle has a maximum range of 0.0002 C. The effect of this improved control on the calorimeter is being evaluated.

The Manager, Radiological Physics spent two weeks at Geneva, Switzerland attending meetings of the Ad Hoc Committee on Quantities and Units of the International Commission on Radiological Units and Measurement (ICRU). The committee prepared a report containing new and/or improved definitions of the concepts, quantities, and units that are fundamental to radiology, radiobiology, radiation protection, and etc. The ICRU is now examining the report. They will distribute it when they have approved it.

The seventh year of teaching Radiology-485 at the University of Washington was completed. An ad hoc committee established at the University (and of which the Manager, Radiological Physics was a member) completed its study of their Health Physics training program. A Radiological Sciences Council was established as part of the graduate school to supervise an MS degree program in this field. The Manager, Radiological Physics, was asked to serve on this council. A graduate student of the

University was hired as a temporary employee to work on an experimental problem of mutual interest. He will use the results of his work for his MS thesis.

Neutron Dosimetry

(J. De Pangher, R. D. McCormick, H. V. Larson)

As a result of the realignment reported last quarter the positive ion Van de Graaff is now operating better than it ever has before. The alignment methods that were developed permit definition of the direction of the undeflected beam to within 0.01 degree. It appears probable that shifts in the foundation of the building will prevent keeping the beam aligned within these limits. The ion source of the positive ion Van de Graaff wore out and had to be replaced. During replacement of the source, the heaters of the palladium leaks were altered to permit better source control. The warning light system was also revised slightly. A check of the alignment of the system showed it had shifted only 0.01 degree during this maintenance work; the shift in the energy calibration of the proton-moment detector was only about 0.015 per cent. Development of a helium ion source for the Van de Graaff continued with emphasis on measurement of the parameters of interest. Another antimony-beryllium neutron source was activated. Since no contamination was found on the source it is available for experiments. A commercially made BF_3 tube of high quality was finally developed for use in the double moderator.

Three precision long counters made independently by three different shops were available for intercomparison. One discrepancy that appeared was traced to different neutron absorption in geometrically similar metal tubes in the different counters. Presumably the metals differ slightly in composition or contain traces of different neutron-absorbing impurities. Aluminum was substituted for brass in these parts of the counters. The variation in counter response was reduced from 3 per cent to 0.1 per cent.

New experimental data were obtained on a fairly large PoBe source, a small PuBe source and an intermediate PuBe source. Any neutron energy degradation in the latter source was below the limit of detectability. A search was made for 7.65 Mev gamma radiation from these sources. Theory indicated that one of the possible alpha-neutron reactions in the sources would lead to a state from which such radiation might be emitted but that it should not be found; the search showed no such radiation. The information acquired recently concerning the energy spectra and the energy degradation in neutron sources was incorporated in a paper to be presented at a meeting of the American Physical Society.

A Perlow spectrometer for neutron energy measurements was put into operation and calibrations begun. A resolution of eight per cent for 886 kev neutrons was achieved. This is as good as has been achieved with such a device. A study of the B^{10} reaction was begun with a spectrometer.

Preliminary tests of normal and enriched LiI scintillation crystals were carried out to indicate their suitability for neutron spectrometry.

A technique was developed for making tritium-titanium targets for the Van de Graaff on tantalum backing material. Tantalum is harder and cheaper than platinum and does not require inventory.

The first attempts at fabricating tissue-equivalent chambers for neutron measurements were made.

Body Monitor

(W. C. Roesch, H. E. Palmer)

A new, more substantial chair was fabricated for the counter. There was difficulty in finding a padding material which was not radioactive. A removable bed was made for the iron room and a screw drive was installed on the holder of the counter. These will permit studies aimed at a scanning method for routine counting.

A series of experiments is in progress to determine how much radioactivity accumulates in the hair and thus how important it is that a shampoo be taken before the whole body count. One female subject was counted both before and after shampooing on eight separate occasions during the past two months. In every case shampooing caused a 100 to 200 per cent increase in the radioactivity in the hair. The increase was found to be due to the radioactivity in the water used. Distilled water does not show the radioactivity that is present in the regular water at the whole body counter. When distilled water was used by the above subject for shampoos, the radioactivity in her hair was decreased below that before the shampoo. Other experiments are being made with a wig of natural human hair. After three days outside in rather stormy weather, the decay products of radium could easily be identified in the wig. When the wig was placed at the position of the head in the standard whole body count, it increased the K^{40} values about 4 per cent, the Cs^{137} values about 8 per cent, and would produce very significant errors in the Zn^{65} values when these are low. An exposure of the wig during milder weather produced even more significant differences but other exposures showed smaller effects.

Two occasions arose that permitted comparisons of the background of the Hanford whole body counter with those of other very similar counters. During a trip to Los Alamos a tape of their background spectrum was obtained. Their crystal is two inches (50 per cent) thicker than ours; this is the only significant difference. Their background is slightly higher due to the larger counter and to their higher elevation with its consequent greater cosmic ray effects. There is also some evidence for slightly higher background from the daughters of radium. The new counter for the animal counting facility of the Biology Operation was also compared with the Hanford whole body counter. This counter is slightly smaller in diameter than ours, and it has only four photomultipliers compared with our seven. Its background is smaller by 53 per cent under the K^{40} photopeak and 29 and 27 per cent under the Zn^{65} and Cs^{137} photopeaks. Tests by Radiological Chemistry

with the Biology counter showed that the background in our iron room is not significantly different from that obtained in a good lead shield. It is of considerable interest that the new Biology counter shows a peak shift phenomenon that is similar to ours: if a source is placed on the photomultiplier side of the counter, its photopeak will appear a couple of channels lower than if it is counted on the other side. Private communications have indicated that this is observed in other large counters also.

The cooperative experiment with Dr. C. L. Finch of the University of Washington continued. Two more people were given intravenous injections of Fe^{59} and counted at the whole body counter.

Changes in the efficiency of the whole body counter multichannel analyzer in its low numbered channels were identified as the source of many of the variations in background that have been observed. When data from the low numbered channels were disregarded, background variations were reduced and were brought into agreement with what might be expected from changes in atmospheric radon due to changes in weather conditions.

The first IBM run of data collected routinely at the whole body counter was completed. This operation will be on schedule with the completion of the June processing of data.

Further tests with the shadow shield counter indicate that the radiation scattered from a person into the counter can be almost completely eliminated by surrounding the subject and counter with sheets of lead 1/4-inch thick. A mockup of a full size shadow shield counter for counting people was built. The arrangement [illegible] as though the roof of the present iron room was used as the shadow shield. The background of this counter was indistinguishable from that of the counter in the iron room at the high energies. At the lowest energies we now use it was only 12 per cent higher.

An attempt was made to detect uranium in a subject having a high and constant bio-assay count; nothing could be detected. An attempt was

made to detect Pu^{239} in a subject suspected of having a high body burden; nothing was detected other than that in the wound (before its excision) that had presumably lead to the burden.

A subject who had inhaled radioactive material while working on the rear face of a reactor was found to contain 0.09 μc of Zn^{65}, 0.01 μc of Sc^{46}, and 0.23 μc of Cr^{51}. Twenty-four days later he contained 0.06, 0.005, and 0.11 μc of the same isotopes. During the latter examination a scan of his body showed that there was a concentration of activity in the upper lung area.

Sodium^{24} was detected in four subjects who had been working on the removal of a ruptured slug. The amounts were between 1×10^{-3} and 4×10^{-3} μc. In another recent case a radiation monitor was found to contain 2.5×10^{-3} μc of Na^{24} after working on a rear face during a shutdown. These cases were discovered through the routine counting program.

Zinc^{65} data obtained in recent months seem to show that the average body burden of employees working in a reactor area increases with the number of reactors upstream from that area.

Three items were received and are awaiting installation on the multichannel analyzer. These items are a paper punch that will make the data from the analyzer immediately available to the IBM machine, a magnetic tape device for spectrum analysis and a key punch for hand entry of data into the analyzer.

Following disagreement on the measurement of the plutonium in a sliver removed from a wound, two other slivers were measured by both X-ray and alpha particle counting. The alpha particle counting was preceded by dissolution of the slivers while the X-ray counts were made directly on the slivers. The alpha particle measurement indicated 6 to 8 times the quantity obtained by the X-ray counts. The difference is due to self absorption of the X rays in the slivers.

For plutonium in very shallow wounds both the 6 kev M X rays and the 17 kev L X rays can be observed. It was found that the ratio of the M to the L X rays can be used as a measurement of the depth of the source for depths to two mm. At this depth, correction for absorption of the L X rays is small.

An artificial dead-time circuit was built for use with plutonium X-ray counters. The object is to eliminate small afterpulses that follow very large pulses and are of just the height to be mistaken for X-ray pulses. Tests indicate that this method of discrimination is only partially successful. Attempts were also made to discriminate against these afterpulses on the basis of their much faster rise time. The space charge saturation method can be used for this purpose but only at voltages much higher than ordinarily used for X-ray counting.

C. Instrumentation (W. G. Spear)

Instrumentation

(W. G. Spear, D. P. Brown, R. A. Kent, M. O. Rankin
E. M. Sheen, J. L. Stringer, G. E. Driver)

The large NaI crystal for the Biology Operation Total Body Monitor was received and tested. The mounting assembly and scanning mechanism for the crystal was designed. Experiments started concerning a Ca^{45} monitor for use in uptake studies from a water solution to fish. A suitable detector device was designed for the work. The previously developed instrumentation for measuring respiratory rates, volumes, and body temperatures of small animals during exposure to radioactive aerosols was fabricated, tested, and is now in satisfactory operation. A development proposal was completed for Biology Operation for a special miniaturized instrument for multi-input scintillation detectors, associated selective gamma energy analysis channels, and necessary addenda amplifiers, scaler, and count-rate meter. The development proposal was for a completely transistorized portable-type unit. Experiments were conducted concerning a

monitor to be used for detection and measurement of Pu^{239} in the bloodstream of animals. Preliminary tests indicate the feasibility of such a system by measurement of the Pu^{239} X rays. Advice and consultation was rendered to Biology personnel concerning purchase of certain commercially obtainable instruments for use in their experiments.

Background investigations of gamma energy spectra were continued in several plant areas and buildings using the multichannel analyzer. All data were reduced and graphically plotted. Such data provide required information relative to background conditions and variations. This information is especially important for health hazard studies, shielding effects, and future instrument development work.

Two different types of personally-carried personnel alarming dosimeters were successfully developed and tested in bench-model experimental form during the quarter. One type utilizes a 10 cc ionization chamber pencil dosimeter as a detector plus a cadmium-sulfide cell and a sensitive relay. By charging the dosimeter correctly, an alarm buzzer is energized after the pre-selected dose is accumulated. A variation of this idea, now partly developed, employs a sliding selector to permit dial selection of the alarm dose level. The range of operation-selection is 10 mr to 200 mr with an accuracy of alarm to ± 5 mr. The alternate personnel dose integrator employs a miniature G-M tube, transistorized circuitry, and a modified watch which indicates the approximate integrated output of the G-M tube. This unit does not, of course, indicate the correct dose due to the use of a G-M tube detector, but it is certainly a useful device for many applications. Both units were tested satisfactorily with Ra^{226} and Co^{60} sources.

All circuitry and detector development-design work was completed on a scintillation, transistorized beta-gamma portable approximating dose-rate meter with a full-scale first range of either one mr/hr or five mr/hr as desired. The prototype instrument is now being fabricated for evaluation and field testing.

Standardization techniques for transistorized circuitry for use in line-operated radiation monitoring equipment continued. A number of amplifiers, binaries, multivibrators, oscillators, gates, and register drive circuits were completed in design-test work and were applied to several types of line-operated alpha and beta-gamma monitors. All circuitry is completely interchangeable and, in many cases, is also directly interchangeable with circuits employed in the family of portable radiation monitoring instruments developed over the past two years. Extension of this type of standardization of circuitry work will be carried out.

Experimental work was completed on the circuitry and detectors for a family of scintillation beta-gamma dose-rate area monitors. Several models of each type were fabricated and successfully tested. A number of production units were also fabricated for general plant use. The family of area monitors included a selectable range linear type with available ranges from 0 - 20 mr/hr full scale to 0 - 20 r/hr full scale; a combined two-detector, high-low, dual-range, selectable alarming unit; and several quasi-logarithmic units covering ranges of five mr/hr to five r/hr or higher as desired.

Development work continued on alpha and beta-gamma scintillation air monitoring systems. The alpha-only air monitor, using coincidence counting techniques to reduce radon-thoron background variation effects, was completed in experimental form except for the final readout circuits. Both analog and digital methods of readout are being investigated. The alpha air monitor sensitivity is calculated to permit alarming (with no radon-thoron false alarms) in three minutes if the detector is exposed for this time to an atmosphere with a continuous Pu^{239} concentration level of 2×10^{-10} $\mu c/cc$. The circuitry development work for the beta-gamma air monitor is complete, and the special lead-shielded detector is complete and has been tested satisfactorily. The calculated beta-gamma air monitor sensitivity will permit alarming, in 40 to 50 minutes, if the detector is exposed for this time to an atmosphere with a continuous mixed fission product contamination level of 10^{-9} $\mu c/cc$.

Investigative experiments continued during the quarter on the silicon surface barrier diode detectors useful for alpha particle detection, alpha energy analysis, neutron detection and etc. Three surface barrier diodes were fabricated with a cover of 50 micrograms/cm^2 of gold. All three worked well for gross alpha detection (Pu^{239}) with geometries exceeding 20 per cent. Two transistorized circuits were developed for work with the diodes and both performed satisfactorily. Extensive further work will be done with these diodes of which ten more prototype units are nearly completed. The diodes can be easily applied to many radiological instrument problems at HAPO.

Experiments continued concerning thermoluminescent dosimeters. These CaF_2: Mn dosimeters can eventually be extremely useful for, especially, finger ring, wrist, and other miniature-requirement dosimeter problems. The work accomplished included a method of inductively heating the detectors during readout; this method permits sealing of the thermoluminescent material against the atmosphere. Experiments included attempts at determination of the proper temperature to obtain the information relative to the accumulated dose of the detector, included a simplified packaging technique design to reduce assembly costs, and included work with a low-vacuum firing procedure to eliminate adverse oxidation effects on the dosimeter.

Experimental circuitry was developed and tested for use with the Total Body Monitor and other similar gamma energy spectrum analysis instruments. This developed circuitry enables the instrument operator to enter numerical data into the memory of a multichannel analyzer. The circuitry comprises a five-place manual keyboard, a three-place address register, and controls for memory address selection.

Consultation, advice, and design work were rendered to many plant groups concerning, principally, radiation detecting and measuring instruments and associated equipment. Specific work of this type included radiation damage effect investigations of glass and ceramic vacuum tubes,

transistors, and tunnel diodes; methods of detection of I^{131} in exhaust air streams in the presence of large quantities of A^{41}; methods of recording gamma energy spectrum data on magnetic tape; design of a number of special instruments for in-cell and cell filter monitoring; design of a special scintillation probe for the detection of Pu^{239}, via X-ray detection, in human wounds; and the design of a special high volume transistorized audio amplifier and loudspeaker instrument for use with certain radiation detection monitors. Another design improvement in both alpha and alpha-beta-gamma scintillation hand and shoe counter probes resulted in a net savings of 25 per cent of the original instruments purchase costs.

VI. RESEARCH AND DEVELOPMENT PROGRESS - CHEMISTRY AND SEPARATIONS PROCESSES (L. P. Bupp, Manager)

A. Radiological Chemistry (W. H. Reas and J. M. Nielsen)

Reactor Effluent Studies

(R. W. Perkins, W. B. Silker, C. W. Thomas)

Slow to fast neutron flux ratios were determined in test positions in the reactor graphite reflector in order to characterize these various fluxes for use in determining the reactions responsible for the formation of some of the reactor effluent radioisotopes, and hence to identify the parent isotopes. The $Al^{27}(n,\alpha)Na^{24}$, $Fe^{54}(n,p)Mn^{54}$, and $Fe^{58}(n,\gamma)Fe^{59}$ reactions were used in the study. The graphite reflector will serve effectively as a thermal column giving a slow to fast ratio of 200:1.

Scandium^{46} can be produced in the reactors by a fast neutron reaction on Ti^{46} or a slow neutron reaction on Sc^{45}. In order to determine which mechanism was responsible for the Sc^{46} observed in the aluminum process tubes, a series of samples of a tube were taken along its length so that a wide range of ratios of fast to slow neutrons flux would be represented. These samples were analyzed for Sc^{46}, Mn^{54} (which is formed by fast neutron reactions only) and Fe^{59} (which is formed by a slow neutron reaction). Scandium^{46} to Fe^{59} ratios are relatively constant while Sc^{46} to

Mn^{54} ratios vary markedly along the tube indicating that a slow neutron reaction on Sc^{45} is the source of the Sc^{46}.

In laboratory tests of the effectiveness of alum flocs in removing arsenic from river water it was observed that 98 per cent of ionic arsenic is incorporated into the floc at pH values from 6.0 to 7.2 and alum concentrations of 40 ppm. At pH values above and below this range the efficiency drops significantly. At 10 ppm alum only about 60 per cent of the arsenic is removed. Since 10 ppm is about the concentration now used in the process water treatment plants, an improvement in arsenic removal and a consequent decrease in As^{76} in reactor effluent water may be possible. Further small scale studies will be undertaken upon completion of a small prototype water treatment plant now under construction.

Two parts per million of EDTA were continuously added to the cooling water of a reactor process tube for three weeks to test the effect of a complexing agent on the process of formation of radioisotopes in the effluent water. Only minor fluctuations were observed in the concentrations of most radioisotopes except for Cu^{64} which rose rapidly to a value 15 times normal and then decreased rapidly to a value only about 25 per cent lower than normal. Studies are continuing to determine if practical beneficial reduction of radioactivity can be obtained as a result of the addition of this agent.

Analytical Methods

(C. W. Thomas, W. B. Silker)

Initial tests of electrodialysis as a means of concentrating large volume water samples showed that about 80 per cent of Sr^{85} from a 50 liter sample could be transferred through an ion exchange membrane into a cathode chamber of 1-2 liter capacity in five hours. The technique was successfully applied to eleven-gallon samples of reactor effluent water and Columbia River water to separate 90, 82 and 77 per cent of Na^{24}, Zn^{65} and Np^{239}, respectively. $Chromium^{51}$ was separated from the river water into the cation chamber, whereas in the case of reactor effluent water about equal

quantities were found in the cation and anion chambers. This substantiates other data which indicates that in reactor effluent water Cr^{51} is present in both anion and cation form. Only the cation form is present in the river water as a result of the reducing action of some material in the river.

In the zinc amalgam procedure for separating radiozinc from reactor effluent water reported last quarter the Cu^{64} which was also recovered was allowed to decay before the measurement of Zn^{65}. For those analyses where Zn^{69} and Zn^{69m} are to be determined, this procedure is not suitable. In this case a pre-extraction with cadmium amalgam was found to effectively reduce the Cu^{64} and allow counting of all three zinc radioisotopes.

Counting Methods

(R. W. Perkins)

A useful application of the Compton Cancelling Spectrometer was made in the determination of As^{76} in reactor effluent water. Arsenic-76 is commonly determined by beta absorption counting since the Cu^{64} annihilation radiation interferes with the gamma of As^{76}. In the Compton Cancelling Spectrometer the counting rate for Cu^{64} at 0.51 Mev is reduced a factor of 15 making possible direct measurement of As^{76} by gamma counting.

Sensitivity measurements were made of various sample sizes with different large sodium iodide detectors to help determine the optimum detector-geometry relationship for low-level environmental samples. The results are tabulated in Table III. The larger volume samples were placed in containers to surround the crystal (except for the phototube end) uniformly to the thickness given.

TABLE III

LARGE CRYSTAL SENSITIVITIES

Crystal	Sample Size	Cs^{137} Sensitivity (99 Per Cent) $\mu c/ml$
3" × 3"	3100 ml (2" thick)	1.3×10^{-8}
3" × 3"	6500 ml (3" thick)	0.96×10^{-8}
9" × 4"	8800 ml (2" thick)	0.41×10^{-8}
9-3/8" well crystal	500 ml bottle	2.1×10^{-8}

These data show the increased sensitivity available with large sample size. As would be expected, little increase in sensitivity is obtained by increasing the sample thickness from 2 to 3 inches.

Bio-assay Methods

(R. W. Perkins)

In a study of the effect of choice of carrier for the precipitation of plutonium in the bio-assay procedure it was observed that use of lanthanum resulted in a background of 17 tracks per week on the nuclear track film. Substitution of yttrium, holmium, samarium, dysprosium, or praseodymium resulted in 9, 1, 91, 2, and 1 tracks, respectively, indicating that for low background work holmium, dysprosium, or praesodymium would be better than lanthanum.

Chemical Protective Agents

(D. R. Kalkwarf)

Determinations of the equilibrium and rate constants for interconversions between the various forms of Erioglaucine present in aqueous solutions were completed for 25 C and 0.5 M ionic strength. It was concluded that Erioglaucine in solution is present in two colored forms and three colorless forms which differ in their extent of hydrolysis at the central carbon atom and protonation at the amino groups. With these data it is now

possible to determine which of the various forms of this dye react most rapidly with free radicals and to adjust conditions for measuring the protection indices of other compounds relative to this dye in non-neutral solutions.

B. Chemical Effluents Technology (D. W. Pearce)

Geological Studies

(R. E. Brown, D. J. Brown)

The basalt series beneath the Hanford Works was appraised in a preliminary way from the viewpoint of the potential feasibility of disposal of wastes at depth there. Procurement of logs of the Rattlesnake No. 1 test well, drilled adjacent to the Hanford Works by the Standard Oil Company of California, makes this appraisal more meaningful. The logs indicated that the well penetrated more than 100 basalt flows separated by interflow zones and tuff beds in the 10,655 feet drilled. The well did not penetrate through the basalt series. Current consideration of disposal at depth at Hanford must be predicted on disposal into the basalt, because information on rocks beneath the basalts here is completely lacking. The basalt flows encountered were evidently typical Columbia River basalts throughout the well. Tuff beds varied widely within the ranges noted in oth wells and from surface mapping. The number of tuff beds and per cent of in the series, particularly at depth, considerably exceeded earlier estima No flows or tuff beds appeared unique enough for use as stratigraphic mar Correlation with flows or tuff beds in other wells will be difficult owing to the present-day lack of knowledge of the geologic structures at depth, and extent, continuity, composition and variations in the tuff beds.

The high permeability of some zones within the basalt series was again confirmed by water level data north of the Hanford Works area. A rise in water level of more than 30 feet in three years was noted in a well penetrating more than 800 feet into the basalt series. The rise coincides with the rise in the unconfined water table in sites three miles north and s miles east. The transmitting aquifer or zone is yet unidentified.

A preliminary geologic map was completed of the eroded, buried surface of the Ringold formation beneath Hanford. This map, combined with structural studies of the Ringold formation, discloses areas where permeable beds of that formation lie at the water table. It thus defines the areas where wastes may enter those beds and the direction in which they might move to greater depth. It indicates where they may rise and either enter the Columbia River directly or move as Columbia River underflow before entering that stream.

X-ray diffraction examinations of geologic samples from outcrops and well construction operations were performed to assist with the identification and correlation of formations. Particular emphasis was placed on examination of samples from various basalt flows and basalt interbeds. One ash interbed that is exposed on the flank of Gable Mountain was traced to the vicinity of the 200 East area through correlation of X-ray diffraction data. Another layer of volcanic ash that occurs in the post-glacial sediments near the surface of the ground was correlated with an ash bed widely distributed throughout the Pacific Northwest. This latter extensive deposit was earlier shown by others to have originated from Glacier Peak in the Cascade Range, with an age of 6,700 years established by radiocarbon dating. This identification provides an important time reference in the chronology of geologic history of the Hanford area.

Soil Physics

(P. P. Rowe, R. W. Nelson, A. E. Reisenauer)

The question of adequately monitoring the water in an aquifer by sampling the contents of a perforated well was studied. Such monitoring wells are frequently perforated throughout the length beneath the water table and thus form low resistance flow paths for water movement between saturated zones having different potential heads. Some vertical variation in potential is usual, particularly in the vicinity of points of ground water recharge, such as cooling water ponds. This cross flow between aquifers

would prevent entry of water from the low-head zone, making it impossible to monitor the water in that zone by analyzing well samples. It is estimated that a head difference of only a few thousandths of an inch would prevent entry of water into a well from a low-head zone having typical Hanford hydrologic characteristics. Methods are being studied for identifyi aquifer zones that cannot be sampled by means of a completely penetrating, perforated well.

Perforated wells that penetrate zones of different potential head also do not provide means of accurately measuring potentials of specific points. By means of a numerical analysis it is possible to estimate the potentials of interior points if the potentials along the boundaries of the region are known As the values of potential cannot always be measured with fully perforated wells, a method was developed for numerically estimating the potentials along the bottom of a flow system. The method consists of solving numerically the partial differential equations that may be derived from the fact tha no water flow is possible across this boundary. The numerical solutions are obtained by using potential images across the boundary and applying the iteration process used for estimating interior points. Tested in a hypothetical example the method appeared to give consistent results.

Equations derived and tested which describe the two-dimensional flow patterns for an aquifer in the vicinity of a point of artificial recharge were extended to include the three-dimension case. The equations will help determine the ground water movement near the water mounds built up beneath Hanford seepage ponds. Approximate solutions to the partial differential equation were obtained by substituting three-dimensional differenc equations that could be solved numerically. Such solutions were obtained for hypothetical examples which tended to confirm the validity of the bounda assumptions and which were gratifyingly consistent with approximations pro vided by other methods.

A computer program for numerical evaluation of transient unsaturated flow in a heterogeneous medium was completed and successfully tested. The mathematical model is for drainage of a column of soil which is completely saturated at time zero. The test cases indicated rapid convergence of the iteration process and resulted in solutions that agreed with experimental data reported by other investigators. It was necessary to develop and apply a technique which provides convergence without oscillation during the iterative solution of the non-linear partial differential equation involved. The computational studies were also extended to the imbibition cycle, i.e., the movement of water into initially dry soil. The computer program had to be revised to accept the negative values of potential and small capillary conductivities encountered in this case.

A laboratory flow model was designed to permit quantitative study of sinking and dispersion phenomena in a system containing miscible liquids of different density. Radioactive iodine was used as a tracer material, the flow pattern being mapped by means of a scintillation detector in a collimating shield. Experiments were completed in which the two liquids were pure water and salt solutions. Solutions having densities of 1.05, 1.07, and 1.10 were examined. In all cases significant sinking rates were detected but attempts to express the effect of density quantitatively in a generalized formula are not yet successful.

Soil Chemistry and Geochemistry

(L. L. Ames, Jr. D. W. Bensen)

From the influence of solution pH on the anion replacement reactions calcite to fluorite, calcite to apatite, gypsum to calcite, and gypsum to apatite it was deduced that the reaction rate is determined by the difference in solubility between the initial and final product. Of the system variables studied, the pH has the greatest effect because these reactions have products containing OH^- and CO_3^{-2} radicals. When a system contains a variety of anions and several final products are possible, only the least soluble under the conditions of the experiment is stable. Changing the conditions (e.g., pH) may

result in a change in the stable product. On the other hand, the concomitant removal of accompanying ions from solution, such as Sr^{+2}, is chiefly governed by the ease with which it fits into the crystal lattice of the stable product.

A reaction in which $CaCO_3$ is replaced by $ZnCO_3$ when contacted by dilute solutions of Zn^{+2} was studied. The reaction was used to test the conclusion that replacement reactions are solubility controlled and to evaluate the mechanism for removing zinc from solution. It was found that zinc is effectively removed from solution in concentration as low as 10^{-7} M and in the presence of as much as 0.3 M sodium ion. Calcite columns flowing at 500 and 5,000 gal/ft^2/day gave a fairly constant zinc decontamination factor of 100. Other variables studied were temperature (25 and 60 C), calcite grain size (0.014 and 0.164 m^2/g), and influent pH (6.0 and 8.0). The data from these experiments substantiate the theoretical conclusion that the rate of mineral replacement reaction is determined by product solubility.

Adequate samples of mineral containing the natural zeolite erionite ($CaO \cdot Al_2O_3 \cdot 6\ SiO_2 \cdot 5\ H_2O$) were obtained from the Nevada State Bureau of Mines to permit experimental studies. The mineral is about 85 per cent erionite. From the reported behavior of the material, erionite was expected to display selective adsorption for calcium and experiments were performed to determine the effect of various cations on the adsorption of Ca^{45}. The results of these experiments indicated a somewhat greater selectivity for adsorbing strontium than calcium and, to a lesser degree, some selectivity for cesium. The selectivity of erionite for none of the ions examined was nearly as marked as that of clinoptilolite for cesium.

Laboratory research to characterize the soil chemistry of rare earth elements is complicated by the fact that some of them, e.g., cerium, are known to exhibit more than one oxidation state. Thus research data obtained with cerium may represent the reactions of Ce(III) or Ce(IV) or a

mixture of the two, which might not be representative of rare earths as a group. For this reason soil chemistry reactions involving radiocerium were repeated with Pm^{147} for comparison since promethium occurs only in the +3 oxidation state. It was found that Pm results duplicated those of Ce over a wide range of conditions, indicating that Ce was largely in that +3 oxidation state throughout these studies. It was necessary to add $NaBiO_3$ to the cerium systems to keep the cerium in the +4 state and obtain significantly greater adsorption than was obtained with promethium. To study the oxidation state of rare earths in an actual waste the adsorption of rare earths from waste samples by soil was determined. Again the adsorption of europium, cerium, and promethium as measured by equilibrium experiments were essentially identical.

Mineral Fixation of High-Level Wastes

(J. L. Nelson, B. W. Mercer)

(Not charged to Biology and Medicine)

Laboratory studies to investigate the possibility of direct decontamination of some types of high-level wastes were continued. Aged Redox high-salt (aluminum) waste was diluted 1:20 with distilled water and passed through four two-foot clinoptilolite columns in series at a flow rate of 50 gal/ft^2/hr. It was found that 98 per cent loading could be attained in the first column before Cs^{137} reached 0.1 MPC in the effluent from the third column. The waste examined was the supernatant liquid from a tank of six-year old Redox waste; the radioactive material it contained was more than 99 per cent cesium-137. The breakthrough curves for strontium and rare-earths were flatter than those for cesium. The cesium from about 50 volumes of undiluted waste can be loaded on one volume of clinoptilolite. Leaching the column with clear water after it was loaded removed about 0.14 per cent of the cesium in 100 column volumes. During leaching the Cs^{137} concentration of the effluent water decreased to less than 0.02 per cent of that in the actual waste. Flushing the column with less than 25 bed volumes of saturated $NaNO_3$ solution eluted essentially all the radiocesium from the bed.

Similar experiments were performed with aluminum coating waste samples. In this case a 240 cm column of clinoptilolite gave an initial decontamination factor of more than 10^7. Again the waste was diluted 1:20 with water because dilution improves the capacity of the mineral for cesium from high-salt solutions. Clinoptilolite column experiments with Purex high-salt waste are not completed, but preliminary results indicate that the cesium from about 30 bed volumes of waste can be loaded on a bed of the mineral. The cesium is readily eluted by 10 M ammonium nitrate solution, essentially all being recovered in two bed volumes of the eluate.

Irradiation experiments revealed an apparent effect of gamma dosages of 10^9 r on the cesium adsorption of clinoptilolite. The effect was studied by means of equilibrium experiments in which distribution coefficients (K_d's) were measured. Using 10^{-4} M cesium solutions at 25 C a 25 per cent reduction in K_d was found when the mineral was irradiated to 10^9 r. At an equilibration temperature of 55 C the irradiated mineral was found to have a K_d that was 29 per cent higher than that for unirradiated clinoptilolite. The reason for this behavior is not known and is the subject of further study.

Ground Waste Investigations

(J. L. Nelson, K. C. Knoll)

A field experiment involving solution discharge to a small 6-inch by 6-inch crib was completed and the soil leached with water. The soil beneath this site is now draining in preparation for additional experiments. It is planned to repeat an earlier experiment with a 2-foot by 2-foot crib at the test site, this time with special measurements of the lateral spread of water beneath the crib. Wells near the crib were probed several times with a neutron moisture detection probe. By the end of the report period very little change in the moisture content of the soil was evident. The calculated volume of water in soil pores beneath a flowing crib at equilibrium was compared with earlier measurements. This estimate of 887 gallons of water contained in soil pores above the water table may be compared with

the estimate of 965 gallons made earlier. These results are considered to agree with the nitrate ion breakthrough which occurred after 800 gallons of solution had been discharged to the crib.

Soil adsorption experiments were performed by means of unsaturated flow through soil columns. The difficulty in maintaining control during these experiments made some of the results uncertain and necessitated numerous repetitive experiments. The study is designed to examine the influence of unsaturation on the utilization of soil adsorption capacity. There is some indication that unsaturation has little effect on the cation adsorption capacity of soils within the moisture content range studied but additional results are needed to confirm this conclusion.

Micromeritics

(A. K. Postma)

Parameters controlling the deposition of particles in conduits due to turbulent diffusion were further clarified. A correlation was established between particle deposition velocity and a function of the important variables. The function is the ratio of particle stopping distance to the thickness of the sub-laminar layer, as defined by particle mechanics and turbulence theory. Correction terms were needed, one for inertial forces acting on the particle and a second empirical factor to achieve the best data fit. The data are in agreement with the expectation that when the sub-laminar layer is thin and the stopping distance large, deposition will be high. The experimentally determined deposition velocities obtained in this study and those reported by others, show satisfactory correlation with this function. A rather wide range of particle diameters, densities, conduit diameters and air velocities are represented. The correlation will prove a valuable aid in estimating losses in sampling lines under a variety of conditions.

An equation was derived which permits estimating the fraction of particles retained in a conduit when the deposition velocity is known. The equation was verified by experimentally determining the fraction of 4 μ ZnS

particles retained in a one-inch diameter 72-foot long pipe. Good agreement was obtained between the actual per cent retained (86 per cent) and deposition predicted (92 per cent). It was further shown that re- entrainment was negligible for the conditions of these experiments.

Single Isotope Analyzers

(C. A. Ratcliffe)

(Not charged to Biology and Medicine)

A chemical separation procedure specific for Np^{239} was selected and the necessary mechanical and electrical components studied to automate the separating, detecting, and recording of this isotope in reactor effluent water. A TTA (thenoyltrifluoroacetone) extraction will be employed with measurement of Np^{239} in the organic phase with a scintillation crystal.

Field Equipment Development

(J. R. Raymond, J. D. McCormack)

The multi-depth water sampler for monitoring wells was tested using seven sampling units in a tandem string. Seven samples were taken simultaneously and brought from a depth of greater than 500 feet below the water table. The sampling unit incorporates a solenoid valve which, when opened from the surface, delivers a water sample to a container. The units are assembled to sections of pipe using quick connect couplings and electrical connections. The sampler will be used to detect and study any vertical stratification of radioactive wastes in the ground water.

A sensitive thermistor flow-sensing element was assembled, tested, and incorporated into an experimental instrument to detect vertical flow of water in wells. Laboratory tests in a simulated well gave sensitivities of the order of 2 ml/min through an eight-inch well casing. The instrument was tested in wells and showed positive flows of the order of 4-8 feet per day vertical movement. Vertical flow is an index to piezometric head differences arising from differing permeabilities in succeeding formations or through the influence of ground water mounds.

Uranium Oxidation and Fission Product Volatilization Studies

(R. K. Hilliard, D. L. Reid)

(In part charged to Biology and Medicine)

All stainless steel parts of the high level experimental equipment were replaced with quartz and Pyrex to facilitate recovery of evolved fission products.

A total of ten oxidation measurements was made under identical conditions of time, temperature and air flow rate. In an air atmosphere at 1215 C furnace temperature for 24 minutes, the 11.5 gram uranium specimens averaged 68 per cent oxidized, with a standard deviation of 3.3 per cent. Four of these specimens had been irradiated to values up to 2.4×10^{17}nvt. The average per cent oxidation of the irradiated specimens was 67 per cent with a 4.3 per cent standard deviation. This shows that irradiation up to 10^{17} nvt does not significantly affect the oxidation rate. Reproducibility of oxidation rates is important since it indicates that constant experimental conditions were maintained between tests. Also, previous work has shown that release of volatile fission products is proportional to the extent of oxidation.

Five fission product release experiments were made, two at 2.4×10^{14} nvt, two at 3.5×10^{16} nvt and one at 2.4×10^{17} nvt. Table IV lists the percentage release for eight isotopes. The tentative conclusion to be drawn from the table is that iodine, tellurium, xenon and cesium are released to a greater extent from uranium irradiated at higher levels. The release of strontium, barium, ruthenium and zirconium appears to be independent of irradiation levels, at least up to nvt values of 2.4×10^{17}.

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TABLE IV

EFFECT OF IRRADIATION ON THE RELEASE OF FISSION PRODUCTS

Isotope	Per Cent Released from Uranium Specimen*		
	2.4×10^{14} nvt	3.5×10^{16} nvt	2.4×10^{17} nvt
I^{131}	71	76	85
Te^{132}	58	64	62
Xe^{133}	80	93	93
Sr	0.2	0.2	-
Cs	17	28	31
Ru^{103}	0.1	0.1	0.05
Ba^{140}	0.3	0.1	-
Zr^{95}	0.03	0.02	-

* Experimental conditions: furnace temperature, 1215 C, time, 24 minutes; atmosphere, air.

Two tests were made to determine the retention efficiencies of "absolute" filters and charcoal beds for air-borne fission products released from uranium heated in air. The filters were Cambridge model ID-100 and a one-inch thick charcoal bed. Both were at ambient temperature. The superficial velocities were about 5 ft/min. The iodine concentration in the off-gas was about 10^{-6} ppm. Table V lists the efficiencies observed.

TABLE V

FILTER EFFICIENCIES FOR REMOVING AIR-BORNE FISSION PRODUCTS RELEASED FROM HEATED URANIUM

Retained On	Per Cent Retained	
	I^{131}	Te^{132}
First Filter	30	99.4
Second Filter	14	92
One-Inch Thick Charcoal	70	50

INTERNAL DISTRIBUTION

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1	F. W. Albaugh
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