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FOR SEPTEMBER 1967

**DIVISION OF PRODUCTION
AND
HANFORD PLANT ASSISTANCE
PROGRAMS**

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C-65, Plutonium Production
(Special Distribution)

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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
SEPTEMBER 1967

Division of Production
and
Hanford Plant Assistance
Programs

by the
Staff of Battelle-Northwest
S. L. Fawcett, Director

October 1967

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

R. S. PAUL

DIRECT AEC SPONSORED PROGRAMS

Summary

Effects of Nuclear Effluent on the Quality of Columbia River Water

Further extensive use of the data and program information developed in this work was made in support of policy studies relative to the proposed standards for water temperature under public review in the State of Washington.

Mechanisms of Environmental Exposure

Measurements of radioactivity in children began at the first of seven Pasco elementary schools on 9/21/67 utilizing the new mobile whole-body counter laboratory. Calibration of this whole-body counter for Na^{24} , K^{40} , Zn^{65} , I^{131} , and Cs^{137} , was completed on 9/15/67. The survey will be conducted in cooperation with the Washington State Department of Game. The first computer run was completed for the program of calculation of environmental radiation doses to the "critical organs" of each individual adult who has a diet record under DIB master file. The initial results appear to be reasonably consistent with expectations, although a few instances of unusual diet information will be investigated for possible keypunch errors.

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DIRECT AEC SPONSORED PROGRAMS

Columbia River Studies

Effect of Reactor Effluent on the Quality of the Columbia River Water

(R. T. Jaske, Environmental and Radiological Sciences Department)

The data generated in this work received further use by the Richland Operations Office in support of their development of policy on the proposed temperature standards under consideration by the State of Washington. Two plan views of the plant reach showing effluent temperature profiles were prepared and forwarded along with interpretive remarks.

Filter plant data collected at the Richland water plant in 1966 have been receiving additional examination for possible correlation with other production plant research. The distribution of floc within the filter media as traced by several radionuclides still promises additional information beyond that obtained last year.

Additional research in optimizing the accuracy of the COL HEAT temperature model have been carried out as a result of direct requests of the RL representative on the Water Temperature Task Force, Pacific Northwest River Basins Commission. At their request the Task Force was given a complete explanation of the simulation program on July 28, 1967. Following this meeting, the Richland Operations Office committed the use of this model system to the Task Force for their use in performing tentative studies of the Snake River Basin. An extensive compilation of plant environmental data was forwarded to the FWPCA in support of a second generation model under development in the same river reach already covered by the existing technology.

Mechanisms of Environmental Exposure

(J. F. Honstead, Environmental Studies Section)

Measurements of radioactivity in children began at the first of seven Pasco elementary schools on 9/21/67 following

computer run also includes a calculation of the percentage contribution of each of several foodstuffs to the radiation exposure received by each of the four critical organs. These latter tabulations reiterated the important contribution to radiation exposure made by consumption of local game birds and Columbia River fish. After the computer run has been spot checked for potential errors, the average dose to each of the four critical organs will be calculated as well as the frequency distribution of dose versus number of persons.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Summary

Mission 1

Study of the simulated rupture test behavior of a high exposure, Mark I, N-Reactor inner fuel component indicates a change in the basic corrosion behavior of the fuel as a consequence of the rupture test. Corrosion tests in boiling water are under way to compare the corrosion behavior of unirradiated and high exposure N fuel.

During the reporting period 45 Mark IV production fuel elements were measured and weighed.

An electrically heated model of a half column of N-Reactor MK-I fuel elements used in earlier experiments was disassembled and examined. This model will be used to extend the range of earlier two-phase pressure drop experiments to higher pressures and inlet temperatures. It has holes drilled through the tube walls to represent spaces at fuel element junctions. The planned experiments require a model with completely separated flow channels. Therefore, plugs for these holes have been fabricated and are ready for installation. The model appears to be in excellent condition and will be ready for use in this present program with only a minimum of additional work.

A test section for measuring hydraulic resistances of various types of obstructions in N-Reactor fuel element cooling channel was completed. The test section provides a rectangular flow channel. Replaceable plugs inserted into one of the channel walls allow the numbers, sizes, and shapes of obstructions to be varied quickly and easily. The results of experiments on this test section will be used to supplement theoretical studies being performed at N-Reactor to establish fuel support design relationships.

Impregnation of the eight graphite target elements with NpO_2 has been completed.

Mission 5

The cross section processing code BARNS-II has been modified to permit the use of negative energy resonances. Preliminary results with U^{235} show a marked improvement in the resulting cross section description.

An error arising in converting the two dimensional transport code 2DF to the UNIVAC 1108 has been eliminated and the converted code will now handle cases with anisotropic scattering.

With successful conversion of ENDF/B library tapes, servicing routines, and the processing code ETOM, multi-group cross sections may now be obtained from the ENDF/B system for use in the slowing down calculations of MUFT and HAMMER.

Methods are being studied to derive Am^{243} and Cm^{244} cross section information from plutonium burnup data.

Mission 7

Radiometallurgy study of high power density fuel elements containing uranium of several compositions and heat treatments has continued. A report document has been written describing the examination of 14 fuel elements, two of which were suspected of having cracked fuel cores. Examination of the fracture surfaces of other cracked fuel cores is planned to help establish the location and possibly the cause of fracture initiation.

Seven Zircaloy-clad nickel-plated uranium cores were prepared by electron beam welding end caps into Zircaloy-2 tubing. These cores will be used in the hot-die sizing operation in an attempt to bond Zircaloy to uranium in the same manner that aluminum is bonded to uranium.

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Mission 1 - Basic Production Mission

Simulated Rupture Testing of High Exposure Mark I Fuels
(J. W. Goffard, Metal Fuels Unit)

Study of the high exposure inner component that was rupture-tested in the Irradiated Rupture Prototype (IRP) facility has continued. This component was initially N-Reactor load Mark I material of standard Fe-Si fuel that successfully attained about 6500 MWd/T exposure (0.75 at.% BU). Its companion outer component with about 8000 MWd/T exposure will also be rupture-tested in the IRP facility, as soon as that facility is returned to operational status after undergoing a safety evaluation. On the basis of metallographic study of the ruptured inner component, it is estimated that about 20 grams of fuel were corroded at the site of the rupture. Some corrosion product remained under the blistered clad. The blistered clad extended over a 2-in. length and 55% around the circumference of the component section. The development of such a large area blister with so small a quantity of fuel corroded resulted because of fuel cracking in the bond zone parallel to the clad. There was no other mode of cracking associated with the fuel corrosion on the rupture damage. Fuel corrosion in the radial direction (depth) was fairly uniform with a maximum penetration of 0.065 in. or about 20% of the fuel wall thickness. Since about 10 hours of exposure in the IRP facility were required to rupture the defected element and since a defected unirradiated element is usually ruptured after an hour of such exposure, it appears that the irradiated fuel has experienced a decrease in corrosion rate. Open-end fuel corrosion tests in boiling water of irradiated and unirradiated inner component wafer sections are under way to determine the effect of high exposure on fuel corrosion rates.

N-Fuel Measurements

(R. K. Marshall, Metal Fuels Unit)

During the reporting period, 45 Mark IV production fuel elements were measured and weighed.

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comparable to those of the spaces between the ends of adjacent fuel elements. The experiments with Rod 328-02 confirmed the beneficial effects of the interjunction flow qualitatively.

Subsequent to the experiments described, the first test section was used in a series of transient experiments investigating various reactor accidents. During these experiments, the model was damaged by high temperatures to the extent that its use for future experiments is unfeasible. The planned experiments to determine liquid and two-phase hydraulic behavior under future operating conditions will require a model with completely separated flow subchannels. Therefore, it was decided to modify the second test section by plugging the inter-channel flow holes. This test section was disassembled and examined and found to be in good condition. During disassembly, thermocouples installed in the fuel model were destroyed and a few ceramic simulated fuel supports were damaged. Plugs for the "junction holes" have been made. Installation of these plugs, new thermocouples, and ceramic supports is in progress.

Effects of Fuel Supports on Subchannel Flow Distribution
(J. K. Anderson, Reactor Engineering Section)

Preliminary design of a test section for measuring hydraulic resistances of various types of obstruction in an N-Reactor fuel element cooling channel was completed.

The purpose of this study is to develop information which can be used in the design of fuel element supports. At present, there is little information about the hydraulic resistance of obstructions such as fuel supports. Support design changes, made to improve strength or fuel stability, can affect the hydraulic resistances of individual flow subchannels and the flow distributions among subchannels. In some cases, this results in imbalances between the flow split and the heat split among the subchannels and can cause tube powers to be restricted to prevent excessive local coolant temperatures.

This test section will be used to obtain experimental measurements of pressure drops across various sizes, shapes,

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PCTR Experiment with Water Flooded Fuels
(D. R. Oden, Reactor Physics Department)

Calculations were started to determine the boron concentration in water required to poison four water lattices of 2.1% enriched uranium coproduct-drivers to a k_{∞} of unity. Hexagonal lattice pitches of 2.4, 2.8, 3.1 and 3.4 in. were chosen for these calculations since critical approach and exponential data are available for these spacings with a water moderator.

Thermos runs for the unpoisoned case were made and the major problem now is the determination of Dancoff coefficients for the tubular geometry.

Vibration Test

(T. J. Davis and L. T. Lamb, NDT Systems Engineering Unit)

The NRD fuel assembly vibration tester has been in use for three weeks, with 100% of outgoing fuels currently being subjected to the test. To date, the tester has been very successful in detecting unsized supports and an analysis of rejected fuel assemblies with no evident unsizing is being conducted.

Modification of 333 Bldg. NDT Equipment

(T. J. Davis, NDT Systems Engineering Unit)

Further analysis was done on the adaptation of the receiving inspection billet enrichment tester for use with new billet configurations. Several electronic modifications were performed. However, it was necessary to magnetically shield both the probe and the billet to obtain optimum count stability. It is anticipated that future testing will have to be performed in a specially constructed shield.

Circuit criteria recently established through associated design efforts on a universal ultrasonic tester will provide for a second attempt at thinclad bond testing using the BNW-developed, high frequency transducers. A passive element switch has been fabricated which reduces both the transmit pulse and its overshoot to an adjustable level of ± 10 to 100

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N-Reactor Component Evaluation; Pressure Tube Evaluation
(M. C. Fraser and R. J. Evans, Materials Application Unit)

The study of the combined effect of fast neutron bombardment and exposure to the N-Reactor stack gas environment upon Zircaloy-2 pressure tubes continued. Five tubular specimens exposed for 122 operating days in an empty N-Reactor process tube channel at temperatures between 600 and 1181°F were previously reported as having their crack propagation strength reduced by over 50%, hydride concentration increased from about 30 ppm to 50 ppm, burst strength essentially unchanged and oxide thicknesses increased from an average of 0.15 mil to one and one-quarter mils on the outside surfaces and from less than 0.01 mil to an average of one and one-quarter mils on the inside surface.

A group of unslotted, slotted, unhydrided and hydrided control specimens is being prepared for exposure to an environmental history minus the irradiation similar to that experienced by the irradiated group. A special furnace and auxiliary equipment to provide the thermal and moist helium environment has been constructed and is undergoing debugging.

Additional tubular specimens have been hydrided to about 200 ppm for irradiation to show the effect of total reactor environment and severe hydriding on tube properties such as fatigue crack propagation strength and burst strength.

Mission 4 - Plutonium-238

Preparation of Neptunium Target Elements
(L. G. Merker, Fuel Process Development Unit)

Two processes for fabricating graphite-NpO₂ target elements for the Pu²³⁸ demonstration program have been tried. Machined graphite pieces, 8½ in. long x 1½ in. diameter, were impregnated by vacuum techniques with 40 grams of neptunium in nitrate solution. These were dried and then high fired in argon at 1000°C for conversion of the nitrate to NpO₂. Four elements were solid pieces and four were machined with a ½ in.

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graphite target elements.

Mission 5 - Other Isotopes

Code Development[#]
(Theoretical Physics Unit)

BARNS-II (K. B. Stewart)

Investigation of the high background calculations by BARNS-II in the resonance region for certain isotopes indicates that parameters for lower energy resonances should be included in the resonance parameter tables to reduce this background. Modifications were made in the code to permit negative energy resonances to be used. Preliminary results with U^{235} show a marked reduction in background when parameters for a negative energy resonance are included. Need for a revision in the unresolved region background is also indicated.

HRG Data Tapes (J. L. Carter)

Several known errors have been corrected in the most recent versions of the HRG library, HRG090767. These errors were introduced during the past few months by incorrect manipulation of data previously calculated by BARNS-II. H_2O , D_2O , Pu^{239} (L)*, and Cu^{63} are affected. Results of a short code to compare data on HRG library tapes in some detail strongly indicate that data for only these nuclides were mishandled. Sources of the errors in H_2O , D_2O , and Cu^{63} were located and corrected; circumstantial evidence points to an 1107 malfunction as the source of the Pu^{239} (L)* error.

Program 2DF (K. B. Stewart)

The computer program 2DF was corrected to allow anisotropic cases to execute on the UNIVAC 1108 computers. Previously, a system error diagnostic terminated execution because

[#] Partially supported by the Division of Reactor Development and Technology.

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in a plutonium fuel are being continued. Analysis of the Am^{243} and Cm^{244} data is dependent upon an adequate description of its plutonium predecessors. Present efforts are directed at improving the plutonium analysis by changing the functional form of $\sigma_a^{40}/\sigma_a^{49}$ (the ratio of the Pu^{240} to Pu^{239} effective absorption cross sections). The necessary analysis codes have been modified and are being tested.

Mission 7 - Target Space Enhancement

Zirconium Plating of Uranium

(R. W. Stromatt, Chemical Separations Unit)

Previous results of the work on electroplating zirconium onto uranium metal out of a molten fluoride salt system have indicated that the production of thin, relatively uniform zirconium plates is indeed possible. These results have also indicated that as the plating progresses, something happens to the melt which interferes with the build-up of a thick zirconium plate. Preparations are now being made to test the concept that continual conditioning of the plating bath, by continually recirculating the melt through a chamber where it can be hydro-fluorinated, will permit the build-up of thick zirconium deposits. Apparatus is being constructed for this experiment. A preliminary trial of a gas-lift melt recirculation system was successful.

High Power Density Alloy Fuel Evaluation

(J. W. Weber, Metal Fuels Unit)

Examination of fuel elements containing alloys being evaluated for high power density applications has continued in Radiometallurgy. A report was written describing the results of the examination of 14 fuel elements for evidence of fuel core cracking which might have occurred during irradiation. Ultrasonic tests in the C Reactor basin indicated that two of these elements had large core cracks.

The results of this examination have been described in previous monthly reports in this series. The conclusions

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with the aluminum assemblies. The assemblies were helium leak checked after welding and no leaks were found.

Aluminum Alloys

(R. R. Studer, Fabrication Metallurgy Unit)

During September, four ingots of 8001 alloy were made using the Direct Chill casting machine built for this project. Before making the drops, a graphite tundish and runner was constructed and a structure to support the tundish was added to the machine.

Of the four drops, the first two were aborted because of difficulties in controlling rate of drop and pouring rate. The third and fourth drops yielded billets of good soundness and surface appearance and had sufficient length to provide an extrudable billet. The fourth drop was the maximum length possible on the machine.

The weaknesses of the casting machine derive mainly from lack of positive reproducible speed control. A larger hydraulic cylinder is being installed to reduce the sensitivity to the control orifice settings.

An aluminum mold sleeve was used for the four successful castings. A mold lubricant of 1/3 castor oil and 2/3 corn oil performed satisfactorily. In view of the large number of "best" mold lubricants in use in different installations, all containing animal or vegetable fats or both, it seems probable that any animal or vegetable fat mixture of the proper viscosity for the existing operating conditions will do an adequate job. In the present instance, a lesser addition of castor oil would have been preferable as the 1-2 mixture was slightly stiff for best ease of application.

The two abortive drops were sectioned for macro and micro-examination. No solidification shrink was found but a large amount of fine dispersed oxides and a few gross inclusions of oxide skins were evident.

Fluxing in the melt crucible was not effective because the rapid cooling while power was decreased during fluxing

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and ν^{25} . Based on these calculated values of $\nu^{28} = 2.73734$, $\nu^{25} = 2.4317$, and $\alpha_{fast}^{28} = 0.39284$, the relationship between ϵ and δ^{28} becomes

$$\epsilon = 1 + 0.5529 \delta^{28}.$$

Depleted uranium foils and 2.1% enriched uranium foils both irradiated in the fuel spectrum environment were used for the determination of δ^{28} . Fission product activity between 510 and 675 keV was measured as a function of time for each foil on a 512 channel analyzer. The ratio at a time t after irradiation, of the measured fission product activity in the depleted uranium foil to the activity of the 2.1% enriched uranium foil, $\gamma(t)$, was thus determined. The number of counts per fission of U^{235} to the number of counts per fission of U^{238} , $P(t)$, was determined for the energy interval between 510 and 675 keV by a correlation of La^{140} counting data with fission product activity in this energy interval. Values for δ^{28} were obtained from the equation

$$\delta^{28} = P(t) \left\{ \gamma(t) \left(\frac{N_D^{28}}{N_F^{28}} \right) \left[1 + \left(\frac{N_D^{25}}{N_F^{25} - N_D^{25}} \right) \right] - \left(\frac{N_D^{28} - N_D^{25}}{N_F^{28} (N_F^{25} - N_D^{25})} \right) \right\}$$

where N^{28} and N^{25} are nuclei densities of U^{238} and U^{235} , respectively. Subscripts D and F refer to depleted uranium foils and 2.1% enriched uranium fuel foils, respectively. The values thus obtained are: $\delta^{28} = 0.082 \pm 0.004$, and $\epsilon = 1.0454 \pm 0.0023$.

Overbore Lattice Experiment in the PCTR

(A. D. Vaughn, Heavy Moderator Reactor Physics Unit)

The PCTR experiment to measure the neutronic parameters of the CMIE fuel geometry in a 7.5 in. pitch graphite lattice is being prepared. All experimental materials are currently on order including the fuel.

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naturally excited vibrations.

Professor G. M. Smith, structural dynamics consultant to the Engineering Mechanics Unit, from the University of Nebraska, during a recent visit to PNL, was briefed on the status of the work. He then gave some constructive suggestions for the study. Main conclusions reached were:

1. The signal to the recorders of our present seismic detection units should be amplified by a factor of 10. (This will make data taken from the recordings sufficiently accurate for constructing acceleration response spectra.)
2. Acceleration response spectra, constructed using data from the amplified recordings, will indicate coupling between the basalt and reactor foundations and also the possible earthquake forces on Hanford structures.
3. We should attempt to find the vibration characteristics of a reactor enclosure structure by taking strain-measurements on the structure during a high wind.

A tentative decision was made to purchase a separate strong-motion seismograph. The chance of a strong-motion earthquake occurring at Hanford is small but the acceleration record of such an event would be invaluable.

Nuclear Safety Reviews

(C. L. Brown, Critical Mass Physics Section)

Nuclear Safety Specification No. 510, covering the use of the Model 44 shipping containers, was reviewed and comments submitted to N-Fuels Engineering. The Model 44 container is used for shipping 2.1 wt% U^{235} enriched uranium and UO_2 to Fernald, Ohio.

Area 14A, a new criticality safety control area in the 333 Building, was reviewed. Area 14A was established for vibration testing, inspection, measurement, and reshoeing Mark I fuel elements designated for the Mark IC program. When the

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Electrode stubs from the primary melting will be used for the pad material for the initial skull build-up melts.

Low Temperature Zircaloy Hydriding

(W. K. Winegardner, Advanced Systems Corrosion Unit)

Zircaloy electrodes from out-of-reactor tests to investigate cathodic charging of hydrogen into Zircaloy-2 were examined after 2275 hours of additional exposure. Analyses indicate a rate of increase of base metal hydrogen concentration in the electrodes of about 0.1 ppm per day. The draft of a report describing hydride formation in Hanford Production Reactor process tubes was completed.

Electrolytic cells of the out-of-reactor test facility consist of platinum anodes surrounded by tubular Zircaloy-2 cathodes. The electrolyte currently being used is reactor process water at about 90°C. In a previous test, 385 hours of exposure with current densities of about 0.4 and 0.005 mA/cm² resulted in a base metal (material underlying the hydride case) hydrogen concentration of 12 ppm, a value not significantly different from that found in material with no exposure. After 2660 hours, analyses of base metal samples from cells with current densities of 0.4 and 0.005 mA/cm² revealed concentrations of 23 to 32 and 23 to 25 ppm, respectively. Total hydrogen concentration (analyses with the hydride case intact) increased from 80 to 84 to 605 to 571 during the 2275 hour exposure with a c.d. of 0.005 mA/cm². Total hydrogen concentration of cathode material from the cell with a c.d. of 0.4 mA/cm² was too great for accurate analyses.

A Zircaloy-2 electrode, previously hydrided to a base metal concentration of 15 to 17 ppm by an applied potential, was exposed during the 2275 hour period with no driving force for hydriding, i.e., with zero current density. Analyses of base metal after the additional exposure resulted in values of 18 to 19 ppm. Prior experience suggests that the indication of a slight increase is probably the result of analytical accuracy or sample nonuniformity, not migration from the hydride case to underlying material.

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four months exposure continue to confirm the initial data reported in June: stress corrosion cracking on 304 SS tubing from an N Reactor steam generator occurs in the liquid and vapor phases of solutions not containing phosphates and on the vapor phase over solutions containing phosphate but not on samples immersed in the liquid phase of any solution containing phosphate ion. Stress corrosion cracking has not occurred on other lots of 304 stainless steel heat treated similarly to the steam generator tubing nor has it occurred on Inconel 600, Incoloy 800, or A212CS. Weight loss on the stainless steel and nickel base alloys is negligible.

Coating Formation on Graphite

(R. C. Giberson, Materials Compatibility Unit)

A study was started to determine the feasibility of depositing a silicon coating on graphite to reduce its oxidation rate. In the first two experiments, small cylinders of TSX graphite (0.43 in. diameter x 1.0 in. long) were treated with flowing helium containing ~1300 ppm of tetramethyl silane (TMS) for one hour at 750°C. The samples were oxidized in air at 650°C before and after the treatment. For comparison purposes, untreated samples were given the same oxidation test. During the coating process, the treated samples gained from 5 to 10 mg. The initial oxidation rate, after coating, was reduced to 1/2-1/10. However, in two to four hours, the rates had returned to the same value as those of the non-treated samples.

Reaction of KC Graphite with Water Vapor

(R. C. Giberson, Materials Compatibility Unit)

The study of the reaction of KC graphite with water vapor was completed. The range of experimental conditions was as follows:

Temperature	800 to 1000°C
Pressure of water vapor	500 to 10,000 ppm
Pressure of hydrogen	0 to 5,000 ppm

Helium gas at one atmosphere total pressure was used as the inert carrier in these experiments. The flow rate was ~500 cc/min.

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and the coated rod was then oxidized in air. Comparison of the rate of oxidation of a similar rod, not treated with TMS, indicated that some protection was achieved with the TMS. In the reactor a compound must have sufficient radiation stability to permit it to come in contact with the hottest graphite. A study of the radiolysis of silane, SiH_4 , indicated that it is rapidly decomposed by gamma radiation yielding a $G(-\text{SiH}_4)$ of 12,000 molecules decomposed/100 eV at 350°C with an activation energy of 70 kcal/mole.

In light of these very-high decomposition rates reported for the silane, it is necessary to investigate the radiation stability of other silane compounds before continuing with the coating studies. Therefore, TMS was exposed to Co^{60} gamma rays for 0.5 hr at a dose rate of 2.9×10^7 R/hr at 350 and 150°C . G values observed for the production of methane were 10 and 6 molecules/100 eV at 350 and 150°C , respectively. This temperature effect corresponds to an activation energy of about 2 kcal/mole. In addition to methane, hydrogen was produced with G values of 3 and 1 molecules/100 eV at 350 and 150°C , respectively. Six other products were observed but were not identified. These decomposition rates for TMS by radiation are about as would be expected and further studies will, therefore, be conducted to determine the feasibility of coating the graphite in the reactor field.

Burnout Monitoring

(N. R. Gordon, Materials Compatibility Unit)

Channels 1675-D and 3567-D, charged November 26, 1966, were discharged July 6, 1967. This was the first period of use for Channel 3567-D in the graphite oxidation program. Channel 3478-D had been used previously but was discontinued because its extended use appeared to be creating non-typical high temperatures, thus resulting in high oxidation rates.

Maximum rates of 1.8% per thousand operating days (KOD) were recorded in Channel 1675, 130 in. from the front face and 5%/KOD 225 in. from the front face. The first maximum is somewhat larger than has been seen in previous charges. It may be due to a very small oxygen leak, but the data are not definitive.

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A small generalized open-channel model is being assembled to determine the sensitivity of the infrared scanning system for determining temperature gradients in models. The model will simulate a short section of a river into which water at selected temperatures can be injected. River flow rates and warm water temperatures and injection rates can be controlled over a wide range.

Radiation Detection and Measurement Instrumentation
(E. M. Sheen, Instrumentation Section)

Liaison effort with personnel of the DUN Research and Engineering Section revealed a definite need for a "neutron rem meter" of portable design in a single package and not exceeding 15 pounds total weight. Harwell has developed such an instrument which may be commercially available from Isotope Developments, Ltd., Reading, England. If so, and if it performs as described in the literature, only minor modifications would be necessary to adapt it to the DUN design requirements. Los Alamos has also considered the design possibilities of a rem-responding, lightweight, 8 in. diameter, cadmium-loaded polyethylene sphere. One of these, together with a suitable neutron detector and associated electronic circuitry, may also meet the DUN operational requirements. Investigation continues into the various possibilities.

Analytical Instrumentation for N-Reactor
(G. B. Barton, Fission Product Chemistry Unit)

The Panametrics Hygrometer was in satisfactory operation when the reactor was shut down. It follows the moisture changes that occur on startup and dryer switch over that were never seen on the older instruments. The elimination of the long sample lines is a major contributor to this improved response.

The Bendix-Greenbriar Company is having difficulty with some of the adjustments of the chromatograph so it will be delayed until some time in October.

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Mission 1

Work continued on the preparation and properties of the nitropropane derivative under study for use in catalyzing the oxidation of NpO_2^+ by HNO_2 - HNO_3 . A preliminary measurement of radiation stability was made.

Using silica-gel adsorption techniques, it was estimated that a current NPH sample contains about 0.05% alcohol.

Work continued on the alpha-radiolysis of plutonium(IV)-nitric acid solutions. The results are summarized.

A Purex Plant production test of a DBBP batch solvent extraction process for recovering Np and Pu from the Purex LW waste stream is scheduled for the near future.

High-purity N-silica shows significantly better corrosion resistance than 30% L to several environments pertinent to fuels processing in the Purex plant.

The experimental Am^{241} Breakthrough Monitoring System, being evaluated for measurement of process material concentrations in the W-14 Column, continued to demonstrate proper operational performance.

Design of a remote sampling system for the Purex HA column waste stream was completed. The system provides a means of batch extracting plutonium and neptunium from the high activity fission products, resulting in a tenfold increase in the amount of sample available for analysis.

Single drop extraction studies in the UNH-n dodecane-30% TBP system showed that the mass transfer rates were too low to detect the concentration limit of density instability by schlieren techniques.

Soil column studies of N-Area decontamination waste showed that DF's of 5.2 for Co^{60} and 3.6 for Cs^{137} were obtained. Co^{60} decontamination was effected by filtration of suspended solids and, although Cs^{137} was exchangeable, the decontamination was not commensurate with that found in other soil-waste systems.

Statistical column studies using the zeolite AW-500 for decontamination of ITS condensate were completed.

Uptake of Cs^{137} by soil previously saturated with organic solvents is being studied. After 100 column volumes of spiked tap water no breakthrough is evident. Studies on the removal of Pu^{239} and Am^{241} deposited on soil by CAW waste are continuing.

Electrolytic oxidation of cerium was carried out successfully in a titanium vessel.

Attempts to define a single NaOH-complexant B-Plant solvent wash to remove M2EHFA and metallic contaminants are promising.

A four stage short section of a packed bed of molecular sieves for removal of moisture and oxygen explosions is being tested. A preliminary effect on

A four circulator system with baffles to divide the tank into four parts is being installed to determine whether the unit cell theory of circulator interaction is valid by comparing baffled and unbaffled systems.

A preliminary analysis of potential organic waste streams from the ARHCO processing plants is being compiled on organic waste streams from the ARHCO processing plants.

Process Technology

The cause of cracking in plutonium nitrate storage bottles is being investigated. Several bottles have been tested at ambient temperature in contact with a surfactant. No cracking has been observed after two weeks. After 30 days at room temperature and 21 days at 40°C in contact with 100% Igepal,

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Mission 1 - Basic Production Mission

Neptunium Chemistry

(J. L. Swanson, Chemical Separations Unit)

Efforts to increase the yield of the NpO_2^+ oxidation rate-accelerating material obtained when HNO_3 is added to the aci-form of 1-nitropropane continued. A several-fold increase in yield obtained this month may have been due to more vigorous agitation of the solution as the HNO_3 was added.

An experiment was done to measure the stability of the rate accelerator in gamma-irradiated 3.5M HNO_3 . The sample was exposed to a gamma field of 8.7×10^5 R/hr for 0.1 hr and its catalytic activity compared with the catalytic activity before irradiation. A 40% decrease was observed. Assuming 100% conversion of the nitropropane to the rate-accelerating material, the accelerator concentrations were 0.018M before irradiation and 0.011M after irradiation. Again assuming 100% conversion, these values correspond to an unusually high "G" value of about 100 for destruction of the accelerator. This probably signifies a conversion yield of nitropropane to accelerator of much less than 100%.

Purex Diluent Studies

(M. R. Young, Heavy Element Chemistry Unit)

Studies of methods for NPH quality determination continued. In work on the determination of alcohol based on adsorption on silica gel, difficulty was experienced from interference of the methanol or ethanol eluants and acetone is now used as eluant. A standard concentration vs. intensity curve is now being made and minimum reagent requirements are being established. Tentative results for a current NPH sample show the alcohol content to be about 0.05%. Olefins are also concentrated by the procedure. By assuming 100% recovery of the olefin, infrared measurements indicate an original olefin concentration of about 0.0075%.

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with plant LWW demonstrate that HNO_3 and oxalic acid scrub behavior of Zr^{95} and Ru^{106} are independent of the order in which the scrubs are performed. Additional work confirmed that oxalic acid strips plutonium and neptunium rapidly from the DBBP phase and that, although practically all the uranium in the LWW solution coextracts with the plutonium and neptunium, only about 10% of it strips into the oxalic acid strip. Finally, extraction contacts with various synthetic LWW solutions demonstrated that the proposed DBBP process will recover both plutonium and neptunium over a wide range of LWW solution acidities, sulfate, and iron concentrations.

Corrosion Studies on Tophet M Alloy

(R. F. Maness, Materials and Process Chemistry Unit)

Weldments of Tophet M alloy were fabricated and corrosion tested in boiling solutions of 65 wt% HNO_3 , Zirflex dissolvent, simulated Purex LWW, simulated Purex Zirflex LWW, and HNO_3 -HF solutions. No preferential attack occurred in any of these tests. Corrosion rates were about 1/2 to 1/5 of those found for 304L stainless steel in similar solutions.

Waste Stream Sampling System

(J. Dunn, Process Systems Development Unit)

Analysis of the HA-Column waste stream in Purex for neptunium and plutonium is complicated by the small amounts of these two elements and the high percentage of fission products. Radiation limits the sample size in the range 200 λ to 1 ml. This quantity is insufficient to permit accurate laboratory analysis for neptunium. In order to provide an effectively larger sample for analysis it is necessary to make a preliminary partition by solvent extraction (or other means, e.g., ion exchange) in the sampler box itself. A system design substituting an extraction cup for part of the piping installed in a jumper in the H-2 sample cell was completed. The system permits a ten-fold reduction in the activity with a corresponding increase in the amount of sample used for neptunium determination. Drawings have been sent to ARHCO for comment.

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of temperature compensation was investigated; it was determined that a temperature compensator must be of a quadratic form for spans on the order to 50°C to achieve accuracies of 1%. The rates of change of conductivity with respect to temperatures are about 1.5% per $^{\circ}\text{C}$ at 25°C and 0.6% per $^{\circ}\text{C}$ at 65°C . Due to the large temperature effect the compensator requires the use of thermistors. In a laboratory-developed conductivity method, conductivity is related to a pulse height which is converted to dc. One method of conversion of pulse height to dc incorporates an operational amplifier with a diode peak detector. A back diode was used to give better sensitivity at low pulse heights. Due to leakage of the diode and the narrow pulse width (0.25 μsec) with respect to the pulse rate, attenuation of the signal is high. The availability of low cost operational amplifiers makes this approach attractive.

Fundamental Mass Transfer Studies

(P. C. Owzarski, Chemical Engineering Analysis Unit)

A series of repeat single drop extractions was carried out on a high concentration UNH (0.45M)-n dodecane-30% TBP forming and withdrawing drop into a stagnant water reservoir at 25°C . Drops with 0.0763 ml volumes were formed at 11.5 per minute at a 0.120 in. nozzle tip. The mass transfer rate is very low, each drop losing 3.8×10^{-7} mole to a 10 ml reservoir.

The significance of the low transfer rates puts a serious limitation on transfer studies from organic to aqueous phases. A much slower pump rate may allow choosing lower UNH feed concentrations to find the concentration limit of density instability observed in transfer in this direction by schlieren techniques. As a result, future runs will be primarily concerned with transfer from aqueous to organic where interfacial turbulence is suspected. The effect of surfactants should be more important in this system.

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The neutron dose rate expected through the back door of A-Cell during curium processing was calculated to be 250 mrem/hr assuming a 60 g curium source five feet from the door. During much of the processing, the neutron thermalization afforded by the resin and water jackets will reduce this dose rate appreciably, but when the curium is on the small columns, or isolated as the oxide, there will be little shielding effect. About 7 inches of water would be required for a reduction to one-tenth.

Mission 4 - Plutonium-238

Extraction of Np and Pu with DBBP-CCl₄

(L. A. Bray, Fission Product Chemistry Unit)

Experiments are in progress to evaluate DBBP solvent extraction for Np-Pu²³⁸ processing (as an alternate to ion exchange for 234-5 Building application). From initial experiments it appears that Np and Pu can be coextracted from Ce(IV) or Cr(VI) oxidized solutions and that they can be adequately separated by ferrous reduction of the plutonium. Additional laboratory and follow-on hot cell experiments are necessary to fully define a flowsheet.

Work was also started on the TBP extraction of Np and Pu--for proposed Purex plant application.

Mission 5 - Other Isotopes

Isotope Separation

(R. E. Lerch, Fission Product Chemistry Unit)

Experiments aimed at evaluating the pulsed current electromigration technique are under way. Preliminary experiments using single pulses and high frequency AC pulses in a silica gel migration column were completed. Overvoltage and cell relaxation time were observed in relation to various current and pulse widths for single pulses. No advantage was gained by long relaxation times and it appears that the previously used square wave is probably close to ideal. Rate of lithium ion migration was greater the higher the frequency; however,

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indicated less than 500 dpm on a poppy. Analytical results have not yet been received for these runs.

The residue from the ambient temperature run appeared to be dry after passing air over the shallow cup for 24 hours. However, after several days of storage in a glove box, the residue showed some surface cracking due to further evaporation. This means that the solution had not completely dried in the 24 hour period. The lack of complete drying was confirmed in the elevated evaporation run at 50°C since 25 hours were required for drying. For this run, the residue surface was cracked and pitted. No apparent change in the appearance of the residue took place after being exposed to the dry glove box air for several days.

Next, the residues were heated to either 700 or 1000°C and air passed over the residues at a higher velocity of 100 cm/sec. The residue initially formed by evaporation at ambient temperature was heated to 1000°C. The residue began bubbling at approximately 530°C and finally formed a single large moss-green flake. The shimstock chimney liner through which existing air passed was rusted over its entire length with the exception of the lower two inches. Microscopic examination of the filter at the upper end of the shimstock revealed some sparsely scattered flecks, some flakes as long as 125μ and one or two lumps approximately 75μ in diameter. The residue from the solution evaporated at 50°C was heated to 700°C. Vigorous bubbling of the sample was noted at approximately 290°C. The remaining characteristics of this run were quite similar to those of the 1000°C run. Analytical work is in progress.

Mission 9 - Waste Management

Organic Waste Oxidation

(K. C. Knoll, Water and Wastewater Research Unit)

Research was started to study the effect of air feed on oxidation efficiency as measured by CO₂ production. The CO₂ is measured by a gas chromatograph. Linde SK 400 (a Y-type silicate with nickel loading) produced the maximum CO₂ per unit

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reviewed with ARHCO personnel. The trailer used for column feed was disconnected from service and decontamination measures are under way.

Organic Waste Disposal to Ground

(K. C. Knoll, Water and Wastewater Research Unit)

Uptake of Cs^{137} by soil previously saturated with an organic solution was measured. Soil columns were prepared and saturated with the following organic solutions:

TBP - CCl_4 , 20-80%
DBBP - CCl_4 , 30-70%
0.4M D_2HEPA - 0.2M TBP in NPH
Fab Oil.

After these columns were allowed to drain by gravity, tap water, spiked with Cs^{137} , was put through by gravity flow. Control columns which had not been saturated with organic were also infiltrated with the Cs^{137} spiked tap water. After the passage of about 100 column volumes there was no breakthrough of Cs^{137} from any of the columns.

Studies of the removal of Pu^{239} and Am^{241} deposited on soil from CAW waste by the passage of DBBP- CCl_4 (30-70%) through the soil were not successful. In order to accelerate the experiment, the CAW was pumped through the column. The increased flow rate caused the high-salt, acidic waste to plug the soil and stop infiltration. This work is being repeated using gravity flow.

Cerium Oxidation

(R. E. Lerch, Fission Product Chemistry Unit)

A successful electrolytic oxidation of cerium was carried out in a titanium vessel, conclusively showing that titanium would be a satisfactory material of construction for a plant-scale oxidizer.

In related stainless steel corrosion rate studies, the dissolution rate was shown to increase cerium concentration,

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Silicone Lubricants

(R. F. Maness, Materials and Process Chemistry Unit)

A literature survey was made to determine the feasibility of utilizing a silicone fluid for lubricating a pump to be used in the in-tank-solidification program (graphite bearing - 416 SS shaft). Both dimethyl and methyl phenyl silicone fluids possess adequate thermal, chemical and radiation stability. However, their lubricity for the graphite-steel combination was reported to be poor. Fluorosilicone and chlorophenyl silicone fluids are appreciably better lubricants. However, their use may be limited by radiation stability.

Hydrogen-Oxygen Safety

(W. E. Willingham, Chemical Engineering Analysis Unit)

It appears that the instrumentation problem has been partially solved by making certain changes in the trigger mechanism in the velocity probes. Several runs have been carried out with both time counters in operation and consistent velocities were obtained. Preliminary work shows that over a very short section of packed bed, the (flame/detonation) wave velocity is reduced by approximately 25%. Additional runs are being planned to cover various bed depths and pressure ratios to see if a detonation wave can be suppressed within the system.

B-Plant Evaporator Studies

(W. J. Van Slyke, Engineering Development Unit)

Three batch evaporation tests were completed with the process in-tube bundle; two runs were made using a typical sludge processing composition and one run was made with an alkaline waste composition. Concentration factors in the range of seven to ten were achieved. The results of heat transfer coefficient measurements show that excellent boilup rates (in the range of 350 to 550 Btu/hr·ft²·°F) can be achieved with these waste compositions at the concentration factor of three which is presently planned for B-Plant operation. The boilup rates drop quite sharply at concentration factors of five and greater.

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In-Tank Circulation Studies

(W. E. Willingham, Chemical Engineering Analysis Unit)

Internal cross baffles are being constructed to permit the study of circulation patterns with and without baffles to test the unit cell theory of circulator interaction.

The various probes for velocity measurements within the tank are being calibrated. First indications are that they will not perform properly within the range of interest.

Land Release Studies

(D. B. Cearlock, Geochemical and Geophysical Research Unit)

The land located within the Horn Rapids Triangle is possibly suitable for irrigation by water supplied from the groundwater. Therefore, consideration was given to the effects on the present groundwater flow system from total development of the groundwater supply for irrigation of this land. It was estimated that the available groundwater would be adequate to irrigate approximately 37% of the total land lying within the Horn Rapids Triangle. The results showed that the water table would be lowered from 10-30 feet beneath the 300 Area, two feet beneath 200-East Area and one foot beneath 200-West Area. Also, the wastes originating from the 200 Areas that were predicted to enter the Columbia River below the Ben Franklin Dam would converge into the pumped area upon full development of the groundwater supply. Radiological health implications should be examined in more detail if such an irrigation scheme is contemplated.

Irrigation of a region lying near the 300 Area ("300 Area Irrigation Project") consisting of approximately 29 sections of land is being simulated to determine the effects on the water table beneath the 200 Areas. The first computer pass indicated drainage will be required. A complicated system of drains might have to be constructed in order to provide adequate drainage since the depth to the water table in much of this area will be less than 20 feet with the construction of the Ben Franklin Dam 400-foot pool. An adequate drainage system is being simulated and the solution to the mathematical model will be obtained and results analyzed.

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BNWL-579

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluation

Some wind drifting of radioactive particulates from previous emission was detected in daily surveys just outside the 300 Area fence.

Radioactive insects were found at night-lighted locations around the 300 Area, but do not constitute human exposure problems.

The concentration of diesel fuel in river water at 100-D has decreased during the month to the approximate concentration found at Priest Rapids (0.15 ppm). Apparently, the enlarged trench at 100-N has been effective in accomplishing this reduction.

Personal Dosimetry

Procedures were developed for evaluating the deposition of uranium in the human lung. Improvement in americium-241 detection level will improve the capability of measuring plutonium-239 in human lungs.

Dosimetry Studies

Development work on a portable neutron survey instrument continued as did studies on the performance characteristics of thermoluminescent materials and radiophotoluminescent glass in dosimetry work.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluation

(J. F. Honstead, Environmental Studies Section)

Daily surveys of control survey plots and special particulate traps around the 300 Area, supplemented by re-surveys of roadways and walkways in the 300 Area south parking lot, were started following the detection of a radioactive particulate emission from the 325 Radiochemistry Building reported previously. A few additional particles have been found near the area fence during these surveys, attributed at present to wind-drifting from areas not yet covered or cleared.

The occurrence of radioactive insects near the river has been a frequent occurrence in past years. Piles of dead gnats around lights burning at night around the 300 Area were found during the month to have activity levels as high as 40 nCi/g on a total beta count. The activity was identified as predominantly Zn^{65} and P^{32} , indicating river water as the probable source. Although the occurrence of some radioactivity in such insects is probably general in locations near the river, off-site as well as on-site, no unusual human exposure would be anticipated.

Evaluation of problems associated with fuel oil entering the Columbia River at 100-D continued. Analysis of river samples from four locations (Priest Rapids, 100-D, 100-F, and Richland pumping plant) indicated a decrease in concentration at 100-D from approximately 0.7 ppm, detected last month, to 0.15 ppm. This is approximately the same concentration detected at the other three locations, and thus it appears that little or no oil is presently reaching 100-D from the 100-N oil spill. Both visual observations and analytical results indicate that the enlarged trench at 100-N has been effective in reducing the amount of oil entering the river.

accomplished at Hanford utilizing the 60 keV characteristic gamma ray emitted from the small fraction of americium present in our production plutonium. Detection limit for americium-241, and, therefore, for plutonium-239, was reduced to about 1/3 (to 0.4 nCi) through the development of a more accurate Compton background correction method. The Compton correction previously utilized was based on the average count in channels immediately above and below the 60 keV americium-241 channels. The new correction utilizes an equation dependent upon the subject's weight and the count in a carefully selected band of channels at about 260 keV. Use of this type of Compton correction is being investigated for other radionuclides whose detection limit could be advantageously reduced.

Accurate location of plutonium or other heavy metal particles within tissue is a necessity for efficient excision. Nondestructive Testing Methods Development was requested to study the feasibility of producing a highly sensitive resonance tuned electromagnetic (eddy current) sensing device for locating heavy metal particles in human tissue. A probe was designed and fabricated that was able to detect 1/16-in. long sections of 1/16-in. diameter Hastelloy embedded in cardboard. This probe was replaced with a more sophisticated dual coil probe. The dual coil probe balances the relative signals from each coil against the other to obtain a null, indicating that the metal detected was equidistant from each coil in that plane. Scanning to maximize the signal defines the location in that plane. Scanning at 90° to the first plane defines the location in two planes. Depth is determined by triangulation. The dual coil technique allows larger coils and higher power to be employed, thereby increasing the sensitivity. The initial development stage detection goal is a 0.010-in. long section of 0.001-in. diameter plutonium (or plutonium equivalent) buried one centimeter deep in tissue. Manganese was determined to be a better electromagnetic substitute for plutonium metal than Hastelloy "C", but pure metallic manganese has been difficult to obtain in a satisfactory form. The detection, by the eddy current method, of other metals buried in tissue is a matter of selecting the optimum resonance frequency for detection of the particular metal of interest.

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Dosimetry Studies

(C. E. Newton, Jr., Biophysical Sciences Section)

Thermoluminescent Dosimeter

The study of the thermoluminescent LiF-Teflon discs was completed and a report on the characteristics of the dosimeters written. Special studies of the response of the LiF-Teflon discs to beta and low energy photon radiation show that 0.030 in. of aluminum effectively shields the uranium beta radiation. With this system it was possible to detect a minimum of 200 mrad exposure to uranium beta. Exposures made with the discs stacked on top of each other did not yield satisfactory results. It was not possible to obtain a good difference reading to determine beta exposure. The low energy photon exposures showed even less difference.

Preliminary design on a small quarterly type badge dosimeter was started. The dosimeter uses two TLD discs; one very lightly shielded from room light, the other shielded with 0.015 in. of iron. With this combination the response of the system to photon radiation from 10 keV to 1 MeV was calculated. Using the ratio of the lightly shielded to the iron shielded readings, it is possible to detect the presence of low energy photons. At 60 keV the ratio is 1.40, and at 30 keV the ratio is 10.

The preliminary idea for payroll code is a series of slots that vary in either width or depth according to payroll number. These slots would be placed in the edge of the device holding the TLD discs. It should be possible to automate this type of payroll number readout. The dosimeter package is small enough to fit in the plastic pouch now used to hold the security credential. Decisions on the use of the TLD discs which have a destructive readout are forthcoming.

Portable Neutron Survey Instrument

The tissue-equivalent proportional counter program continued with the first attempts to use the sealed 2 in. counter.

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BFQ instrument an estimate of the neutron dose was obtained that was about 15% of the dose interpreted by the fission fragment neutron dosimeter. It is known that the BFQ response falls off as the energies of the neutrons go above 5 MeV.

Neutron Criticality Alarms

Technical assistance was given regarding the selection of a commercially available BF₃ tube for the neutron criticality alarms. The tube selected is expected to improve the signal-to-noise ratio and thus decrease the power line noise sensitivity. Should the tube provide the necessary improvements, minor mechanical and electrical modifications will be required on the moderator, pulse amplifier and high voltage supply.

Multichannel Analyzer System

Specifications are being written for a multichannel analyzer system for use by the Environmental Surveillance Section of the Environmental Health Department.

Aquatic Biology Services (H. A. Kornberg, Biology Department)

Eighty-eight jack chinook salmon (precocious males) were obtained from Priest Rapids. These fish will be acclimated to different temperatures and challenged with high water temperatures to determine the response of adult salmon to sudden exposure to warm waters. No data are presently available on the thermal tolerance of our local chinook adults. Such data will be useful in evaluating the effect of thermal plumes.

Pre-immune yearling rainbow trout were obtained from the Natches, Washington trout hatchery, were immunized during the past 2 months, and transported to the Bonneville, Washington ladder. Thirty immunized and control trout were placed in a trough with the circulating water from the fish ladder and exposed to columnaris infection from the organisms present naturally in the water. A similar exposure test was set up locally under laboratory conditions. All fish were cold marked

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requirements of a given sampling system. Once the purpose of the system is clearly established, a system can be designed to adequately meet the imposed requirements.

Meteorological Services

(E. H. Phinney and D. E. Jenne, Atmospheric Sciences Section)

Meteorological services, viz., weather forecasts and observations and climatological services were provided to plant operations and management on a routine basis.

SEPTEMBER 1967

Forecasting

<u>Type</u>	<u>No. Made</u>	<u>% Reliability</u>
Production	90	85.1
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