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TECHNICAL INFO

FOR FEBRUARY 1968

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

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Classified by R. J. Sorenson

C-65, Plutonium Production
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PACIFIC NORTHWEST LABORATORY
Classification Cancelled and Changed To
MONTHLY ACTIVITIES REPORT

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FEBRUARY 1968

By Authority of CG PR-2 (PR 24),

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and

Hanford Plant Assistance

Programs

by the

Staff of Battelle-Northwest

F. W. Albaugh, Director

March 1968

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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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

Measurements of radioactivity in 277 children were completed at the fourth Pasco Elementary School on February 16. Whole-body counting of children at the fifth elementary school began on February 21, following lectures to 12 classrooms in grades one through six.

Analysis of pheasants, quail and chukar shot locally revealed that only birds collected in the vicinity of the Columbia River contained detectable p^{32} and Zn^{65} . In decreasing order of concentrations the three species ranked chukar > pheasant > quail. Heads of the birds consistently contained more of these two radionuclides than the edible flesh by factors varying up to 20.

An ca 5% sample of local hunters was drawn from Washington State Game Department records in February. A questionnaire designed to gather data on hunting and consumption of game birds by local residents will be mailed to these 657 people in March. The results will aid in defining the importance of game-bird consumption to local environmental radiation doses.

Diet distributions of Richland and Benton City elementary school pupils were computed this month. Consumptions of various foods were determined for various age groups of boys and of girls. Annual environmental Radiation doses to Hanford employees residing in the Tri-Cities for the year 1966 were re-computed during February. Doses for whole-body, GI tract, thyroid and

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DIRECT AEC SPONSORED PROGRAMS

Columbia River Studies

Effects of Nuclear Effluent on the Quality of the Columbia River Water

(J. F. Honstead, Environmental Studies Section)

A study was started to adapt the existing digital simulation model for emergency application. A simultaneous collection of samples of effluent from all operating production reactors was performed in order to define the total curie input of Na^{24} . A multichannel analyzer with a 5 in. x 4 in. NaI crystal was installed at the Automatic Columbia River Monitoring Station (ACRMS) in order to determine the hourly concentration of Na^{24} at that point over a period of several days. Two sample traverses were conducted, one above the 300 Area and one at the Richland ferry landing in order to relate the concentration of Na^{24} at the ACRMS to the average concentration found in the river. The hourly data from the ACRMS will be adjusted to reflect average hourly river concentrations.

Mechanisms of Environmental Exposure

(J. F. Honstead, Environmental Studies Section)

Measurements of radioactivity in 277 school children in grades one through six at the Longfellow Elementary School, Pasco, Washington, were completed on February 16. Lectures describing the whole-body counting program were given to 12 classrooms at Robert Frost Elementary School. Whole-body counting began at this school on February 21.

Dietary distributions were computed for Richland and Benton City elementary school children from the data accumulated on the master file from the whole-body counting program. Calculations of intakes of various foodstuffs were made for boys and girls separately for each year of age from 6 through 14. Some 3,000 total cases are included in the distribution. The results of these distribution calculations are being prepared for a formal report.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Summary

Mission 1

Mark IV tube-in-tube end-spider supported fuel assemblies are being prepared for flow loop testing. The test will provide pressure drop, inner component motion (frequency, amplitude), and end-spider performance data.

Nickel-silicon deposits ca 2 mils thick on uranium metal showed satisfactory porosity after being heated to 600°C. Thinner deposits, ca 1 mil, were prone to blistering and rupture on heating.

The continuous recirculating purification technique for the molten salt Zr plating bath was shown to be workable. A leak resulted in catastrophic corrosion and loss of the apparatus. No further effort is planned.

The uranium enrichment tester at receiving inspection has been restored to stable operating condition. Addition of a spectrum stabilizer to the system was required.

Radiography and other nondestructive testing services were furnished to N-Reactor in support of various modification and maintenance projects in the steam generator cells.

Mission 2

N-Reactor fuel failure #42 was examined and determined to be similar in nature to previous failures #39, 40 and 41. The cause of failure was enhanced, localized clad corrosion resulting from impaired heat transfer at the locked support of the inner component.

Driver elements adjacent to fuel failure #40 have been examined and it is concluded that impaired heat transfer at the locked support existed over a span of at least 4 assemblies

is on hand but modifications will have to be made to existing foils for these measurements. Design of templates for the existing PCTR water tank and design of a mockup assembly for structural tests are in progress.

Preliminary design work was started on a 3/1 highly enriched uranium separate LiAl target PCTR experiment. The design is based on the use of materials already on hand which include:

1. 7.5 in. K lattice graphite for a 6x6 array of cells;
2. Standard Zircaloy process tubes for K lattice;
3. Standard size LiAl targets, 2.93 wt% Li (42.8% Li⁶).

The experiment will be performed on a 6x6 array since this yields a square shaped center cell with the correct fuel cell to target cell surface ratio (3/1).

The experimental procedures and measurements necessary to determine the infinite neutron multiplication factor of the CnLE overbore fuel element in the PCTR were completed. These experimental measurements included variations due to four different degrees of flooding conditions from the dry cell through maximum flooding in process tubes, hole bar, and moderator gas vents.

Mission 8

The "B" Unit is again in working order. Gain in the "A" Unit vertical channel is still a problem. A survey of normal operating background vibrations at 105-B was made.

Gas permeation from a crib into which water from a reactor loss-of-coolant accident would be disposed was studied. Streamline and equipotential surfaces were determined preparatory to solving for time displacement of gas into the soil adjacent to the crib.

Constant level balloons are now operational and trajectory studies using them will be initiated in the immediate future.

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Corrosion observed on nozzles removed from Tube 2455 in October 1967 is similar to that observed on nozzles removed in May 1966.

Corrosion penetrations on samples exposed fifteen months in the secondary system condensate loop ranged up to 0.2 mil. Stress-corrosion cracking was not observed on stressed samples but extensive pitting up to 5 mils deep was found on carbon steel samples.

Potentiostatic tests confirmed that the mechanism responsible for reducing galvanic corrosion currents in laboratory tests between K Area diesel engine powered emergency coolant pump components was cathodic polarization due to oxygen removal.

Long-chain organic polymer additions are under investigation as a method of reducing tuberculation on carbon steel in filtered Columbia River water.

Water levels in wells close to the 100-BC trench disposal site showed no appreciable increases during the month; more distant wells continue to show slight increases in elevation and, for the first time, in temperature. No changes in radioisotope concentrations in riverbank springs were noted that could be directly attributable to the disposal test.

X-ray diffraction results on sediments near the Columbia River showed that the sediments could be used in simulated Columbia River water turbidity experiments. Preliminary results of nine experiments on turbidity-alum requirements and associated zeta potentials were related by a least squares fit of the data.

Detailed neutron calibration measurements commenced on the portable solid state prototype rem meter developed for application in radiation protection monitoring in the reactor areas.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Mission 1 - Basic Production Mission

End-Spider Support Development

(J. W. Goffard, Metal Fuels Unit)

A charge of Mark IV tube-in-tube N fuel assemblies with end-spider supports, replacing the inner component supports and locking hardware, is being prepared for ex-reactor flow loop testing. Based upon earlier work on this concept, the design parameters to be explored are the fatigue behavior of the Zircaloy-2 spiders and the projection welds under dynamic simulated operating conditions. Pressure drops across the column of test assemblies will also be measured for assembly orientations that are expected to produce the maximum and minimum resistance. An instrumented end-spider supported assembly will be included in the test charge so that the nature (frequency and amplitude) of the flow induced motions of the spider supported inner component can be determined.

N Fuel Measurements

(R. K. Marshall, Metal Fuels Unit)

One hundred twenty-eight fuel elements of the 23 in. size were measured and weighed.

Zr Plating of Uranium

(R. W. Stromatt, Surface and Electrochemistry Unit)

The continuous recirculating purification technique for the molten salt electrolyte was shown to be feasible from an operational point of view. It was demonstrated that the melt could be pumped from the electrolysis cell to the HF sparge cell using a gas lift. From there the melt flowed under gravity over an overflow to an Ar sparge cell and by the same process back to the electrolysis cell. There was no difficulty encountered in controlling the flow or in sparging the cells.

Weld Radiography

(T. J. Davis, NDT Systems Engineering)

Repair and modifications to the injection water system in cell 2 of N-Reactor required radiography of carbon steel pipe welds. A 4 to 2 inch, schedule 160 reducer required repairs in the four inch OD weld after two successive radiographic examinations and was accepted after the third examination. The delay occasioned by the repairs made it necessary that installation be completed at the next outage. Sixty end cap welds for flow splitter fuel elements and a Graylock fitting were also radiographed for N-Reactor and were of acceptable quality.

N-Reactor Simulation

(R. D. Benham, Systems Analysis and Simulation Unit)

Analog runs were made with the overall N-Reactor simulation model for further development and testing of simulated reactor operator models. The operator model was put into the system to perform the function of manual take-over of the pressurizer pressure control system in the event of failure during a severe turbine trip transient. The operator's action, in this case, is necessary to prevent a reactor scram from excessive pressurizer pressure by opening the backup and cold spray valves.

Digital computer display and readout routines are being written for use with a multi-node reactor core model of N-Reactor. A polynomial fit of ten flux level values programmed on the PDP-7 gave an excellent display of the flux profile along the core.

Radiation Detection and Measurement Instrumentation

(E. M. Sheen, Instrumentation Section)

Delivery of the prototype rem ratemeter and battery charger to the External Dose Evaluation Unit for calibration tests completes the initial developmental effort by the Instrumentation Section on the project. Performance evaluation will be conducted following specific calibration procedures to determine the future utility of such an instrument.

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Mission 2 - Coproduct Mission

N Reactor Fuel Failure Analyses

(J. W. Goffard, Metal Fuels Unit)

N-Reactor failure #42 occurred on January 2, 1968, to a 2.1% enriched co-product driver element occupying position #4 in tube 0750. This element had attained about 3300 MWd/T exposure, had experienced the 4800 MW reactor power demonstration of last summer and, at rupture, the reactor was operating at about 3/4 of its design level of 4,000 MW. cursory examination in N-basin indicated that the failure was very similar in many respects to the previous three failures.

Radiometallurgical study revealed that coolant leakage occurred through a highly localized corrosion penetration of the inner clad at the downstream side of the crevice between the locked solid target support and the fuel clad. Fuel corrosion had undercut the fuel-clad bond and the increased volume of the corroded fuel had collapsed the inner clad against the target and had buckled the outer clad and fuel outward to fracture. The fuel side of the clad at the corrosion penetration displayed a 1/16 in. diameter white spot situated at the apex of a 3/8 in. diameter dimple (blister on coolant side). The coolant side of the failure site was mashed by the solid target support and the apparent site of the coolant entry was a small white spot. The area around the failure site was not heavily filmed or crudded. The solid target supports were recovered from the failed end without damage and they appeared to be normal and "as manufactured"--i.e., without evidence of surface grinding to accommodate the assembly operation. There did appear to be a crud deposit, however, on the surface of the support that formed the crevice around the point of support contact. It has been concluded that this failure was caused by the same mechanism responsible for previous failures #39, 40 and 41. High temperatures and other environmental conditions in crevices on heat transfer surfaces have enhanced clad corrosion to the extent that complete penetration to the fuel has resulted.

of at least four elements or 8 1/2 feet involving positions #2 to #6.

Mission 4 - Plutonium-238

NpO₂ Target Elements

(R. J. Anicetti, Fuel Process Development Unit)

Target elements for the irradiation of NpO₂ are fabricated by homogeneously mixing NpO₂ with an inert, ceramic or metallic "carrier" material. An investigation is under way to determine the acceptability of carrier materials which have a low neutron absorption cross-section, have acceptable reactor physics characteristics, and are amenable to separations chemistry and are resistant to water at 150°C and 170 psi. Low material and fabrication costs are also an important consideration.

Previous target elements employed graphite or aluminum as the carrier but, because of undesirable separations chemistry (graphite), and neutronic considerations (aluminum), a study of alternative candidate carriers was started.

Tests on lead monoxide (PbO) compacts exhibited corrosion resistance to water at 150°C and 170 psi. However, due to the low thermal conductivity of PbO, other materials are being investigated.

Both cold-pressed (97% TD) and sintered (450°C, 97.5% TD) magnesium compacts were resistant to water under the test conditions (150°C, 170 psi). This appears to be the most promising candidate at this time; however, acceptance will depend on the corrosion rate which will be determined by tests now in progress.

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GAMBLE-V - K. B. Stewart

The computer program GAMBLE-V, a fast running, 2D diffusion code, permitting upscattering, has been imported from Gulf General Atomics and is being adopted for use on the CSC Univac 1108.

Mission 7 - Target Space Enhancement

Aluminum Alloys

(R. R. Studer and W. E. Gurwell, Fabrication Metallurgy Unit)

All of the alloys under consideration as possible candidates for cladding resistant to ledge and groove corrosion have been delivered. A total of 17 alloys were produced in two forms, namely, extruded tube and rolled corrosion coupons. These alloys include the Al-Ni-Fe alloys, a high silicon age-hardenable alloy and a high Ni-high Fe aluminum powder metallurgical alloy. The casting alloys were made by direct chill casting a billet, pre-extrusion to break up the structure, then final extrusion into thin wall tubes and sheet bar. The sheet bar was hot rolled at 400°C to a thickness of 0.063 in. thick, then water quenched. All extruded tube was heat treated at 400°C and water quenched to assure uniformity of product. Some defects were noted on the interior of some tubes. These defects may have been due to either a breakdown in lubricant or else resulted from the slight taper that was present on the mandrel. The powder metallurgical alloy was fabricated from atomized powder by hot pressing in evacuated cans, then machining off the can and extruding into tubes and sheet bar stock.

Duplex Aluminum Alloys

(R. R. Studer and W. E. Gurwell, Fabrication Metallurgy Unit)

A total of 60 feet of tubing and 10 feet of sheet bar were delivered for evaluating the performance of the duplex alloys in Hot Die Sizing Tests and Corrosion Tests. The duplex alloy concept is to clad the functional material with a layer of anodic metal, in this case a layer of X8001 is coextruded over an AlSi tube. All compounds were direct chill cast, pre-extruded,

20-fold depleted uranium, 93% enriched uranium in aluminum, and 4% dysprosium in aluminum.

Lattice parameters to be measured are the infinite medium neutron multiplication factor (k_{∞}), thermal disadvantage factor, thermal utilization, resonance escape probability, and fast fission factor. The measured concentration of boron in the moderator required to reduce k_{∞} to unity will be reported as it is useful in criticality safety studies. Both the null reactivity technique (with boric acid in the water as a neutron poison) and measurements of the adjoint weighted excess neutron production cross section will be used to determine k_{∞} .

Since the maximum weight of fuel to be contained in the water tank is as much as eight times the previous maximum amount, a mockup is being made to test the structural integrity of the lattice plate structure and water tank assembly before loading into the PCTR. Planned fuel loading procedures will also be tested in the mockup.

Highly Enriched Uranium - Separate Target PCTR Experiments

(D. F. Newman and D. R. Oden, Heavy Moderator Reactor Physics Unit)

Preliminary design work was started on a PCTR experiment utilizing highly enriched uranium in aluminum fuel with separate targets of 2.93 wt% lithium (42.8% Li^6) in aluminum, in a three-to-one supercell. The design is based on the use of materials already on hand which include:

1. A 6x6 test cell array of 7.5 in. pitch K-lattice graphite,
2. 36 standard Zircaloy K-lattice process tubes, and
3. 9 columns of standard size lithium in aluminum targets (1.502 in. target core OD).

The experiment will be performed on a 6x6 test array since this geometry lends itself to the use of a square central supercell and has the correct fuel cell-to-target cell surface ratio (3/1). The redistribution of the flux in the graphite near the cell cavity when the central supercell is removed is minimized by the use of the square central supercell.

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MASS OF COPPER NECESSARY TO ACHIEVE UNIT MULTIPLICATION
(PCTR EXPERIMENT WITH CMLE OVERBORE FUEL)

<u>Lattice Condition</u>	<u>Mass of Copper (Mo)</u> <u>(grams)</u>
Normal cell with coolant	- 53.35 ± 3.27
Flooded process tube channel (simulated with polyethylene wrap around process tube)	- 64.31 ± 3.06
Flooded process tube channel and gas vents (simulated with polyethylene wrap around process tube and Lucite strips in gas vents)	- 48.38 ± 2.93
Lattice without coolant	+366.06 ± 7.48

Mission 8 - Nuclear Safety

Response of Hanford Reactor Systems to Seismic Excitation
(F. R. Vollert, Engineering Materials & Mechanics Section)

A new "B" Unit synchronous recorder motor was installed by a technician from the Hathaway Instrument Co., manufacturer of the recorder. Also, at the same time the Hathaway technician installed some electronics modifications designed to make the recorder run cooler.

Before B-Reactor was put on standby, the "B" Unit was used to monitor or survey the normal operating background vibrations at the 105-B Reactor Building. Data from the locations surveyed will be analyzed with the power density spectra program CALSPEC in an attempt to determine the natural periods (T) of certain B-Reactor components.

A minor circuit change designed to eliminate the drift or gain in the vertical channels of the "A" and "B" Units due to normal changes in room temperature was made in both units. The

Initially unirradiated fuel elements will be used to obtain metal-water reaction rates for autoclaved Zircaloy-2 surfaces and any disparities in the rate curves due to distortions of the fuel and cladding during the prolonged heating cycle. Temperatures of 1000, 1100 and 1200°C will be used to obtain rate equations as a function of temperature.

Release and Control of Fission Gases Under Reactor Accident Conditions

(A. K. Postma and L. C. Schwendiman, Gaseous and Particulate Waste Research Unit)

A study was started to describe noble gas permeation from a crib typical of one which might be used for disposing of water following a reactor accident. Gases initially contained in the water are conservatively assumed to be released into the void of the crib and then moved out through the soil as the level of water raises in the crib. Streamlines and the equipotential surfaces in the soil were established. Penetration of the gas along the flow lines as a function of time is yet to be determined.

A second paper study is being carried out to estimate the fraction of noble gases which will be dissolved in water in the proposed quench basin. This estimate will be used to calculate the amount of gas expected to enter the crib with the water.

100-N Atmospheric Study

(H. G. Daubek, Theoretical Meteorology Unit)

Preparation for constant level balloon studies continued. When the temperature of the weigh-off shelter is within a degree of the temperature desired, the balloons will fly in the desired height range. Under these conditions, we are prepared to make tests in the 100-N Area.

It is not always possible to have the shelter within 1° of the desired ambient air. When the temperature of the shelter is on the order of 10° from ambient, the balancing of the balloon is much more difficult. It is presently the objective to decrease the rate of rise now created by this temperature difference.

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Water Treatment Variables

(W. B. Silker, Radiological Chemistry Unit)

"Cat Floc", a positively charged polyelectrolyte, is now being used as the coagulant in place of alum in the Water Treatment Pilot Plant. The main objective in this test is to eliminate the aluminum that passes through the filter. It has been theorized that the residual aluminum may furnish the bulk of the material that forms the film on in-reactor surfaces, and as such, furnished impurities either through codeposition or exchange. The initial experiment involved "Cat Floc" addition at a rate (ca 0.5 ppm) sufficient to maintain neutral surface potential. At first, Separan was added as a filter aid, but high filter effluent turbidities prompted elimination of this additive. Even so, the filter effluent turbidities remained higher than the normal 0.05 Jackson unit (J.U.). It increased after backwash to about 0.4 J.U. and decreased through the run to 0.1 J.U., probably due to the formation of a "schmutzdecke."

Mission 10 - Columbia River

Effluent Control Program

(J. R. Divine, Reactor Water Chemistry Unit)

In previous reports we have discussed the use of the analog and digital computer in developing our model of the kinetics of activity buildup on reactor process tubes, fuel elements and in the coolant. In the initial work with the analog computer, we found that the coefficients relating to the deposition and release of material from the surfaces were interrelated.

The relationship obtained is

$$B = aA + b$$

where A and B are the release and deposition coefficients and a and b are assumed to be constant. Furthermore,

$$a = \frac{m}{k_e}$$

Using the above data, while realizing that some of the numbers are of more qualitative than quantitative significance, the values for k_e , fk_i and λ_o were calculated. Additional assumptions made were that fk_i and λ_o are the same for both tubes for a given species and that $1 - V = 0.02$. The values calculated are:

Term	Aluminum Tube		Zircaloy Tube	
	Species		Species	
	(1)	(2)	(1)	(2)
k_e (cm ⁻¹)	0.2	1.9×10^{10} to 0.4	0.006	0.18
fk_i (cm/sec)	0.15	0.08	0.15	0.08
λ_o (hr ⁻¹)	0.05	0	0.05	0

All of the values calculated for k_e are much higher than the value of 0.0003 cm⁻¹ obtained from an independent experiment for aluminum. Also, the k_e data for the Zircaloy tube are smaller than those for the aluminum tube which is contrary to expectation.

The kinetic deposition factors, fk_i , are of the order of calculated values. The calculated factors were obtained using the assumption that the particles were of the order of 0.2 μ in diameter.

No evaluation of the λ_o values will be made at this time owing to insufficient data.

Process Technology

Low Temperature Zircaloy Hydriding

(W. K. Winegardner, Corrosion Research Unit)

Investigation of the effects of continued exposure on existing zirconium hydride distributions in production reactor process tubes continued. This investigation currently consists of the comparison of the hydrogen content of selected process tubes with base-line data obtained from the analysis of eight tubes that had been installed in-reactor for 42 to 44 months. Attempts

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were located were usually similar to those found in the upstream portions of the tubes, i.e., no noticeable hydrogen pickup.

Corrosiveness of Phosphoric Acid Decontamination Solution
(A. P. Larrick, Corrosion Research Unit)

A large quantity of Turco 4512A (inhibited phosphoric acid) was left over from the October 1967 full-reactor decontamination of N Reactor. A laboratory test was conducted to determine if the corrosiveness of the solution was changing during storage because of inhibitor degradation, iron pickup, etc. The visual appearance of the solution when received on September 21, 1967, was clear and colorless; when it was sampled on January 26, 1968, it was light amber colored. Corrosion penetrations calculated from weight losses after a one-hour exposure to a 10% solution at 71°C were 0.0069 mil on A245 carbon steel, 0.015 mil on A212 carbon steel welded to 304 stainless steel, and 0.0014 mil on 304 stainless steel. These losses are comparable with losses found in the fresh solution, indicating that the solution corrosiveness is not rapidly changing. Pitting up to 5 to 10 mils wide and 1 to 2 mils deep on weld samples exposed eight hours to the aged solution was slightly greater than on the samples exposed to the fresh solution.

In-Reactor Corrosion Monitoring
(A. P. Larrick, Corrosion Research Unit)

Monitoring corrosion coupons in the N Reactor primary and secondary coolant bypass test sections has begun. Coupons are being fabricated from nineteen alloys in stock and material for fabricating samples from ten other alloys is on order.

Corrosion of Reactor Nozzles
(A. P. Larrick, Corrosion Research Unit)

The front and rear steel nozzles removed from Tube 2455 in October 1967 are being examined for corrosion. The nozzles were removed shortly after the full-reactor decontamination; therefore, no oxides were present. Numerous pits were present in patterns similar to those observed on nozzles removed in May 1966.

Carbon Steel Corrosion in Filtered Columbia River Water
(A. P. Larrick, Corrosion Research Unit)

Catalyzed sodium sulfite has been found to reduce pitting and tuberculation on carbon steel exposed to filtered Columbia River water but there are some inherent drawbacks in the use of this material. Alternate methods of reducing pitting corrosion or tuberculation are being investigated. Currently, a long chain polymer, Betz DE-287B, is under investigation in the TF-21 loop. Betz DE-287B is normally used as a sludge dispersant but it is also reported to reduce tuberculation better than phosphate-containing materials. Results are inconclusive after two weeks' exposure because tubercule formation has not occurred on either the test samples or the control samples exposed to untreated filtered water. Metal weight losses were somewhat lower on the control samples.

Water Filtration Studies
(D. E. Olesen, Water and Waste Management Unit)

Approximately 15 gallons of Columbia River water was passed through a continuous high-speed (25,000 rpm) centrifuge to remove particles down to 0.1 μ which produce turbidity. This material was compared by X-ray diffraction with sediment samples collected from the river bank at Richland. The analysis showed the materials to be similar so that the river bank sediment can be used to produce turbid water for the pilot plant experiments.

Using the water filtration pilot plant, experiments were conducted to determine alum requirements at various turbidity levels and the associated zeta potential. A least squares fit of the data yielded the following equation:

$$y = 10.22 + 0.725X$$

x = turbidity, JTU
y = Alum Conc'n, mg/l for 0 zeta potential

These data are based on 9 experiments at turbidity levels from 1.6 to 17 JTU. They are preliminary and will require additional information before being fully accepted because of the

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Mission 1

The (G_T) value for radiolytic production of gases in plutonium nitrate solutions was reduced by the presence of oxygen. A negative $G(O_2)$ value was observed when n-butanol was present in the plutonium nitrate solution. A Deoxo catalyst for recombination of H_2 and O_2 over plutonium nitrate solution apparently became poisoned by water vapor after about three weeks.

Indications are that reaction of aci-form 1-nitropropane with HNO_2 and HNO_3 may produce more than one material which accelerates oxidation of $Np(V)$. Emphasis has been given to defining conditions which produce a single product of low extractability and sensitivity to destruction by radiation.

Further work continues to show that the proposed batch contact process for recovering Np from Purex acidified sludge by DBBP extraction will recover greater than 90% of the Np present along with about 75% of the Pu .

Recent experimental work has involved the testing of different sizes of electrophoresis apparatus and different applied voltages to the migration rates of $U(VI)$ and $Nd(III)$, the search for mixed solvents which will move inorganic ions in thin layer chromatography, and the study of thin layer shapes to facilitate separations.

Techniques are being sought for the separation, concentration, and determination of Np and Pu in Purex waste. The ultimate goal is to develop a remote, automatic system of instrumentation to accomplish these tasks. Methods investigated included coprecipitation, solvent extraction, ion exchange-, reverse phase-, and thin layer chromatography, electrophoresis and electrodialysis, atomic absorption spectroscopy, gamma spectrometry and X-ray spectrometry using high-resolution energy dispersion.

phase was small, 73 ppm, whereas their total concentration in the NaOH phase was about 500 ppm. The nickel content of the bismuth phase was 10 to 20 ppm. The estimated maximum permissible concentration is 100 ppm. Essentially the same results were obtained with 304L as the containment material.

A study was started to determine the impact of wastes which will be derived from processing enriched fuels in the Purex plant on the Hanford Waste Management Program.

Mission 8

Preliminary analyses of experimental data from water-reflected slabs of 195 g Pu/l plutonium solution having 23.3 wt% Pu²⁴⁰ isotopic concentration indicate the critical heights of 8.0, 7.3, and 6.8 in. slabs to be 12.9, 15.3, and 18.1 in., respectively. The difficulty encountered in analyzing the experiments with dilute plutonium solutions has been partially resolved and satisfactory agreement between calculation and experiment is now being obtained for the reflected slabs. Discrepancies still exist for the bare systems and are being investigated.

Four boiling regimes were characterized and initial experiments conducted to measure the release of plutonium aerosols from a boiling solution containing plutonium nitrate.

Work was started on obtaining first-order levelling and triangulation data to be evaluated for possible changes (and trends) caused by local earth movement.

Mission 9

Inconsistencies were noted in the calculation of permeability distribution using the one and the two dimensional versions of the STREAM program. Correction of the problem may result in calculated travel times showing closer agreement with field-measured travel times.

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A study was started to determine the cause of spontaneous burning of nitric-acid-leached Zircaloy cladding observed at the Nuclear Fuels Services plant during processing of Hanford N-Reactor fuels. The presence of pyrophoric epsilon phase U-Zr intermetallic compounds on the leached cladding might account for the observed burning. To date, no evidence for this pyrophoric phase has been found in laboratory-scale leaching studies with nonirradiated N-Reactor fuel pieces.

Miniature mixer settler studies testing proposed Purex process second and third plutonium cycles are in progress. The proposed flowsheets, if successful, could permit replacement of the present plutonium ion exchange cycle with an all-solvent-extraction system allowing higher plutonium processing capacity.

Modifications and repairs to the 325-A Bldg. C-Cell have been completed and processing of irradiated neptunium has been resumed. One batch of N-Reactor elements was processed and a second batch dissolved.

No separation of europium from americium was obtained in a laboratory-scale test involving eluting a solution containing these elements through an ion exchange column with NTA as the elutant and zinc ion as barrier.

Techniques are being developed for separating zinc from americium and americium from curium by precipitation. The procedures are needed for use in recovery of desired constituents from impure fractions left from the Am-Cm recovery operations conducted in the 325A cells.

A sampler was devised to take an average sample of the non-condensable gases over a long period. Collection of the iodine and organic iodides will be by liquid nitrogen trap. Traps for removal of water and NO₂ before the liquid nitrogen trap are being developed and tested for possible associated iodine loss.

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Mission 1 - Basic Production Mission

Radiolytic Gas Generation in Plutonium Nitrate Solutions
(J. C. Sheppard, Separations Chemistry Unit)

The investigation of the radiolytic gas generation from plutonium solutions was continued. The hydrogen radical scavenging potential of oxygen was determined. A G_T value of 0.07 was found for an oxygen saturated, 4.0M HNO_3 solution, compared to the air saturated value of 0.10. These results are consistent with those found for the argon experiment and suggest that the reactions

play a small but significant role in the radiolytic generation of hydrogen and oxygen. More work would have to be done to quantitatively explain the results.

The gas generated from the radiolysis of the 0.5% n-butanol in the 4.5M HNO_3 -plutonium solution was analyzed. $G(\text{H}_2)$ and $G(\text{N}_2)$ were in agreement with other 4.5M HNO_3 results, while $G(\text{O}_2)$ was actually negative, indicating the radiolytic or chemical consumption of oxygen. A possible series of reactions involves the reduction of plutonium(IV) to plutonium(III) by radiolytic or chemically produced butylaldehyde, followed by the reoxidation of the plutonium(III) to plutonium(IV) with oxygen. Carbon dioxide was also found. Radiolysis or chemical degradation of the chemically produced butyric acid probably is the source of carbon dioxide.

The experiment in which radiolytically generated hydrogen and oxygen were recombined to form water using a Deoxo catalyst was terminated. It was found that the pressure over the 4.5M HNO_3 -plutonium(IV) solution could be maintained constant for 26 days. After this the pressure started to build up, indicating

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The extractability of accelerating material was studied in two solutions obtained by allowing the product of low temperature runs to stand before being diluted to quench the reaction. In one case a single equal-volume contact resulted in the extraction of no accelerating material (within the uncertainty of the measurement) and in another case three equal-volume contacts resulted in the extraction of 25% of the accelerating material from 3.1M HNO_3 , using pre-equilibrated organic.

The radiation stability was also investigated in the two solutions used. In the first case, a dose of 10^5 rad to a 3.1M HNO_3 solution containing 0.0035M RNO_2 (by make-up) resulted in destruction of about 20% of the accelerating material. In the second case, the 10^5 rad dose destroyed all the accelerating material in a 3.1M HNO_3 solution containing 0.0013M RNO_2 and 35% of it in a 0.0082M RNO_2 solution.

DBBP Extraction of Np from PAS Solutions
(W. W. Schulz, Separations Chemistry Unit)

Experiments were continued to develop a satisfactory process for batch DBBP extraction of neptunium from Purex Acidified Sludge (PAS) solutions. The process currently visualized involves successive batch contact (in a tank farm vault) of three portions of PAS with one batch of 25% DBBP-NPH to extract at least 90% of the neptunium and as much of the plutonium as feasible. Subsequently, the loaded organic phase would be routed to the Purex plant for oxalic acid stripping of the actinides and the solvent, after carbonate washing, would be returned to the tank farm vault. This process thus couples closely with the DBBP extraction process proposed for use in the Purex plant to recover neptunium from LWW solution.

Experiments performed with actual PAS solutions demonstrated the feasibility of the DBBP neptunium recovery scheme. PAS solutions were obtained by HNO_3 leaching of water-leached sludge from Tanks 241-104-A and 241-101-A. Portions of each PAS solution were adjusted to 0.5M HNO_3 and 0.05M ferrous sulfamate and allowed to stand 30 min at 25°C. Subsequently, in each case, a 25% DBBP-NPH solvent was contacted (10 min,

immediate goal is to develop laboratory techniques that can accomplish the above steps and also be the basis for the automatic instrumentation method.

A consideration of all the instrumental methods currently developed for trace analysis, and of the size, cost and arrangement of these instruments, makes a simple radioactive counting technique seem most desirable for the final determination. The method is among the most sensitive and it will require very little apparatus with no moving parts to be located remotely at the sampling station. Lithium-drifted silicon diode detectors have energy resolutions of less than 1 keV. These diodes cannot be used with high energy gamma radiation but will measure the low energy X rays produced by Np and Pu isotopes, provided these X rays are sufficiently intense to count in a reasonable time period. Other possible methods will be considered and will be used in the laboratory experiments to evaluate separation and concentration techniques.

A second objective of the study is to provide automatic monitoring of plutonium losses in Purex HAW waste. The technique developed for Np may, with only a little additional effort, be made applicable to the simultaneous determination of plutonium.

Electrophoresis and Chromatography

(M. C. Lambert, Chemical Sensors and Instrumentation Unit)

Several electrophoresis tests have been run in order to try out three different sizes of apparatus, different lengths of strips, and different voltages. With uranium and neodymium (and all inorganic ions in general) high voltages are desirable to accomplish significant ion mobility. The power supply on hand will supply a maximum of 500 v at 150 ma. However, this may be sufficient for the short thin layer "chromagram" strips to be used.

Several mixed solvents have been tested with U, Ag, La and Nd ions and thin layer "chromograms". Some solvents reacted somewhat with the cellulose nitrate backing, but fiber glass

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Electrodeless Conductivity

(O. H. Koski, Chemical Sensors & Instrumentation Unit)

Temperature compensation of the pulse type electrodeless conductivity system being developed for HAW acidity measurement was studied. It was determined that attenuation of the drive voltage by a temperature sensitive resistive network at the probe is feasible. This method is of advantage due to the restriction on the available conductors through the cell wall.

Range selection can be accomplished by drive voltage or amplifier gain adjustment. Output linearity has been found to be impaired at solution loop resistance less than 10 ohms. The nominal range of the instrument is thus from 10 ohms to 800 ohms solution loop resistance.

Thermodynamics

(J. D. Christian, Physical and Theoretical Chemistry Unit)

Present results on the measurement of the vapor pressure of $\text{UCl}_3(\text{c})$ by a transpiration method may be fitted to the equation

$$\log p_{\text{torr}} = - \frac{9212}{T} + 7.415.$$

The calculated values of $\Delta H_{\text{subl}}^{\circ}$ and $\Delta S_{\text{subl}}^{\circ}$ are compared in the table with values obtained by previous workers.

Temp. °K	Investigator	Method	$\Delta H_{\text{g}}^{\circ}$, kcal mole ⁻¹	$\Delta S_{\text{g}}^{\circ}$, Eu	$\Delta S_{\text{v}}^{\circ}$, at b.p.
992	This work.	Transpiration	42.2	20.7	23.4±0.7
960	Sheckarev, et al (1956)	Knudsen effusion	51.0	27.5	29.8±1.8
1070	Altman (1943)	Knudsen effusion	54.9	32.6	36.7±2.7

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to 50 grams, in hoods and glove-box installations. Additionally, the improved discrimination provides a versatile instrument for investigation of new product areas in connection with possible Pu^{238} , Am^{244} and Np^{237} processing.

Mission 4 - Plutonium-238

Iodine Release During Target Dissolution (R. E. Lerch, Separations Chemistry Unit)

Additional hot cell experiments have been carried out to determine the relative amount and form of I^{131} released during target element dissolution and to determine the effectiveness of Hg^{++} for the control of the amount of iodine released during dissolution of various types of fuel elements. Four types of fuel elements have been investigated: 1) pressed graphite - neptunium oxide, 2) impregnated graphite - neptunium oxide, 3) $\text{UO}_2\text{-PuO}_2$, and 4) aluminum-neptunium oxide alloys. Organic iodides, varying from 1.5 to 30.5% of the total amount of released iodine, were released for all four types of fuel elements. The results of these experiments suggest the following:

1. The presence of Hg^{++} (0.005M) in the leach solution (or in the acid used to acidify the products of caustic dissolution) is relatively ineffective in controlling the amount of I^{131} released. Results with mercury were not reproducible.
2. The formation of organic iodides probably takes place during the dissolution operation (as opposed to being formed during reactor irradiation).
3. The presence of nitrite ion in a caustic dissolution solution significantly reduces both the total amount of I^{131} released and the % released as organic iodide.

Only a small amount of I^{131} was released when nitrite ion was present in a caustic dissolution solution. Upon neutralization of the caustic with nitric acid, a 14-fold increase in the amount of iodine released was noted, and the % released as

Isotope Separation by Electromigration
(R. E. Lerch, Separations Chemistry Unit)

The laboratory work is finished on pulsed current migrations and a final report on all isotope separation work is being written.

Mission 6 - Enriched Fuels Reprocessing

Reprocessing of Uranium-Zirconium Alloy Fuels
(R. F. Maness, Corrosion Research Unit)

The corrosion resistance of 304L, 310L, HAPO-20, BNW-1, Corronel 230 and Tophet M Alloy was determined in HNO_3 -HF- $\text{Al}(\text{NO}_3)_3$ solutions suitable for the dissolution of Zr-2 cladding and U-Zr alloy fuel cores. Variables were temperature (65°C to boiling), nitric acid concentration (1 to 4M), fluoride ion concentration (1 to 3.5M), dissolved Zr(IV) concentration (0.0 to 0.5M), and Cr(VI) concentration (0.0 to 0.2M). Preliminary data indicate that corrosion would limit the use of 304L as a containment material to a maximum process temperature of about 70°C . Corrosion of 304L was inhibited in the acid fluoride systems used by the presence of Cr(VI). However, the effect is of limited value since Cr(VI) is reduced to Cr(III) as Zr-2 dissolution proceeds. Of the alloys investigated, only HAPO-20 appears to possess adequate corrosion resistance to suitable boiling acid fluoride solutions in the absence of Cr(VI). Corrosion rates for HAPO-20 are about 2.5 mils/mo in the solvent as mixed. However, promising results were obtained with 310L in the presence of Cr(VI). The corrosivity of the dissolvent solution decreases as the dissolved zirconium concentration increases due to a reduction in HF concentration. Rather careful process control will be necessary to maintain the F/Al/Zr ratio within solubility limits.

The dissolution rate of 0.5 in. OD, 0.035 in. wall, oxidized Zr-2 tubing (three day exposure to 400°C steam at 1500 psi) was determined in 1M HNO_3 -2MHF-1M $\text{Al}(\text{NO}_3)_3$ solution at 70°C and at the boiling point and in boiling 1MHNO₃-1MHF-1MAL(NO_3)₃ solution. An induction period of about six hours was observed with the boiling solution containing 1MHF and with the solution containing

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Mission 8 - Nuclear Safety

Atmospheric Release of Radioactive Particles

(L. C. Schwendiman, Particulate and Gaseous Waste Research Unit)

Four boiling regimes were characterized through observation, heater temperatures, and boil-off rate for experiments in which the airborne release of plutonium salts from such solutions are to be measured. First experiments were performed using plutonium nitrate for the two more vigorous boiling regimes. These two regimes have been characterized visually as "vigorous rolling boil" and "steady continuous breaking of the surface". The heater input and temperature at a reference point are used to assist in reproducing the conditions employed. Analytical results are not complete, but the indications are that a significant fraction of plutonium will be airborne under the conditions imposed.

Additional data were obtained for the release fraction upon heating plutonium nitrate residues at 400 and 1000°C. The release was less than 4×10^{-3} wt% per hour. Use of significantly lower analytical blanks obtained from those in earlier experiments may allow an even lower release rate to be reported. Blanks which include gloved box handling are being determined.

Seismic Evaluations

(D. D. Tillson and D. E. Peterson, Hydrogeology Unit)

Work was begun on obtaining historical first-order leveling and triangulation data that will be evaluated for possible changes (and trends) caused by earth movement. Discussions with U. S. Coast and Geodetic Survey personnel in Seattle revealed that this type of study has not been done previously to any degree in the Pacific Northwest. Similar studies done for other areas, notably California, have proved to be very useful in providing information on magnitudes and rates of earth movements over the last century.

M2EHPA removal is favored by low aqueous phase sodium concentrations while the tendency toward third phase formation is less at high sodium concentrations. Selection of an optimum wash composition and volume thus involves a compromise between these two opposing criteria.

Tests were also performed this month with a 0.3M D2EHPA-0.2M TBP-NPH solution containing 0.026M iron (III) to evaluate the effects of wash composition and volume on precipitation of $\text{Fe}_2\text{O}_3 \cdot x\text{H}_2\text{O}$. As expected, the concentration of tartrate required to solubilize the iron depends on volume ratios and on the NaOH concentration of the wash. Thus, only trace amounts of iron precipitated when the solvent was washed with an equal volume of 2.5M NaOH-0.25M sodium tartrate but large amounts precipitated at 0.15M (or lower) tartrate concentrations. Also, even 0.25M tartrate did not prevent precipitation of large amounts of the iron when the solvent was washed with one-half volume of 2.0 or 2.5M NaOH-0.25M sodium tartrate. For various reasons, solvent obtained in actual B-Plant processing of PAW may contain a different iron concentration than that of the solvent used in these studies. Repetition of these tests with plant solvent, when available, will be performed to firm up tartrate requirements.

Waste Packaging

(T. S. Soine, Materials Chemistry Unit)

The literature search to determine the most attractive strontium and cesium compounds for packaging was continued. The compounds investigated included:

SrF_2	SrOZrO_2	SrCO_3	SrSO_4
SrO	$\text{Sr}_3\text{P}_2\text{O}_8$	$\text{Sr}(\text{NbO}_3)_2$	SrB_6
SrTiO_3	$\text{Sr}_3\text{P}_6\text{O}_{18}$	SrOSiO_2	SrS
Sr_2TiO_4	SrHPO_4	$3\text{SrO} \cdot \text{Al}_2\text{O}_3$	SrMoO_4
CsCl	$\text{CsW}_x\text{Nb}_{1-x}\text{O}_3$	$2\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 9\text{SiO}_2 \cdot \text{H}_2\text{O}$	
Cs_2O	CsSO_4	$\text{Cs}_2\text{O} \cdot \text{B}_2\text{O}_3 \cdot 4 \cdot 2\text{SiO}_2$	
CsNbO_3	Cs_2TiO_3	$\text{Cs}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$	
CsTaO_3	$\text{Cs}_2\text{U}_2\text{O}_7$	$\text{Cs}_{0.32}\text{WO}_3$	

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Compound

Bases for Selection

Cs_2O	High Cs density, easy to prepare (may require an inert atmosphere), high water solubility and reactivity, possible losses due to volatility of Cs_2O .
CsNbO_3	Fair Cs density, low water solubility, processing somewhat difficult (high temperature fusions).

Burning H_2 - O_2 Mixture in Closed Containers
(W. E. Willingham, Process Engineering Unit)

Series "D" runs in the test H_2 - O_2 burning chamber were completed. Hydrogen-oxygen mixtures were ignited in a 4 ft long pipe six inches in diameter. In the D series, the pipe contained a 3 ft long packing of AW-500 zeolite supported by a wire screen five inches from the bottom of the pipe. Results were essentially the same as in the previous series of runs in which the zeolite bed depth was two feet. No detonation waves were observed; burning waves occurred in some cases.

Process Technology

Zirflex Decladding Process Improvement
(D. G. Carter, Separations Chemistry Unit)

Several preliminary experiments were conducted to determine the effect of pH (of the Zirflex dissolver solution) on the dissolution rate of uranium. $(\text{Et})_4\text{NOH}$ was used to increase the pH of the Zirflex dissolver solution (5.5M NH_4F - 0.5M NH_4NO_3) to pH 7.85. $(\text{Et})_4\text{NOH}$ concentration was ca 0.04M . The reaction vessel was a 3000 ml Teflon-coated stainless steel beaker fitted with a Teflon top connected to a glass water-jacketed condenser. About one liter of dissolver solution was used. The uranium was allowed to remain in contact with the boiling ($100 \pm 4^\circ\text{C}$) Zirflex dissolver solution for one and two hour periods. Preliminary results indicate dissolution rates (in flat portion of curve) of $1.2 - 1.9 \text{ mg/cm}^2 \text{ min}$. The average of these numbers represents a reduction in the rate of reaction on the order of 54%. Additional and more refined experiments must be conducted for reproducibility and accuracy and some attention

Initial dissolution tests with unirradiated N Reactor fuel pieces do not support the hypothesis that the fires are due to epsilon phase residues. In these tests, hemispherical, one-half inch thick wafers cut from both heat-treated and non-heat-treated N Reactor fuel elements were dissolved in boiling 7.5M HNO_3 to yield 2M HNO_3 -1M $\text{UO}_2(\text{NO}_3)_2$ solutions. (The "heat treatment" referred to above involves heating the coextruded U-Zr element to 730°C and then rapidly quenching into a low temperature salt bath.) At frequent intervals the test pieces were removed, allowed to dry, and touched with the spark from a Tesla coil to detonate or ignite any explosive residues. No such residues were observed in these tests or even when non-heat-treated material was anodically dissolved in a large volume of 1M HNO_3 at 30°C . The electrolytic dissolution technique was shown in the 1954 work to yield especially sensitive residues.

The present chemical evidence indicates that either our techniques are too insensitive to detect the epsilon phase in unirradiated fuel or that this phase is not present in such fuel. At least for heat treated material the latter conclusion appears more likely since the phase diagram for the U-Zr systems shows the epsilon phase does not exist at 730°C ; rapid quenching from 730°C would be expected to yield a material free of epsilon phase. Failure to detect chemically any epsilon phase material in non-heat-treated material is not understood because from metallurgical considerations some would be expected; however, direct metallurgical evidence for the existence of the epsilon phase in the bond layer between the uranium and Zircaloy can in non-heat-treated fuel apparently does not exist.

Metallurgists have expressed the opinion that N Reactor irradiation at temperatures around 400°C should produce epsilon phase material even in heat treated fuel. At month end such irradiated fuel was being procured to search further for combustible residues.

Purex Process 2nd and 3rd Pu Cycle Flowsheet Tests
(W. W. Schulz and D. G. Bouse, Separations Chemistry Unit)

ARHCO is considering replacing the present anion exchange

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have been a source of product contamination. In addition, the 6 in. column was repaired and all leaks stopped.

The first tube of K Reactor NpO_2 -Al elements was processed. This batch contained ca 320 g Np which had a reactor discharge date of September 26, 1967. Because of the long cooling time, no iodine problem was encountered. The processing went very well with no unusual incidents. The dissolution of the elements was surprisingly rapid. Some elements were completely dissolved in less than 20 min and no induction period was noted, i.e., the dissolution began almost immediately on insertion in the dissolver.

The second batch of elements was also dissolved and made ready for processing.

Plutonium-238 Accountability

(F. P. Roberts, Separations Chemistry Unit)

The resin used for all the Np-Pu processing through December 1967 had been stored in two waste cans in C cell. This resin was sampled prior to disposal and the Pu content determined. The 60 liters contained only 12g Pu^{238} and therefore does not represent a significant fraction of the material unaccounted for (MUF).

Processing Shippingport Fuels Am-Cm Ion Exchange End Cuts

(L. A. Bray, Separations Chemistry Unit)

After completion of the successful Shippingport Am-Cm separations programs in the 325-A ion exchange facility, several partially purified fractions remained for further processing. One of these contained 80 g Am^{241} , 0.01 g Cm^{244} and was highly contaminated with Eu^{154} . Because laboratory data were incomplete for NTA vs DTPA separation of Eu from Am, a short ion-exchange run was performed for the dual purpose of determining if Eu can be separated from Am by NTA elution and to convert most of the 80 g of americium to Eu-free product. The Am-Eu feed was loaded on H^+ -form resin and eluted with NTA to a second ion exchange column containing a zinc barrier. No separation

Waste Storage Tank Circulation Studies

(W. E. Willingham, Process Engineering Unit)

Instrumentation (thermistor probes and a Doppler Flow Meter) for measuring flow velocity in the 324 Bldg. model waste storage tank was received and installed. Efforts were directed to calibration and adjustment of this instrumentation.

Criticality Studies with Plutonium Nitrate Solution in Slab Geometry; Effect of Pu²⁴⁰ on Criticality*

(R. C. Lloyd and C. A. Rogers, Critical Mass Physics Section)

Criticality experiments were continued for determining critical thicknesses of "infinite" slabs of plutonium solution having a 23.3 wt% Pu²⁴⁰ isotopic concentration. The experimental vessel, a variable thickness slab tank with 42 in. dimensions, was water-reflected. Although the chemical analyses have not been completed, the solution contained about 195 g Pu/l at a nitric acid molarity of about 1.5.

Critical height measurements were made on slab thicknesses of 8.0, 7.3, and 6.8 in. Preliminary values for the critical solution height in each of these three experimental assemblies are 12.9, 15.3, and 18.1 in., respectively.

Theory and Analysis*

(R. C. Lloyd, L. E. Hansen, and R. D. Johnson, Critical Mass Physics Section)

As reported previously, the agreement between measurements and calculations for critical thickness of infinite slabs of dilute plutonium solution has not been satisfactory, even though the same calculational model (HFN code) reproduces the experimental data for these same solutions in spherical geometry quite satisfactorily. A thorough investigation of the experimental facility revealed a consistent inaccuracy in the slab

* Work also funded in part by Division of Reactor Development and Technology.

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and input preparation write-ups will be prepared following program completion and debugging.

A version of the two-dimensional STREAM program was modified to run on a one-dimensional fit of a streamline. If procedures using this modified program prove successful and applicable, a quite beneficial time-saving can be made by tailoring the whole program to one-dimensional analysis.

Organic Waste Disposal

(G. E. Benedict and V. L. Hammond, Advanced Chemical Engineering Unit)

Separation of carbon tetrachloride from TBP and DBBP was investigated experimentally. A simple batch approach was used in that the solvent was mixed with water and distilled through a 10-plate tower. The distillation was cut off when the tower outlet vapor temperature reached the boiling point of water. The degree of separation desired is 100 ppm of impurity in each component; chemical analysis is still in progress. The results of this preliminary distillation study will be used to determine the material of construction of the incinerator condenser. Additional work will be done to optimize the separation of the carbon tetrachloride diluent from the TBP and DBBP and to evaluate the feasibility of recycling the carbon tetrachloride to the process.

The solvent incinerator system, presently envisaged, will consist of a burner, a surface condenser, an impingement recirculating scrubber, an electrostatic precipitator and an absolute filter--all operating at negative pressure to prevent out-leakage of radioactive aerosols. Numerous manufacturers of incinerators for burning liquids were contacted and information was obtained on the availability and design characteristics of small incinerator systems.

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Evaporator Condensate

(B. W. Mercer, Water and Waste Management Unit)

Analytical results of an experiment with 242-T evaporator condensate and a 1 in. diameter by 15 in. column of 20 x 50 mesh AW 500 show that Cs¹³⁷ decontamination factors of 35 to greater than 100 were attained at a flow rate of 20 column volumes per hour. Since the evaporator condensate contained 5.2×10^{-4} μCi of Cs¹³⁷/ml a decontamination factor of 26 was sufficient to meet drinking water limits for Cs¹³⁷. It is of interest to note that the Cs¹³⁷ removal increased during the latter portion of the run as the AW 500 bed became plugged by the fine material that goes through the 5 μ Cuno filter. This indicates that some of the Cs¹³⁷ may be tied up on the fine suspended matter which is more effectively removed as the AW 500 bed becomes plugged.

The AW 500 bed received 5500 bed volumes of condensate before termination of the experiment because of plugging. Previous experiments with a small (8 cm³) AW 500 bed showed no Cs¹³⁷ breakthrough to 18,000 bed volumes. Since only a small Cs¹³⁷ decontamination factor is normally required, upflow operation of an AW 500 bed may be possible to avoid filtration. The extent of the Cs¹³⁷ sorbed on the suspended matter would have to be determined to establish the feasibility of this approach.

B-Plant Emergency Water Supply

(J. R. Raymond, Hydrogeology Unit)

Assistance was provided on the design and construction of an additional emergency water supply well for the B-plant fission product recovery project. Drilling samples were sieved and the well screen sized and specified. Procedures for a quantitative aquifer pumping test were outlined and results will be evaluated following well completion and testing.

Waste Tank Sludge Monitoring

(J. R. Raymond, Hydrogeology Unit)

Work was undertaken to determine the consistency of the sludge in the 116-TX tank. A drive-point connected to a string

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Similar experiments will continue; however, buffered systems at $\text{pH} > 8.2$ will be used to eliminate adsorption competition by soluble soil calcite.

Coating Formation on Graphite
(R. C. Giberson, Materials Compatibility Unit)

A sample of TSX graphite 3 in. long x 0.44 in. in diameter was coated by the MARQUARDT Company. Details on the coating are not known but it is thought to be silicon carbide. The coating thickness at one end, the center, and the other end of the sample was 0.0074, 0.0068 and 0.0063 in., respectively. The sample ends were not coated.

The as-received sample, number 1P, was placed in a recording thermobalance and oxidized at ca 920°C in 6,000 ppm of water vapor in helium. The temperature varied from 900°C at the gas inlet end of the sample to a maximum of 928°C at the center and 915°C at the gas outlet end of the sample.

The initial oxidation rate was 4.0×10^{-5} g/g-hour, increasing to ca 9×10^{-5} g/g-hour after 100 hours of oxidation. This rate can be compared with an uncoated sample rate of ca 1.8×10^{-3} g/g-hour for similar oxidation conditions. The rate remained in this range for an additional 50 hours of oxidation. The sample lost 1.1% of its original weight after 150 hours of oxidation. The weight-loss rate agreed well with the product formation, 65 ppm of CO and H_2 for the reaction $\text{C} + \text{H}_2\text{O} \rightarrow \text{CO} + \text{H}_2$. No CO_2 was detected during the oxidation.

During the first 3 hours of heating in the moist helium, the sample gained about 1 mg. A gas sample taken during the weight-gain period contained 8 ppm CO_2 , 31 ppm CO, and 75 ppm H_2 . This result is not presently understood.

After removal from the oxidation furnace, the color of the sample was red at the low-temperature end, blue in the center, and blue-green at the other end. Associated with the color change was an increase in diameter of 0.9 mil and 1.3 mils at the ends but no measurable increase occurred at the center.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluations

The calibration of aerial survey equipment for both Columbia River and land masses was begun.

New fuel oil analytical procedures show that no measurable quantity of fuel oil from the 100-N spill is currently reaching the river.

Increases in river concentrations of I^{131} from reactor effluents have resulted in a calculated incremental thyroid dose to Tri-City infants of about 4 mrem to date. Slight but persistent increases in shoreline dose rates above McNary reservoir are attributed to continuing shoreline escape of leaking effluents from K Area.

Dosimetry Technology

Development work continued on a thermoluminescent dosimetry system for personnel not routinely exposed to radiation.

Personnel Dosimetry

Annual radiation exposure reports were issued responsive to AEC Manual Chapter 0502. Instrumentation is available for estimating Pu^{238} deposition in liver.

Instrumentation Services

Drift tests with a surface contoured diode and transistor-tunnel diode discriminator, as slated for application in wide dynamic range radiation monitoring, revealed variations of only $\pm 5\%$ with a charge discrimination of 3×10^{-13} coulomb per pulse, equivalent to gamma discrimination between 125 and 360 keV.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluation

(J. F. Honstead, Environmental Studies Section)

Aerial Survey

The aerial survey study was begun. The study includes three distinct portions: (1) calibration of existing gross measurement equipment using a planar source, (2) development of a capability to perform gamma energy analysis of isotopes in the Columbia and (3) development of similar capability over land areas. Over 400 Cs¹³⁷ sources are to be used in simulating a planar source. Measurements over the planar source at various altitudes will be compared to measurements over point sources of varying energies. At least three different NaI crystals sizes (3 x 5, 5 x 4 and 4 x 9 inches) are to be investigated for the gamma energy analysis. Measurements were made with the analyzer over a section of the river at the same time a river sampling traverse was conducted. The analytical results will be compared to the aerial survey results.

Oil Contamination in Columbia River

The solvent extraction-infrared procedure for analyzing fuel oil in water samples was improved to the point where it is statistically possible to identify 0.15 ppm. Since the first of the year when the results averaged around 0.3 ppm the concentration has decreased to the point where it is not detectable. The infrared spectra were identical for both Vernita and 100-F samples. Thus, it appears the fuel oil from the spill at 100-N is no longer reaching the river.

Environmental Evaluations

Increased concentrations of I¹³¹ have been observed in continuous water samples at Richland and Pasco since January 16. The maximum concentration in any weekly sample was 17 pCi/l in river water at Richland, about twice the level expected from recent experience. The initial results could be correlated with

a more satisfactory result can be obtained with about 0.005 in. of iron as a filter. Measurements with 0.005 in. of iron and with the disc shifted more over the tantalum filter are being conducted to provide the necessary response data.

An engineering drawing was prepared of the TLD carrier to be used in the personnel dosimeter. The drawing is currently being detained for approval until the energy response study for the filtered TLD is completed. An attempt is being made to obtain a response from 16 keV photons that is about 35% of the response from radium gamma and linear for energies greater than 50 keV.

Personnel Dosimeter System for Routinely Exposed Workers

Development work continued on neutron dosimetry using the activation foil dosimeter. TLD-700 ribbon (on loan from the Douglas Laboratory) indicated a response characteristic from PuF_4 exposures that was similar to duPont 508 emulsion film. Further development work will be continued when the 1/8 in. x 1/8 in. TLD-700 ribbon now on order is received.

A supply of $\text{Li}_2\text{B}_4\text{O}_7$ Teflon discs was received and gamma response studies started. Lithium borate shows promise of being useful as a neutron dosimeter using backscatter from the body. The sensitivity of $\text{Li}_2\text{B}_4\text{O}_7$ is 1/3 or less than the sensitivity of LiF . The $\text{Li}_2\text{B}_4\text{O}_7$ must be used in combination with a dosimeter responsive only to gamma radiation in order to measure neutron exposure.

Three TLD readers of different manufacture were observed in operation: the EG&G peak reading instrument, the Harshaw integrating instrument, and the Madison Research integrating instrument. The latter was tested in the laboratory and found to have no regulated heat cycle, an inadequate analog to digital converter and an intermittent trouble in the counter logic. The EG&G reader provided only a glow curve versus time with no digital output (a must in our TLD system). The Harshaw integrating instrument (which has preheat, rate, time, and maximum temperature control on the heat cycle) has advantages for our particular purpose.

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Organ Counter Studies

Design continued on an apparatus employing conventional six-inch diameter NaI crystals, low-background photomultiplier tubes and appropriate shielding which should not only detect organ burdens of various radionuclides, but also should provide information on the movement of radionuclides between organs.

The sensitivity and response characteristics of our Nuclear Diodes, Inc., dual Ge(Li) detector to various gamma-emitting radionuclides contained in the liver, kidney and lungs of a REMAB phantom were examined to establish the usefulness of these detectors in organ counting.

Personnel Dosimetry

(H. V. Larson, Personnel Dosimetry Section)

All year-end occupational exposure reports were issued to Hanford contractor organizations with the exception of the individual exposure reports which will be issued to all employees the week beginning March 11, 1968. Also, annual radiation exposure reports summarizing Hanford internal and external occupational exposure experience for 1967 were issued responsive to AEC Manual Chapter 0502.

Personnel exposure data for (1) duPont employees at Hanford for the period 1944 through 1946 who subsequently transferred to General Electric and remained at Hanford, (2) General Electric employees at Hanford for the period 1946 through 1964, and (3) Vitro employees at Hanford for the period 1962 through 1964 were summarized and forwarded to HEHI for inclusion in the Mancuso Study. The total radiation exposure from external sources received by each person during his employment at Hanford was summarized by year. Exposure received at locations other than Hanford was summarized separately, by years. Internal depositions were listed separately for each known intake of bone-seeking radionuclides.

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Biological Services

(W. L. Templeton, Aquatic Ecology Unit)

Columbia River Water

Columbia River water, as presently delivered to the 146-FR, is supersaturated with nitrogen and has caused some problems in holding adult fish at elevated temperatures. The percent saturation has reached 140% compared with 120-125% in the river.

The problem has been reduced to some extent by pretreatment of the water for each experimental trough by aeration in a series of bubble trays. Reductions to 92-105% have been achieved.

Thermal Requirements of Young Chinook Salmon

The experiment on the effect of different temperature regimes on young chinook continued. All lots have hatched, and except for the control lots, the fish have started feeding. The mortality data through hatching show no temperature effects, but we expect to differentiate the temperature effects when all lots develop past the critical initial period of active feeding. Our past work indicates that a latent mortality can be expected for adverse temperatures.

The variation between females or spawns has been wide. For egg mortality, the controls showed a range of a low of 8% to a high of 44%.

Instrumentation Services

(W. G. Spear, Instrumentation Section)

Wide Dynamic Range Radiation Monitoring with Surface Contoured Diode Detectors

Drift tests, conducted over a two-week period, with a surface contoured diode and transistor-tunnel diode discriminator, revealed variations of approximately $\pm 5\%$. No general fatigue trends were apparent. A charge discrimination level of

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