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C-65, Plutonium Production
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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

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JUNE 1968

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By Authority of CG PR-2 (CP 824) Division of Production

US Lewis 2-10-64.

and

By Opie Mullen 2-15-64.

Verified By J. E. Sorenson Hanford Plant Assistance

2-23-64

Programs

by the

Staff of Battelle-Northwest

F. W. Albaugh, Director

July 1968

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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

Estimates of the relative contribution of various species of Columbia River fish to the radiation doses received by local consumers (as a group) were calculated from the results of a household survey conducted in 1967. Probable species composition of typical and maximum fish diets is being estimated from diet data previously collected at the time of routine employee whole-body counts.

About 80% (500) of the game-bird questionnaires mailed to local hunters have been returned. Telephone interviews of a few nonrespondents are under way to characterize the nonrespondent group as a separate stratum of the local population of hunting license holders.

Additional tests of the digital simulation model for emergency application were undertaken.

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DIRECT AEC SPONSORED PROGRAMS

Columbia River Studies

Effects of Nuclear Effluent on the Quality of Columbia River Water

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

Evaluation of two previous tests of the digital simulation model for emergency application continued. Comparison of hourly ^{24}Na measurements in the river to predicted concentrations based on river flow conditions and ^{24}Na input data should be possible now that all data have been prepared for input to the program. Comparison of special isotopic measurements undertaken at McNary Dam to the predicted concentrations is awaiting completion of isotopic analyses.

Another test of the program is planned with ^{131}I as the isotope of interest. Data are being obtained on the measured concentrations of ^{131}I in the Columbia River both at 300 Area and Richland that resulted from the recent release of fission products from 105-KW. An excellent set of arrival time and concentration data is available for comparison with predicted data.

Mechanisms of Environmental Exposure

(J. F. Honstead, Environmental Studies Section)

Recalibration of the Whole-Body Counter Laboratory (Mobile Unit B) for ^{40}K was satisfactorily completed at the end of May. Reconstitution of the ^{40}K sources eliminated the erratic results previously obtained for various phantom sizes. Some difficulty with highly variable background was noted during some of the calibration runs. In June it was found that operation of the Negative Ion Accelerator (Building 3745A) could lead to exposure rates as high as a few tenths of a milliroentgen per hour inside of the whole-body counter shadow shield. However, there did not appear to be any correlation between the high background readings observed during the earlier ^{40}K calibration and operation of either the negative or the positive ion accelerator.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Summary

Mission 1

Flow testing continued on a column of end-spider equipped Mark IV fuel assemblies in the 189-D flow facility. Flow exposure of 415 hours and 9×10^7 cyclic deflections have been experienced by the spider support system without failure.

Excellent quality convoluted style end-spiders have been blanked from 0.030, 0.040, and 0.050 in. thick Zr-2 sheet stock. Slight die modification should permit equally satisfactory blanking of end-spiders from 0.060 in. stock.

Process development was completed for the casting, machining, assembly, and welding of Pu-Al fuel elements.

Tests were completed on two tubular test sections which simulated the central coolant subchannel of an I&E fuel element, and a total of 16 boiling burnout conditions were defined. The remainder of the experiments which will consider the annular subchannel were scheduled.

The data obtained in earlier N-Reactor fuel element experiments were analyzed using the THOR computer code to correct for test section heat losses and instrumentation calibrations. Comparisons of the corrected data with data of an earlier experiment which used an identical test section with no "W" spring supports indicated that the "W" spring support does not adversely affect the boiling burnout limits of this fuel.

Two high-frequency, ultrasonic, bond test systems were completed and performed successfully during eight weeks of continuous on-line operation. These units were used to examine a recent extrusion run of thin clad N-Reactor fuel.

Radiography with X ray and gamma ray was performed on welds and pipes for various on-site customers with satisfactory results. Magnetic particle examination of welds and pre-

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sample which had been oxidized to 4.3% burnoff showed a reduction of compressive strength of 25%. This is similar to that obtained from a non-coated sample oxidized to the same extent.

A gas recirculator for use in the Co gamma facility was completed and some preliminary work begun. A molecular sieve trap and a water diffusion cell allow continuous removal of water or holding the water concentration constant. Irradiation of a 50-50 helium-nitrogen mixture, with about 50 ppm of water vapor, in the presence of TSGBF graphite at 400°C showed considerable ammonia formation after 1 hour, but much less after 3 hours.

Investigation continued on the effects of exposure on existing zirconium hydride distribution in production reactor process tubes. Analysis of tubes, charged with unanodized spacers throughout their exposure, revealed that the average total hydrogen content at selected longitudinal positions of comparison tubes with 52 to 58 months exposure was either similar or significantly less than corresponding values of total hydrogen for base-line tubes with 42 to 44 months exposure.

Corrosion losses of titanium and 6063 aluminum were nil in the chromic-sulfuric acid solution planned to be used in cleaning the N-Reactor control rods.

Analysis of N-Reactor pressure tube 2455 for hydrogen showed a level of 53 to 68 ppm in the area 2 to 8 feet from the inlet end compared to 7 to 18 ppm in the remainder of the tube. A similar hydrogen peak was found in the previous tube examined.

A vacuum sampling device was built and tested for extracting samples from the reactor water treatment plant filter beds. Initial samples were size-graded and studied for the effect of temperature on bed expansion during backwashing.

A study indicated that disposal of a decontamination waste to the ground would not affect subsequent waste disposal to the

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Mission 1 - Basic Production Mission

End-Spider Support Development

(J. W. Goffard, Metal Fuels Unit)

An additional 65 hours of flow testing of the charge of Mark IV tube-in-tube N-Reactor fuel assemblies with end-spider support hardware have been accomplished in the 189-D flow loop. Inspection of the 14-element column after the total test period of 415 hours at 110% of normal flow revealed no failures in the end-spider or in the attaching projection welds. The charge has experienced 9×10^7 hydraulically-induced cyclic deflections of the end-spider supported inner component.

N-Reactor Section reported that an end-spider equipped Mark IV assembly was drop-tested onto a discharge trampoline and that the assembly performed well in surviving the deceleration forces. The inner component was displaced axially about 0.5 in. bowing the end-spider without failing any of the connecting webs or the attaching projection welds. The drop test involved a 50-foot air free-fall onto a water-submerged trampoline. Under similar testing, the inner components utilizing the solid stop W-spring support system are usually loosened because of permanent deformation of the W spring.

Tooling for the fabrication of Zr-2 end-spider hardware was completed and trial fabrication runs were made. Convolute end-spiders (an inner and outer rim connected by 4 spiral webs) of 0.030, 0.040, and 0.050 in. thickness were blanked successfully, except that the die would not clear or strip properly with the 0.050 in. stock. Fabrication of additional 0.050 in. thick spiders and 0.060 in. thick spiders will be attempted with slightly modified dies that should effect proper stripping of the die cavities. The blanked spiders were free of cracks, tears, or localized deformations and only light burring was evident on one face of the spiders from the blanking operation. This burred edge can be readily removed by tumbling, blasting, or etching.

<u>Pressure</u> (psia)	<u>Mass Velocity</u> $\times 10^6$ (lb/hr ft ²)	<u>Subcooling or</u> <u>Quality at Burnout</u>	<u>Boiling Burnout</u> <u>Heat Flux $\times 10^6$</u> <u>Btu/hr ft²</u>
<u>0.7 in. ID Tube</u>			
215	2		
	2	58°F	1.54
	4	102°F	2.45
	4	14°F	1.56
	7		
	7	3.2%	1.69*

* Physical burnout

In the subcooled region, the burnout heat flux decreased as the enthalpy at the location of the burnout increased. Near saturation, two inflection points were noted in the curve of burnout heat flux versus enthalpy for a given mass velocity. Just prior to saturation, a minimum burnout heat flux occurred followed by a slight increase to a maximum value at 5 to 10% quality. Beyond this point, the burnout heat flux again decreased steadily with increasing enthalpy. The burnout heat flux maximum was generally 10-20% greater than the minimum value depending on pressure and mass velocity. These effects were tentatively attributed to the increased local velocities which occur near saturation under low pressure conditions and large void fraction changes resulting in transitional flow regimes. This behavior cannot be attributed to pressure-flow oscillations because the low pressure facility was designed to minimize or eliminate these. Although small pressure oscillations were noted at the test section exit in the slug-two-phase-flow regime at low qualities, the flow rate was held very constant under these and all conditions studied. Further analysis of the data is required before complete results can be reported.

The remainder of the experiments which will consider annular coolant subchannels with equivalent diameters of 0.254 and 0.354 in. were scheduled.

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The results were compared with the results of an earlier experiment in which an essentially identical test section without spacers was used. It was found from this comparison that the test section with W springs showed consistently higher boiling burnout heat fluxes by approximately ten percent over the full range of variables considered in these tests.

High Frequency Bond Test

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

A second high frequency, ultrasonic, bond test chassis was fabricated at N-Fuels facilities, duplicating the BNW-developed prototype. Both units have undergone two months of continuous on-line operation, successfully completing the bond testing of a recent extrusion of thin clad fuels. Current efforts are being devoted to the compilation of a report describing the use and maintenance of these new tester systems.

Plant Support Testing Service

(C. B. Shaw, NDT Customer Applications Unit)

Ultrasonic wall thickness measurements and weld examinations were completed on the 1310 decontamination storage vessel in the 100N Area. The weld examination revealed several spurious ID indications. The customer concluded that these indications were the result of normal weld erosion.

Plant Support Testing Service

(C. B. Shaw, NDT Customer Applications Unit)

Radiographs were taken of four six-inch, schedule 40 welds in conjunction with the installation of an orifice assembly for Boiler #3 in the 165 KW Bldg. Four more such investigations will be conducted over the next few months.

Twenty-four one-inch, schedule 40 carbon steel socket pipe welds were radiographed under ASME code standard Section VIII.

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The experiments used an electrically heated model of the downstream half of an N-Reactor MK-I fuel column provided with a typical pressure tube and downstream piping. Additional upstream piping and instrumentation measure and control the coolant entering the individual subchannels.

The experimental program covered outlet riser pressures of 1200, 1350, and 1500 psig at a test section power of 2500 kW and an outlet riser pressure of 1500 psig at a test section power of 1000 kW. Experiments were made under conditions of reducing the total flow to the test section and under conditions of reducing individual subchannel flows. A total of 125 sets of data points was obtained.

The preliminary analyses done thus far have been directed primarily to resolving and reconciling differences in the results of this test section and one used previously. The differences were due to slight variations of dimensions, and while not large, they were greater than expected.

A normalization technique was developed to enable the reconciliation of the data and to facilitate its application to reactor fuel elements. This normalization technique uses two ratios, the ratio of the experimental pressure drop to the experimental pressure at the flow at which boiling starts and the ratio of the flow to the flow at which boiling starts. Using this technique, the data for the two test sections agreed quite closely; all but two data points out of sixty fell within a $\pm 5\%$ band.

The successful application of this normalization technique shows that the differences in the data can be eliminated and also provides a rational method of applying the data to the reactor.

PCTR Water Tank Experiment on 2.1 wt% ^{235}U NPR "Base" Rod-in-tube Fuel Elements

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

Experimental work was completed and analysis of data

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components can be measured with the neutron absorber placed in the lattice. Coolant water will then be added to the process tubes in the test lattice. The same series of measurements performed on the dry lattice will be repeated on the wet lattice.

Mission 8 - Nuclear Safety

Release and Control of Fission Gases Under Reactor Accident Conditions

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

The study of noble gas behavior following loss-of-coolant accidents involving the low pressure Hanford reactors was extended to include a single cross-header accident, an appraisal of the relative behavior of iodine, and study of a method for leak testing an installed crib.

The study has shown that, for a single cross-header accident, the path followed by the fission gases will be similar to that found for a larger scale accident in which reduced flow of coolant to the entire reactor was assumed. For the larger scale accident, it was concluded that the fission gases would largely follow the effluent water circuit.

Iodine is much more soluble and reactive than the noble gases and hence would release from water to a much lesser extent. It was concluded that, for the quench-tank crib disposal system considered, the I_2 would not represent as great a hazard as the noble gases. Since the rate of atmospheric release of the noble gases was predicted to be minimal, I_2 would be of even less importance.

Leak testing of an installed crib over a time period of several hours appeared to be feasible based on present technology. The cost of carrying out a test based on a proposed method would not be prohibitive. The proposed method is based on atmospheric sampling using 3 different noble gas radioisotopes simultaneously. Two of the isotopes would provide limits on the atmospheric dilution and the third would be used to trace the leak. Leak rates of 1% or greater over a

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was oxidized by 6,000 ppm of water vapor to a total of 4.3% burnoff at temperatures of 800, 850, 900 and 950°C. About 0.5% burnoff was obtained at each temperature and the test repeated to assess possible effects of burnoff. No significant increase in rate was observed with burnoff. The oxidation results indicate a protection factor of 3 to 4 at 950°C decreasing to less than 2 at 800°C.

These samples were tested for compressive strength at the conclusion of the oxidation tests. Some comparative results are shown in the following table:

Compressive Strength of TSX Graphite

<u>Sample Description^a</u>	<u>Compressive Strength lb/in²</u>
Coated, oxidized to 4.3%	3200
Coated, non-oxidized	4210
Non-coated, oxidized ^b	3750
Non-coated, non-oxidized	4997

^a Samples are 1 in. long x 0.43 in. diam cylinders of TSX.

^b This result calculated from data provided in BNWL-SA-989 assuming a non-coated, non-oxidized strength of 4997.

No conclusions should be made regarding the absolute strengths of the coated samples versus the non-coated because sample to sample variation on compressive strength data may be this large. It may be significant, however, that a similar amount of oxidation on the coated and non-coated samples produced a reduction in strength of about 25% in both cases.

Irradiation of the Graphite-Helium-Nitrogen-Water Vapor System
(E. D. Jenson, Materials Compatibility Unit)

Construction of a gas recirculator for use with the gamma facility was completed and placed in operation. The cell consists

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The combination of molecular sieve trap and water diffusion cell allows maintenance of the water concentration at a constant level. Water vapor is added by the diffusion cell, reacts in the cell, is removed by the molecular sieve, and is then added by the diffusion cell to the same moisture level again. By-passing the molecular sieve trap would allow the water concentration to build up to a steady state level approaching its vapor pressure at that temperature.

Preliminary experiments using a 50-50 mixture of helium and nitrogen with about 50 ppm of water vapor added and the cell at 400°C showed 46,000 ppm of NH_3 formed in one hour (corresponding to a G value of 250) followed by the ammonia yield dropping to 3300 ppm after 3 hours (to a G value of 6). Carbon monoxide was observed with a G value of 2.5 after 3 hours but was not detected after 1 hour. For this run both the water diffusion cell and molecular sieve trap were bypassed after establishing the water concentration. This would allow the water concentration to decrease as reaction occurred. The amount of ammonia observed is far more than could have come from the gas phase water. However, it possibly represents the contribution of water or hydrogen absorbed on the walls and on the graphite even after having been under less than 1 micron pressure. This drop in NH_3 concentration may explain why no ammonia was detected in an ampoule experiment of over 50 hours duration. Apparently radiolytic reactions of ammonia remove it from the system. At the conclusion of the run oxygen was found in the vessel, but not hydrogen. The ammonia formation could, therefore, have been formed with a nitrogen oxide intermediate.

A second run was made with the water diffusion cell and the molecular sieve trap in line. The changes in gas composition were similar to the first run. Again no hydrogen or other gases were detected but a large amount of oxygen was present.

Low Temperature Zircaloy Hydriding

(W. K. Winegardner, Corrosion Research Unit)

Effects of continued exposure on existing production

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by 4 ma/cm² for 333 hours. This relatively short exposure increased the total hydrogen content to about 300 ppm. These case-hydrided samples, with controls, are currently being exposed in process water sparged with gaseous hydrogen at 95°C.

Corrosion in Chromic-Sulfuric Acid Cleaning Solutions

(A. P. Larrick, Corrosion Research Unit)

Plans were made to chemically clean the coolant passages in the N Reactor control rods to restore flow to original conditions. Corrosion tests were conducted on titanium and 6063 alloy aluminum, the two major materials of construction, in a room temperature solution of 12.8% sodium dichromate, 4.4% sulfuric acid, 82.8% water solution. This solution has been effective in cleaning similar equipment in the past and is being considered for the control rod cleaning. Corrosion calculated from weight losses was nil for the titanium and 0.07 mil for the aluminum for a 24-hour exposure period. No nonuniform corrosion was observed.

N Reactor Pressure Tube Examination

(A. P. Larrick, Corrosion Research Unit)

N Reactor Pressure Tube 2455 is currently being examined to determine the amount of corrosion and hydrogen absorption which occurred during in-reactor operation. The tube is an original tube which was exposed from the time of reactor startup to November 1967.

Although not complete, the hot vacuum extraction hydrogen analyses are confirming findings of a previous tube examination, i.e., higher hydrogen levels occur at an area starting about 2 ft from the upstream end and extending at least 6 ft downstream. The fuel charge starts approximately 11 ft from the upstream end. At the upstream end, and from the center of the tube downstream to the outlet end, the hydrogen levels are 7 to 18 ppm. At the area of high hydrogen levels near the upstream end, hydrogen levels as high as 68 ppm were found. These levels are a few ppm higher than in the previous tube

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<u>Mesh Size</u> (mm)	<u>Weight % Less Than Mesh Size</u>			
	<u>Filter #5</u>		<u>Filter #10</u>	
	<u>Anthracite</u>	<u>Sand</u>	<u>Anthracite</u>	<u>Sand</u>
1.41	51.7		45.2	
1.68	67.2		65.3	
2.00	81.3		86.5	
2.38	88.3		96.2	
3.36	100.0		100.0	

<u>Filter</u>	<u>Effective Size</u>		<u>Uniformity Coefficient</u>	
	<u>Sand</u>	<u>Anthracite</u>	<u>Sand</u>	<u>Anthracite</u>
5	0.44	0.98	1.50	1.54
10	0.49	1.02	1.39	1.54

The filter material from filter #10 was placed in a glass column with 24 in. anthracite and 6 in. sand to study the effect of temperature on bed expansion during backwashing. Following are the results from these experiments.

<u>Temperature,</u> <u>°C</u>	<u>Backwash Rate,</u> <u>gpm/ft²</u>	<u>Bed Expansion,</u> <u>%</u>
6	12	8.7
	14	12.3
	16	16.0
	18	19.7
	20	23.3
10	12	7.0
	14	10.7
	16	14.3
	18	18.0
	20	21.7
15	12	5.0
	14	8.7
	16	12.3
	18	15.7
	20	19.3

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Mission 1

NPH as delivered to the Purex Plant contains a small amount of aromatic and olefinic impurities. The effects of these specific impurities on the quality of the solvent had not been investigated previously. A series of experiments in which the concentration of these impurities was varied indicates that they have a direct relationship to fission product retention. Specifications pertinent to the olefinic and aromatic content should be reappraised and incoming NPH diluent carefully monitored to assure adherence to specifications.

In the DBBP solvent extraction process for recovering Np from Purex process Zirflex 1WW waste, Na₅ DTPA and Na₄ EDTA were found to be slightly superior and Na₃ NTA and Na₃ HEDTA inferior to citric acid for scrubbing zirconium from the organic.

The presence of the rate-accelerating material (RAM) for oxidation of Np(V) to Np(VI) had no significant effect on the behavior of ZrNb and Ru in batch contacts simulating Purex HA column extraction and scrub sections. Extraction coefficients E_a^0 for RAM were 0.02 for extraction contacts and 0.2-0.6 for scrub contacts. Satisfactory RAM yields have not yet been obtained in pilot scale equipment studies.

Studies aimed at elucidating the "Zirflex tonic effects" were continued. The improved decontamination noted in the Purex plant when a small amount of zirconium cladding dissolver solution was blended with aluminum cladding dissolver solution was confirmed in laboratory studies. Addition of inert zirconium to Al-clad dissolver solution did not improve decontamination; addition of an Al-Zr-F solution did.

In organic aqueous drop coalescence studies, anionic and cationic surfactants and transfer of uranium from the organic

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higher with 0.3M HDEP-0.2M DBBP-NPH as solvent than with 0.3M HDEP-0.2M TBP-NPH. The increase was about 20% for U-Al alloy process waste. Cerium(III) E_a^0 values were lower with DBBP instead of TBP in the solvent for both wastes.

Further studies on the Alniflex dissolution of U-Zr alloys indicate that the dissolvent must be at or near boiling temperature for satisfactory dissolution rates.

On the basis of solution stability, Zircaloy-2 dissolution rate and the time required to penetrate the oxide layer present on Zircaloy-2 clad fuels, the optimum initial dissolvent for Zircaloy-2 clad U-Zr alloy fuels appears to be 1M HNO_3 -0.5M $\text{Al}(\text{NO}_3)_3$ -1.25M HF. Assuming a dissolved Zircaloy-2 concentration of 0.5M, the required terminal HF concentration is 2.5M. Stable solutions are produced under these dissolution conditions and the induction period required to penetrate the oxide layer is only about one-half hour. HAP0-20 alloy corrodes in the initial and in the terminal dissolvent at rates of about 2.5 and 0.7 mils/mo, respectively.

Mission 8

Further experiments to evaluate the airborne release of plutonium from burning materials showed that a maximum of 1% would be released under the conditions imposed. The nature of the materials burned did not have a decisive effect on release within the spread of the data.

A literature search and compilation of abstracts on incidents involving loss-of-control of plutonium was under way.

Mission 9

Absorption of nitric acid from dilute solutions reported last month was found to be a property of the carbon substrate rather than of the sorbed DBBP. Uranyl nitrate was not sorbed by the DBBP-carbon sorbate, but a DBBP-uranyl nitrate complex was. Uranium was easily removed by washing with water and DBBP was removed by washing with acetone.

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lower than values obtained from a dummy exposed in an aluminum tube.

Process Technology

Seventeen grams of heart-cut curium oxide have been shipped to the Savannah River Laboratory.

Burning, dissolution and processing of 7 1/2 impregnated graphite-neptunium oxide elements were completed in the 325A Bldg. high level cells. Burning of 7 1/2 pressed graphite-neptunium oxide elements was completed.

Continuing study of the properties of B-plant solvent has not yet disclosed the reason for the gradual decrease in Sr extraction capacity of the solvent.

Laboratory studies on N-Reactor fuels sheared at ORNL show that a zirconium fire initiated at a feather edge on a massive piece of sheared fuel (with uranium removed by nitric acid leaching) will propagate into a mass of needles and fines produced by shearing.

Laboratory and engineering scale studies continue to indicate that thorium wafers can be dissolved in Purex annular dissolvers without the use of air lift circulators and that, because a large size charge and heel could be used, short dissolution times are possible.

Mini mixer settler runs testing proposed Purex plant 2nd and 3rd plutonium cycle flowsheets were continued. Both ferrous sulfamate and sulfuric acid stripped plutonium satisfactorily under proposed 2B column conditions. Erratic and generally unsatisfactory plutonium stripping was obtained with either hydroxylamine nitrate or hydrazine-stabilized ferrous nitrate.

Graphite, silicon nitride and boron nitride continue to show promise in laboratory work to determine their usefulness as materials of fabrication for reusable crucibles in the reduction of plutonium oxide to metal.

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Negotiations are in progress with the Coast and Geodetic Survey to obtain horizontal earth movement correlations for the Hanford area.

Infrared imagery collected recently over the Columbia River is being processed in an attempt to detect ground water entering the river.

A comparison for cesium removal from PSN solution was made between Davison 700 and Linde AW 500. AW 500 cesium loading to 10% breakthrough from PSN was 5.2 meq Cs/100 g and for Davison 700 was 3.6 meq Cs/100 g.

Trace cesium and strontium adsorption on soils was determined in a multicompeting cation system at two levels of soil clay content. Cesium sorption showed a strong correlation with clay content; however, no significant difference was obtained for strontium.

Experiments to study radionuclide behavior in specific retention systems were initiated. Complexed ^{60}Co is being used to study the movement of waste fronts in soils.

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Mission 1 - Basic Production Mission

NPH Quality

(E. C. Martin and T. W. Lawell, Organic and Natural Product Chemistry Unit)

Although NPH has proved quite satisfactory in the Purex Plant and specification tests have been devised to permit purchase on a bid basis, some aspects of the specifications now accepted are not completely satisfactory. In particular, the 98% minimum NPH and the 0.2% maximum aromatic are somewhat arbitrary. If the nature of the small amount (2% or less) of impurity in the NPH were better identified and if these impurities were then evaluated in a realistic use test, then perhaps a more relaxed specification could be issued. Other major gains would be an improved confidence in laboratory testing and information on which to base solvent washing procedures for the Purex Plant.

It has been found that silica gel has the property of removing olefins and relatively high molecular weight aromatic compounds from NPH. Since this is one of the areas of concern in the list of specifications for NPH, it seems reasonable that the effect of these impurities be examined with some care. The present use test experiments were set up to examine the NPH as received after silica gel treatment and after spiking with additional impurities obtained from silica gel removal process.

The use test is designed to resemble as closely as possible the operations in the Purex Plant, clean up, extraction, scrub and stripping steps. Before the test experiments were undertaken, the 30% TBP solutions in NPH were subjected to a chemical degradation by contact at 70° for 24 hours with an equal volume of 8M HNO₃ and 0.05M NaNO₂ in a closed system. The degraded NPH solution was then cleaned up by successive washes with water, dilute (0.025M) KMnO₄ and Na₂CO₃, HNO₃, Na₂CO₃ and finally HNO₃.

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and with RAM present (0.015M RNO_2 from a preparation that gave $k_2(\text{RAM})/(\text{RNO}_2) = 0.6 \times 10^4$). To exaggerate any effect that might occur, RAM was added to the scrub solutions as well as to the aqueous phase before extraction. Nominal phase compositions were 0.015M U + 3.1M HNO_3 in the aqueous and 0.23M U in the organic. With RAM present, decontamination factors of 24,000 for ZrNb and 6,500 for Ru were obtained, compared with 15,000 to 20,000 for ZrNb and 3,000 to 6,300 for Ru for duplicate experiments in the absence of RAM.

The extractability of the RAM itself was also studied in the experiment. The extractability was too low to be measured by difference in the amount of RAM present in the aqueous before and after extraction; instead, it was necessary to use the amount of RAM found in subsequent scrub and strip solutions as a measure of the amount extracted.

A 0.12M RNO_2 + 3.1M HNO_3 solution prepared from 7M HNO_3 backmix reactor product was contacted with three volumes of pre-equilibrated solvent which was then scrubbed twice with 3.1M HNO_3 (A/O = 0.4) and stripped three times with water (A/O = 0.8). Values of $k_2(\text{RAM})$ in the aqueous after extraction and in the scrub solutions were 730, 30, and 5. From the $k_2(\text{RAM})$ values found in the aqueous phases and the volume ratios employed, decreases in $k_2(\text{RAM})$ in the organic phase of 12 and 2 resulted from the two scrub steps and a decrease of 3 occurred on stripping three times with water. Assuming that all of the RAM was removed from the organic by this sequence of steps, RAM extraction coefficients (E_a^0) of 0.02, 0.2, and 0.6 were obtained for the extraction and two scrub steps.

Pilot plant scale equipment was assembled and tested for full-scale production of RAM. To date satisfactory yields of RAM have not been obtained. Inaccurate temperature measurement during the mixing of the aciform nitropropane and the HNO_3 - HNO_2 streams is thought to be the problem. Equipment modifications to overcome the problem have been made and further test runs were planned.

A point of concern at present is that decontamination from Nb appeared to suffer markedly in the two experiments in which inactive zirconium was added, to the extent that decontamination from ZrNb apparently suffers. It is felt that this apparent result may be caused by the experimental procedure; in all of the experiments there has been about an hour delay between the extraction and the first scrub contacts. During this time the amount of Zr that decays to Nb is calculated to be greater than the amount of Nb present in the organic after the fourth scrub contact. This situation would not exist in the plant columns so a difference in apparent Nb behavior is not very unlikely. The fact that the apparent worsening of Nb decontamination showed up only in the experiments containing inactive zirconium indicates, however, that the complete explanation may be more complex.

Data illustrating the points discussed in the preceding paragraphs are given in the following table.

Zirconium Scrubbing Behavior

Feed	Zr Behavior on Extn. Step		DF Through Extn. and 4 Scrubs (x 10 ⁻⁴)			4th Scrub	
	<u>E_a</u>	<u>DF</u>	<u>Zr</u>	<u>Nb</u>	<u>ZrNb</u>	<u>Zr</u>	<u>E_a</u>
Zr-clad	0.039	8.5	11	36	20	0.15	
Al-clad	0.021	16	0.9	3.5	1.5	3.4	
Al-clad	0.024	14	1.2	8.2	2.4	1.2	
Blend(1)*	0.038	8.7	5.1	40	11	0.4	
Al-clad + 0.002M Zr*	0.060	5.6	0.74	1.0	0.8	1.1	
Al-clad + 0.01M Zr + 0.01M F							
+ 0.03M Al*	0.086	4.0	2.5	1.1	1.5	0.4	

(1) 0.16 Zr-clad + 0.84 Al-clad on U basis.

* Digested about 6 hr at 50-60°.

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<u>Aqueous Phase</u>	<u>Coalescence Behavior</u>
Demineralized water	Rapid coalescence
10 ppm sodium docecylsulfate (A)	No coalescence
10 ppm di-n-butylphosphate (B)	Rapid coalescence
10 ppm trimethyl dodecyl ammonium (C)	No coalescence
1 ppm A	Slow coalescence
1 ppm B	Rapid coalescence
1 ppm C	No coalescence
10 ppm A, 0.01M HNO_3	No coalescence
10 ppm B, 0.01M HNO_3	Rapid coalescence
10 ppm C, 0.01M HNO_3	No coalescence
1 ppm A, 0.01M HNO_3	Slow coalescence
1 ppm B, 0.01M HNO_3	Rapid coalescence
1 ppm C, 0.01M HNO_3	No coalescence

When the aqueous was demineralized water and the organic was 30% TBP-NPH-0.1M UNH coalescence did not occur; at 0.01M UNH in the organic, coalescence was slow; addition of 10 ppm DBP to the aqueous did not change the coalescence behavior.

In summary, anionic and cationic surfactants and transfer of uranium out of the organic phase had a depressing effect on coalescence of the two drops.

Stress Corrosion Cracking of Mild Steel

(R. F. Maness, Corrosion Research Unit)

Mild steel weldments, 1 x 3 ft, were fabricated from ASTM A-515, grade 60 (3/8 in. plate) and A-283, grade C (1/2 in. plate) and exposed in duplicate to four compositions of boiling NaNO_3 - NaNO_2 solutions at pH 9.5. All weldments were braced to produce a restrained seam weld and all contained a simulated weld repair. The test solutions all contained 5M sodium ion with nitrate and nitrite concentrations varying from 1-4M. These solutions simulate the NO_3^- - NO_2^- compositions found in

of construction. Three inexpensive operational amplifiers can be combined to provide preamplification and pulse height to DC conversion. Nominal useable solution loop resistance appears to be from 1 to 1000 ohms.

Development of Alpha Scintillation Sensors

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

Further examination and testing of cerium activated Vycor alpha scintillation detectors was conducted in an effort to define alpha vs beta-gamma sensitivity as a function of cerium diffusion depth and diffusion gradient. Electron microprobe examination revealed that the gradient zone varied from 5 to 20 μ across one typical sensor and 25 to 30 μ on another. The depth of the diffused zone varied a maximum of 41 μ across a 3 in. sensor indicating that grinding of the sensor during manufacture was not precisely parallel to the diffusion band. A selected sensor was ground to reduce the diffusion zone from 13 mils (331 μ) to 1 1/2 mils (38 μ) without an appreciable effect on alpha sensitivity but with a reduction in beta sensitivity to 1/500. This sensor will be used as a standard for comparison with others to determine the reduction in the diffusion band necessary to produce an equivalent.

Improvement in alpha to beta-gamma sensitivity by an estimated factor of five may be possible through choice of a manufacturing process to produce a narrow gradient zone and grinding parallel to the diffusion band to form a uniform activated layer equal to the penetration depth of the alpha particles, i.e., 13 μ for 4 MeV alpha particles.

Mission 5 - Other Isotopes

Extraction of Po from Bi by Fused NaOH

(G. F. Schiefelbein, Separations Chemistry Unit)

The final large-scale full-level test of the fused NaOH process for removing ^{210}Po from irradiated bismuth was successfully completed.

	<u>Total Radiation, R</u>	<u>Conc. DBP ($\times 10^{-2}M$)</u>	<u>Conc. MBP ($\times 10^{-3}M$)</u>
TBP (blank)	2.3×10^7 R	0.94 ± 0.03	0.83 ± 0.13
Benzene	"	0.63	0.76
Napththalene	"	0.70	0.67
Xylene	"	0.78	0.72
Phenanthrene	5.2×10^7 R	0.52	0.85
Biphenyl	"	0.87	1.0
Anthracene	"	0.86	0.93
TBP (blank)	5.4×10^7 R	1.3	1.3
Xylene	"	1.2	0.92

In every instance there appeared to be a reduction of the TBP decomposition. Compounds such as phenanthrene appear to exercise a very strong protection for this system. Additional studies to determine the fate of the protective agent and other techniques to monitor the TBP concentration are under way.

Processing Enriched Fuels Wastes in the Hanford Waste Management Complex

(J. A. Partridge, Separations Chemistry Unit)

Initial determinations of E_a^0 Sr values for the U-Al and U-Zr alloy waste solutions for HDEHP solvent extraction (B-Plant process) were made with the aqueous feed containing 0.20M hydroxyacetate (HA) and 0.025M tartrate. More recent data, however, show that the hydroxyacetate and tartrate are not necessary for buffering in the presence of the high citrate required for complexing in these solutions since citrate itself has a buffer region around pH 4. Furthermore, the E_a^0 Sr values are higher in the absence of the HA-tartrate, about 35% higher for the U-Al waste solution and about 15% higher for the U-Zr waste solution for feed solutions in which the citrate to total metal ion ratio is one or greater. Deletion of hydroxyacetate and tartrate would have the twin advantages of more favorable strontium extraction and reduced chemical costs.

Some scouting experiments were performed to check the extraction of Sr into 0.3M HDEHP-0.2M DBBP-NPH (vs 0.3M HDEHP-

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oxide layer is only about one-half hour. HAPO-20 alloy corrodes in the initial and in the terminal dissolvent at rates of about 2.5 and 0.7 mils/mo., respectively.

Recirculating Dissolvers

(W. V. DeMier and G. Jansen, Engineering Analysis and Economics Unit)

Program BARD (FORTRAN V, Univac 1108) to calculate compositions in batch and recirculating dissolvers has been coded and compiled and most of the debugging completed. Preliminary runs with all types of dissolver and reservoir configurations promise a less than two minute solution time per case for dissolutions with single reaction rate equations.

Mission 8 - Nuclear Safety

Atmospheric Release of Radioactive Particles

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

Additional experiments were performed to determine the plutonium released from combustible materials. Release during the burning of fine shavings, cardboard, rubber scraps, and cheese cloth was found to range from about 0.02 to 1%. These, with previous experiments, have shown that the release can vary through this range without showing a significant dependence on the nature of the material burned. It thus appears that 1% is the maximum amount which would be airborne when these kinds of materials are ignited and burned under the conditions maintained.

Three experiments were performed to investigate the fraction of plutonium made airborne from sand onto which plutonium nitrate had been dried. In these experiments the sand, contained in a shallow dish, was heated to 1000°C or 1200°C and the airborne material collected on a filter. The conditions imposed were selected to simulate a possible accident involving loss of integrity of a shipping container and subsequent fire in which the liquid spilled into the soil was subjected to

DBBP completely free of uranyl nitrate. This parallels the behavior of the uranyl nitrate-DBBP complex in solvent extraction technology, where water (or 0.01M HNO_3) strips the complexed uranyl nitrate from the organic phase.

Carbon sorbates of DBBP will not be effective for removal of uranium (and presumably plutonium) from dilute nitric acid streams. Carbon will, however, efficiently remove DBBP from aqueous solutions. Acetone efficiently displaces DBBP from the carbon.

Z-Plant Organic Wastes

(R. A. Hasty, Applied Chemical Research Unit)

The flowsheet for Z-Plant PRF was studied to determine methods by which the amount of organic waste can be reduced. Organic wastes originate from two principal sources: the low level concentrations of organic matter dissolved or entrained in the aqueous waste streams and from the TBP- CCl_4 or DBBP- CCl_4 solutions which no longer possess suitable solvent extraction properties. The first of these sources arises because of the finite solubility of the organic materials in aqueous solutions and incomplete phase separation. Removal of these organics being investigated in the study of Z-Plant aqueous wastes. The second of the two sources is a cause of larger concern because it involves the disposal of large volumes of organic solutions. Disposal by incineration is being investigated but recovery might be more economically attractive.

A brief examination of the flowsheet of the PRF suggests the following areas for further consideration, beginning with a literature survey.

1. The rates of both hydrolytic and radiolytic decomposition and the identity of the products of hydrolysis and radiolysis for the TBP- CCl_4 and DBBP- CCl_4 systems in contact with both alkaline and acid nitrate solutions.
2. The suitability of CCl_4 recovered by steam distillation for use as a diluent for the PRF.

This study of the hydrolysis in a single phase creates considerably more confidence in the rate of hydrolysis derived from two-phase systems.

The products of gamma radiolysis of TBP alone or in CCl_4 are reported to be

I. Gases

H_2
 C_4H_8

C_4H_{10}
 C_3H_8

C_2H_6
 CH_4

II. Liquids

DPB
Butyl alcohol
Butyl ether

MBP
 H_3PO_4

$\text{C}_n\text{H}_{2n+2}$; $n > 5$
Polymers

The radiolytic decomposition products of carbon tetrachloride were reported to be chloride ion, hexachloroethane, and phosgene. The rate of chloride ion production was reported to be 0.07 g Cl^- per watt/hr of energy absorbed. The presence of benzene was reported to stabilize TBP toward ionizing radiation.

Customary solvent treatment removes major hydrolysis and/or radiolysis products (DBP, MBP, and phosphoric acid). Little attention has been given to the presence of the other hydrolytic or radiolytic decomposition products. The effect of minor decomposition products on organic to aqueous distribution of inorganic and organic components needs to be determined.

Burning Tests on ARHCO Waste Solvents

(G. E. Benedict, Advanced Chemical Engineering Unit)

In the tests made in late May at John Zink Company, the solvents incinerated were: 30% TBP in NPH; 20% TBP in CCl_4 , and 100% TBP containing 500 ppm CCl_4 . No difficulties were experienced in burning these solvents (auxiliary gas fuel was

Cesium Recovery from Acidic Wastes

(L. A. Bray and R. A. Bircher, Chemical Separations Unit)

Laboratory cesium loading studies as a function of temperature, flow, and feed variation have again been started with acid-stable inorganic resins (as contrasted with the alkaline-stable resins currently in use). Upon completion of the cold tracer work, at least the two resins ABEDEM-TiA and the AMP resin sent to us by Dr. J. van R. Smit of South Africa will be tested in the 325 High Level Radiochemical Facility using both Zirflex and PAW wastes.

Waste Packaging

(T. S. Soine and H. T. Fullam, Materials Chemistry Unit)

Efforts continued on flowsheet development for the preparation of strontium fluoride and cesium chloride.

Synthesis of strontium fluoride using simulated B-Plant product as the starting material is being studied. The following table gives the presently anticipated composition of the feed material.

Composition of Strontium Nitrate
Solution from the B-Plant Process

<u>Material</u>	<u>Molarity</u>	<u>Mole %</u>
Sr	0.293	62.9
Ca	0.029	6.1
Fe	0.029	6.1
Al	0.029	6.1
Mg	0.088	18.8

Impurities make up better than one-third of the total metallic ions and, except for the aluminum, they will all precipitate as either the hydroxide or fluoride when strontium fluoride is precipitated.

precipitate also. Apparently $\text{Cs}_2\text{U}_2\text{O}_7$ must be made from Cs_2CO_3 and a nearly neutral solution of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$. The Cs_2CO_3 would provide the means for obtaining the basic solution necessary for the precipitation.

Cesium phosphotungstate was prepared by combining CsNO_3 and phosphotungstic acid. The precipitated cesium phosphotungstate filtered rather slowly. After drying overnight at 400°C , the powder density was 1.6 g/cc. The powder step-pressed to a density of 2.8 g/cc, which gave a cesium density of about 0.3 g/cc.

Movement of Radionuclides through Hanford Soils

(D. B. Cearlock, D. R. Friedrichs and A. E. Reisenauer, Hydrogeology Unit)

Permeability distribution determination by the energy dissipation method involves forming the permeability integral by taking first and second partial derivatives of a planar function which describes the water table surface. The derivatives with respect to the vertical direction are assumed to be zero since the surface has only two independent (space) variables. This assumption can potentially result in significant errors in the calculated permeabilities and associated travel times if the curvature of the water table surface changes rapidly. Equations for determining the first partial derivatives were developed based on the orthogonal relationship between potential and stream surfaces (the water table is a stream surface). Efforts are now under way to determine how to form the second partial derivative on the water table surface (rate of gradient change). This work has application in improving the accuracy of flow system analysis.

Several bids have been received on the light-pen and display equipment for use in hydrologic analysis in conjunction with the PDP-9 computer. Discussions were conducted with several of the vendors to clarify specifications and technical requirements of the equipment. Several new program tapes were received that will have application in integrating the display equipment-computer system.

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B-Plant Solvent Quality

(W. W. Schulz and L. C. Neil, Separations Chemistry Unit)

B-Plant solvent extraction operation with radioactive feeds began in early February. Concurrent with plant startup a program was inaugurated to follow, routinely, certain important properties of the plant HDEHP-TBP solvent. As of June 1, the plant extractant had received an average exposure of about 700 watt hr/l. Radiolytic destruction of either HDEHP or TBP has not been excessive. Nevertheless, the strontium extraction capacity of the solvent (as measured by a standardized strontium E_a^0 test) has steadily decreased until it is now slightly less than half that of unirradiated 0.3M HDEHP-0.2M TBP. Addition of virgin HDEHP to the solvent increases the strontium extraction capacity. However, even when the plant solvent is adjusted to 0.4M HDEHP (and 0.3M TBP) the strontium E_a^0 is still only about 80% of what it is for the unirradiated 0.3M HDEHP-0.2M TBP extractant.

The properties and behavior of the irradiated plant solvent parallel very closely those observed in earlier laboratory cyclic ^{60}Co irradiation studies. In both cases the washed irradiated extractants did not appear to contain any H2DEHP. The latter compound is the primary radiolysis product of HDEHP and even small concentrations have a depressing effect on strontium extraction. The reason for the decrease in strontium extraction capacity of the plant solvent is not known. It is theorized that radiolytically generated material similar to H2DEHP is tying up the HDEHP molecules through hydrogen bonding and making it less available for strontium extraction. The experimental program aimed at elucidating degradation mechanisms and preventives or cures is continuing. Meanwhile, preparations are under way in the B-Plant to butt the entire solvent inventory to 0.4M HDEHP and 0.3M TBP.

N-Reactor Fuels Cladding Fires

(W. W. Schulz, Separations Chemistry Unit)

The investigation into the causes, mechanisms, and significance to present and future Hanford Purex plant operations of

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Engineering scale studies on the dissolution of thorium wafers are in progress. Present studies are on wafers (800 lb) obtained from DUN. They are production wafers normally used for capping canned thorium powder.

Initial loading in the EDL-102 Mark V dissolver was 403 lb (2370 wafers). Most of these were dark colored, about 175 were light in color. Two approximately 100 pound cuts were made using, for each cut, 143 liters of dissolvent initially 12.2M HNO_3 -0.1M $\text{Al}(\text{NO}_3)_3$ -0.025M HF. Cut No. 1 was 1.146M in thorium and cut No. 2 was 1.353M. In both the time at boiling was 3 hours.

An additional 200 pounds of wafers was added to the dissolver and two more ca 100 pound cuts were made. This procedure was repeated and then further cuts were made to clean out the dissolver. At the start of Cut No. 3 much frothing and foaming occurred and off-gas vacuum control was nearly lost. After about 30 minutes, the frothing subsided and the run proceeded normally. The same frothing occurred at the start of Cut 4 but did not occur during succeeding cuts. No explanation for the frothing has yet been found. Cut No. 3 reached 1.448M Th in 108 minutes and Cut No. 4 reached 1.431M in 110 minutes at boiling. Analytical data for the remaining cuts, now up to 8, are not yet available. These results correspond to thorium dissolution rates in the range of 200 pounds of thorium dissolved per hour per ton of thorium in the dissolver.

Some fine material was produced during these runs. This material appeared to be readily suspended during dissolving. Some of this fine material and some wafers of near original size remain in the dissolver after the eighth cut. These will be examined to see if they are unusually difficult to dissolve.

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

Certain process advantages may be obtained by replacing the present Purex plant tail-end plutonium anion resin process

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3. Tantalum - The slag wets the surface of the tantalum.
4. Titanium diboride - The boride is not wetted by the slag and there is no evidence of attack.
5. Boron carbide - The surface is wetted by the slag.

Bomb reduction tests were carried out using three different crucible materials which had shown up well in the compatibility tests. Uranium tetrafluoride was used as a stand-in for PuF_4 in the reductions. Calcium (with iodine) was the reducing agent. The crucible materials tested were graphite, silicon nitride, and boron nitride. The results were as follows:

1. Graphite - The slag did not adhere strongly to the crucible. The crucible was not affected by the thermal shock.
2. Silicon nitride - The slag did not adhere to the crucible; however, the crucible cracked due to thermal shock.
3. Boron nitride - The slag did not adhere to the crucible. Slag and button were removed after tapping the crucible. The crucible was not affected by thermal shock.

Additional reductions are planned with vitreous carbon, silicon nitride, silicon carbide, and graphite crucibles. The graphite crucibles will be coated on the inner surfaces with oxides such as UO_3 or CaO to prevent carbon pickup in the metal button.

Reflector Savings for Enriched Uranyl Nitrate Solutions (C. A. Rogers, Critical Mass Physics Section)

Experiments were continued for determining the reflector savings of enriched uranyl nitrate solutions utilizing the Remote Split-Table Machine and a plutonium bearing core reflected with various thicknesses of uranyl nitrate solutions.

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Previous calculations on reflected systems utilizing the HFN multigroup diffusion code and the DTF-IV multigroup transport code have now been extended to include "best" derived values for critical dimensions of unreflected spheres, infinite length cylinders, and infinite slabs.

Vessel Vent System Studies

(L. A. du Plessis, Applied Chemical Research Unit)

A sampling train for Purex off-gases was fabricated. It consists of absorption tubes for water and NO_2 , and a cold trap (liquid nitrogen) for organic iodides and other condensible gases. The stream to be sampled is sucked through the train by an air-jet vacuum pump via a surge tank fitted with a vacuum regulator. To avoid condensation of oxygen (less volatile than nitrogen) the cold trap is operated at reduced pressure. Reduction of pressure is achieved by drawing the gas through a thin tube whose resistance to flow also serves to fix the gas flow through the train. Another thin tube causes a small pressure drop between the cold trap and the surge tank in order to avoid momentary surges of air back into the trap. Tests have shown the pressure in the trap and the gas flow to have the designed values.

Laboratory equipment was designed to study the rate of reaction of gaseous alkyl iodides with AgNO_3 crystals as a function of temperature and iodide pressure.

A photomicrographic study of AgI layers on AgNO_3 was made in order to observe the diffusion of the two substances into each other. No diffusion was observable at room temperature over a period of a few days. At 99°C , the structure became uniform in 46 minutes. These few observations should be supplemented by a more quantitative study, as diffusion in the solid (or melt, since the lowest eutectic of the AgNO_3 - AgI system is at about 100°C) may control the reaction rate between AgNO_3 and iodine compounds.

A cursory survey of thermodynamic data showed that silver is unusual in having a higher standard free energy of formation

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Air Flow, CFM	Water Flows, gal/min			Aerosol Input, mg/m ³	ΔP , in. w.c.	Cleanup Efficiency, %
	Orifice	Impingement	Scrub			
72	2.5	2.5	5	1.34	61	82
77	2.5	0	0	1.33	59	81
80	0	2.5	5	1.33	59	65

Effect of Air Flow and Water Flow on Scrubbing Efficiency

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

At the higher air flows and higher ΔP , little effect of scrub water flow rate was noted in the scrubbing efficiency of uranine aerosol. At the design air flows, changes in the water flow caused noticeable changes in the scrubbing efficiency.

Air Flow, CFM	Water Flows, gal/min			Aerosol Input, mg/m ³	ΔP , in. w.c.	Cleanup Efficiency, %
	Orifice	Impingement	Scrub			
78	1.5	1.5	4.0	1.33	59	83
72	2	2	5	1.4	61	82
68	2.5	2.5	5	1.34	60	80
67	1	1	2.5	1.6	35	63
57	1.5	1.5	4	1.8	37	66
50	2.5	2.5	5	2.15	39	72

The air flows change when the water flows are changed, more markedly at the lower ΔP , and this probably accounts for the greater change in efficiency under these conditions. A damper system on the blower and permanent air flow measuring devices would increase the usefulness of this off-gas system.

Effect of Aerosol Loading on Scrubbing Efficiency

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

Since it was difficult to change one variable in the equipment as set up without changing another, the aerosol loading variable needed evaluation under constant air flow and water flow conditions. No change in scrubbing efficiency was noted when

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The combined efficiencies of the Peabody scrubber and Cottrell precipitator on the uranine aerosol were from 99.85 to 99.93%.

P₂O₅ Aerosol Cleanup

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

A phosphorus pentoxide aerosol was prepared by burning TBP in a propane torch and quenching with cool air. The resulting aerosol proved to be much more difficult to scrub out than was the 0.4 μ mean mass diameter uranine. Under the same operating conditions and pressure drop, the Peabody scrubber removed 85% of the uranine and only 40% of the P₂O₅. The electrostatic precipitator removed from 97 to 99% of the P₂O₅ from the scrubber off-gas versus about 99% for the uranine under similar conditions. It is not known whether the lower scrubbing efficiency for the P₂O₅ is a function of particle size alone or some combination of particle size and chemical differences.

TK-28-4 Incident

(L. K. Mudge, Engineering Analysis and Economics Unit)

Thermal and flow calculation indicated that inadvertent admission of 40 psig steam through a mislabeled steam jet caused the excursion rather than a fire in the tank.

Waste Tank Structural Temperatures

(G. Jansen and J. R. Nance, Engineering Analysis and Economics Unit)

Transient temperatures and vertical temperature gradients in the concrete near a model hot spot were estimated. Wall temperatures were calculated for a hemispherical hot spot by program SPHERE (Univac-1108) assuming that most of the heat flows to the boiling liquid. Vertical temperature gradients in the concrete were then calculated by using these temperatures as input to the one dimensional heat transfer program THRM which can accept arbitrary transient wall temperatures.

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contractors. Accelerometers will be located on basalt above and beneath the water table, at appropriate surface stations, on tank farm and other structures to obtain information on coupling between bedrock, overburden and structures.

Negotiations are in progress with the U. S. Coast and Geodetic Survey to have them utilize their specially developed programs for the simultaneous adjustment of a series of regional triangulation networks to determine the history and extent of horizontal earth movement in the Hanford area. Such information is relevant to nuclear facilities siting and seismicity relationships.

Infrared Imagery Processing

(J. R. Eliason, Hydrogeology Unit)

A hybrid computer program for rectifying infrared imagery was developed using the PDP-7 Beckman 1132 computer system. Infrared imagery collected with the Bendix Thermal Mapper operated by Cartwright Aerial Surveys, Inc. over the Hanford Project will be processed using the new program. The program corrects for the distortion inherent in infrared imaging systems and allows a pre-selected number of scan lines to be averaged to reduce the signal-to-noise level and increase the temperature sensitivity of the system. Imagery collected over the Columbia River on the Hanford Project is being processed with this program in an attempt to detect ground water entering the river from the 200 Areas.

Comparison of Davison 700 and Linde AW 500 Column Performance for Cesium Removal from PSN Solution

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

A sample of specially formulated, 20 to 40 mesh, beaded, Grace Davison Company 700 zeolite was used for column performance evaluation and comparison with Linde, 20 to 50 mesh AW 500 zeolite for removing cesium from PSN waste. A synthetic PSN solution was prepared for use in the study.

400 cc columns, 35 cm by 3.8 cm in diameter contained 310 g of AW 500 and 276 g of Davison 700. Column flow rate

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$$K_d = 6.5 - 2.31X_{NH_4} - 1.33X_K + 0.59X_K X_{NH_4} + 2.76Y - 0.94X_{NH_4} Y - 0.44X_K Y \quad (1)$$

in which

$$X = -1.5 + 10C$$

C = concentration of the competing ion

$$Y = -2.143 + 0.2857 (\text{clay } \%)$$

The following regression equation for soil No. 1 (reported earlier),

$$K_d = 3.7 - 1.31 X_{NH_4} - 0.84X_K + 0.48X_K X_{NH_4}, \quad (2)$$

can be used to verify the validity of this method. Introducing -1 for Y at 4% clay into Equation (1) will result in an expression with coefficients essential equal to Equation (2).

Statistical treatment of the data obtained for trace strontium adsorption showed no significant effect due to clay content. This was not expected; however, if additional studies with other soils and solutions confirm these results, prediction of strontium adsorption will be simplified.

A study was initiated to determine the effect of the disposal of a TM-30 caustic cleaning solution on the sorption of U by the Hanford sediments. Column breakthrough curves are being determined for the processed condensate waste. The U concentration was increased by a factor of 7 to facilitate the measurement of U in the column effluent. Loaded columns will be leached with TM-30 caustic cleaning solution to simulate the pulsing of the crib with this solution. The effect of TM-30 on ^{90}Sr and ^{137}Cs sorption by the Purex facilities will also be evaluated.

A study was continued to determine the effect of the prior disposal of an uncontaminated solution on the specific retention disposal of wastes to cribs.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Studies

A release of radioiodines to the river took place on June 19, following a fuel element cladding failure at KW Reactor. An estimate from preliminary data indicated that about 20 Ci of ^{131}I and 200 Ci of ^{133}I passed Richland following the fuel failure. The estimated thyroid dose received by the Maximum Richland Child was < 5 mrem. A release of radioiodines to the atmosphere from KW Reactor occurred on June 20 during disassembly of reactor piping. The estimated thyroid dose received by the Tri-Cities population was negligible.

Personnel Dosimetry

Field testing of the prototype TLD dosimeter for non-radiation workers indicates a routinely achievable accuracy of $\pm 20\%$ at the 95% confidence level in the dose range from 0.1 to 10 R. Several new phantoms were designed for studying the effects of chest tissue thickness in lung counter measurements.

Dosimetry Technology

The field study comparing dose estimates from film and LiF thermoluminescent dosimeters (TLD) within the present Hanford badge continued. Audit results indicate very good agreement with calibrated exposures. An extension of the current field program was suggested in order to more closely examine solutions to fading observed with the TLD.

Studies were undertaken to examine the response of selected TLD materials to neutrons. Significant modification of a recently purchased commercial TLD reader was undertaken in order to provide capabilities for semi-mechanized or mechanized readout of the new dosimeter for personnel routinely exposed to radiation.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Studies

(J. F. Honstead, Environmental Studies Section)

At approximately 11:00 P.M. on June 19, a fuel element cladding failure occurred at KW Reactor. The failure was such that only volatile fission products (radioiodines) were released to the river. An initial analysis of reactor effluent water indicated that perhaps up to 500 Ci of ^{131}I had been released to the river. Preliminary analyses of Columbia River water, however, indicated that about 20 Ci of ^{131}I and 200 Ci of ^{133}I passed Richland between approximately 9:00 A.M. on June 20 and 12:00 noon on June 21, 1968. The unusually high ratio of ^{133}I to ^{131}I (ca 10:1) occurred because of a short irradiation time given to the fuel prior to cladding failure. An initial estimate of the thyroid dose received by the Maximum Richland Child (2 g thyroid) via the drinking water pathway was 5 mrem (including ^{131}I and ^{133}I).

During the afternoon (ca 3:30 P.M.) on June 20 a release of radioiodines to the atmosphere from KW Reactor occurred during disassembly of process piping. No concentrations of radioiodines above detection levels were observed in the Hanford environs. The estimated thyroid dose received by the Tri-Cities population as a result of this release to the atmosphere was negligible.

Modification and testing of the EMA program is essentially complete. Parallel runs were made with the new EMA and EMB programs and compared to output from the existing EMA program. A second set of parallel runs is planned as insurance. The definition, sorting, criteria, contents and tentative format were prepared for 28 different monthly reports to be generated by the new program. A cost estimate is being prepared for the completion of this work after July 1. A special program was written to correct certain general and specific location codes and literals and to correct certain group and type codes and literals. These corrections, which are identified by EMA number, will be made in both the Table Master and Result Master

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area, and cannot be used for this purpose. A similar phantom is being constructed of wholly synthetic materials which will more nearly represent a standard man.

Scintillation lung counters were installed in the lead room at the whole body counter. These counters were calibrated for ^{238}Pu , ^{239}Pu and ^{241}Am .

A set of four 5-inch diameter by 3/8-inch thick NaI(Tl) scintillators were matched and mounted on a cot in a reproducible geometry for lung counting examinations. These detectors have about 45% resolution for 17 keV X rays. This counter will also be installed in the lead room for use in lung measurements of plutonium, americium and uranium.

Accurate control of the sled drive rate in the shadow shield whole body counter is essential to obtain accurate internal deposition information. An electronic controller was designed and fabricated utilizing integrated circuit construction and a stepping motor sled drive. Initial tests indicated no electronic noise problems. Analyzer dead-time drive rate, distance of travel, and fast travel controls functioned according to design. The controller is being installed on the portable shadow shield counter for final testing.

Washington State Inspectors made an inspection tour of Battelle-Northwest radiation exposure records on June 5. They were primarily interested in assuring that no BNW employee had exceeded any of the permissible exposure limits. The overall record system was also reviewed for them as this was their first official inspection.

An inspection of Hanford dosimeters was completed. Over 18,000 dosimeters were inspected. Of these, 198 had at least one component missing. All of the missing components were replaced.

There were 314 bioassay samples submitted for plutonium analysis during the month including 73 from new employees who had either previously worked with plutonium or who were assigned to work with plutonium in their assignment at Hanford. In

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This field-study program was originally scheduled to coincide with monthly film exchanges for April, May and June; the suggestion has been made that it be extended through July. We have noted that the decay from TLD is more than originally anticipated; therefore, a study will be conducted with the TLD fully annealed.

Personnel Dosimeter for Routinely Exposed Workers

The study of neutron response of several TLD materials was started. The accelerator was used as the neutron source. Exposures were made with TLD-100, -200, and -600, materials in air, B-10 covered, cadmium covered, bare, and in front of a phantom. The neutron energy was 0.73 MeV. Results taken with TLD-100 and -600 inside the B-10 ball placed on the moderator show no response at all. The B-10 ball must absorb essentially all intermediate and thermal neutrons impinging on it. Exposures inside the cadmium cover show a significant response to cadmium-capture gammas.

Preliminary results with BeO discs show a response to radium γ of about 1 reader unit per mR. The results were quite scattered, however, with a $\pm 30\%$ range on 10 measurements.

The TLD neutron dosimeter incorporating the use of TLD-100 and -700 ribbon was investigated. All exposures were made in the tin-cadmium filter system earlier described. The response per unit of fast-neutron dose was about a factor of 3.3 times greater than obtained from TLD-700 in the rhodium filtered dosimeter. The energy dependences for both systems were found to be virtually identical. A feature in favor of the tin-cadmium dosimeter would be a cost savings of about \$17.00 per dosimeter. Further development work will be continued when the Harshaw reader is back on the line.

Preparation of the Harshaw thermoluminescent dosimeter (TLD) reader progressed for the reader's dual role of involvement in experimental and routine work. Many of the controls were very hard to adjust because of temperature drift or extreme sensitivity of the controls themselves. Suppression of dark current,

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gain, and meter function were interlocked with the cycle-start switch to prevent lost or erroneous readings. The interlock safeguards all routine readings of the LiF extrusions read on the 10-second cycle. All non-routine programs for experimental readings require a momentary bypass of the lockout circuit. The circuit cannot be bypassed, however, when the meter is in the zero adjust position. Previously, it was possible to lose readings by leaving the meter in the zero position and running the reader through readout cycles. This fault is extremely dangerous on a destructively read dosimeter like TLD.

Tissue Equivalent Proportional Counter (TEPC)

The tissue equivalent proportional counter program continued with studies of the response as a function of the distance from the accelerator targets. This study was done to determine how close to r^{-2} dependence the TEPC measures. Exposures were made at 30, 40, 50, and 60 cm from the target. Results indicate that the measurements deviate only about 10% from r^{-2} dependence. The measurements are off in such a manner as to place the effective center of the TEPC farther away from the target than the geometrical center.

An exposure was conducted to 4.0 MeV neutrons using the new 1024-channel analyzer. The data show a clear resolution of the gamma-secondary peak from the electronic noise and the neutron secondaries. This is better resolution than was possible in previous experiments using the 512-channel analyzer.

Mössbauer Effect Studies

The 30 mCi ^{57}Co source arrived and allows a much more rapid accumulation of data than before--an adequate spectrum of the complete iron hyperfine structure can be obtained in 1 1/2 to 2 hours and shows an effect of about 2%. A DC restorer is needed to permit the system to accommodate higher count rates, thus taking more advantage of the stronger source. This instrument would improve amplifier resolution so that less of the unwanted background would be analyzed, thereby increasing the percentage effect observed.

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or contain little food. This suggests that fish moving seaward in midstream currents during darkness do little active feeding.

Columnaris Studies

Fifteen spring run adult chinook salmon were sampled at Rocky Reach Dam for columnaris exposure and antibody development during upstream migration. Thirty-three percent of the migrants had positive agglutinin titers similar to those observed at Astoria and McNary Dam. Gill and body lesion swabs from the salmon were negative. Water samples and coarse river fish were also free of isolates. The disease is probably of no consequence to the spring chinook run during upstream migration in the Columbia River. No evidence of columnaris exposure or definite increase in titer magnitude was observed during migration. If columnaris exposure in the tributary spawning grounds is minimal, the disease is not a problem for adult spring chinook salmon.

Semi-monthly largescale sucker samples are being taken at Hanford slough to determine the earliest presence of the pathogen in this section of the Columbia River. Evidence of the disease in river water is required at the earliest possible moment to determine initial exposure of our orally immunized juvenile salmonids. The average titers for largescale suckers have increased from 1:130 to 1:288 during the past three months; however, no other indication of new exposure has been observed. The percentage of zero-titered fish has remained constant and no organisms have been isolated from the fish. It would appear improbable that antibody production is triggered solely by increase in river temperatures. These observations suggest that many of the coarse river fish with columnaris titer may be carrying residual organisms that are activated by warm water temperature. This hypothesis would explain the sudden increase of the pathogen on river fishes when temperatures reach the low 60 values, and substantiates earlier observations on the presence of carrier fish.

Juvenile coho salmon and rainbow trout in the Laboratory have been fed daily for the past month with *C. columnaris*

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Instrumentation Services
(W. G. Spear, Instrumentation Section)

Wide Dynamic Range Radiation Monitoring with Surface Contoured Diode Detectors

Completion of the assembly of the experimental logarithmic count rate meter allowed evaluation of the instrument for the range from 10 to 10^8 counts/minute with achievement of an equivalent linear accuracy of $\pm 2\%$ full scale. Incorporated time division techniques, coupled with additional faster integrated circuit binaries, should increase the upper end of the scale to 10^9 counts/minute. The development approach provides a significant improvement in the dynamic range of the experimental radiation monitoring system.

Laundry Scaler Table Design Modifications

Completion of fabrication and bench tests for the improved laundry scaling table permitted installation of the system at the ITT/FSS Laundry Facility for field evaluation. Line noise, associated with the existing equipment at the facility, created an operational problem during the initial tests; however, addition of more filtering circuits eliminated the difficulty. The parts and materials required for the remaining scaler tables were also delivered to the facility for fabrication of the remaining units.

Laundry Monorail Modification for ^{147}Pm Monitoring

Fabrication of twelve signal conditioning, printed circuit cards and completion of layout of cards for the main tripping rate meters highlighted progress on the low energy beta monitoring portion of the system. Inclusion of a signal conditioning circuit comprising a 3-stage (integrated circuit) pulse amplifier, pulse height discriminator and low impedance line driver for each photomultiplier tube assures full independence of the counting systems with minimum interference from electronic noise sources. Output signals from the twelve line drivers trigger integrated circuit binaries and buffers that operate diode pump

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