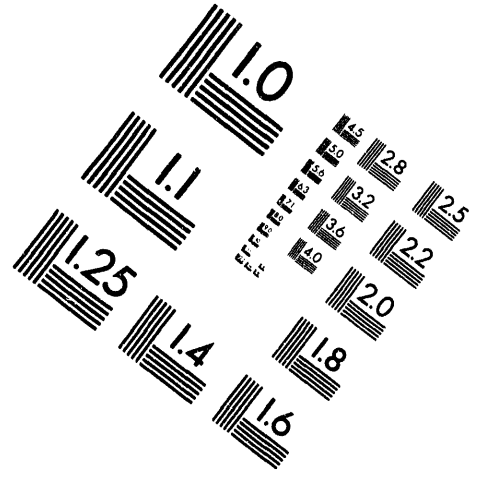
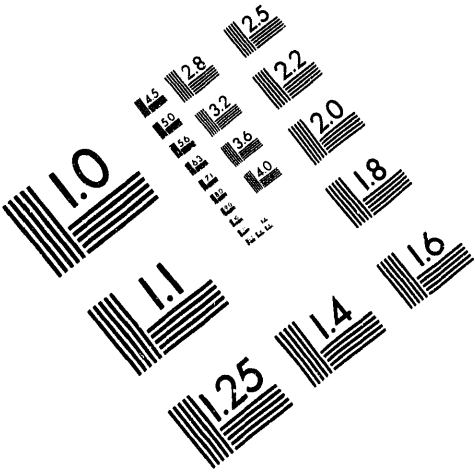




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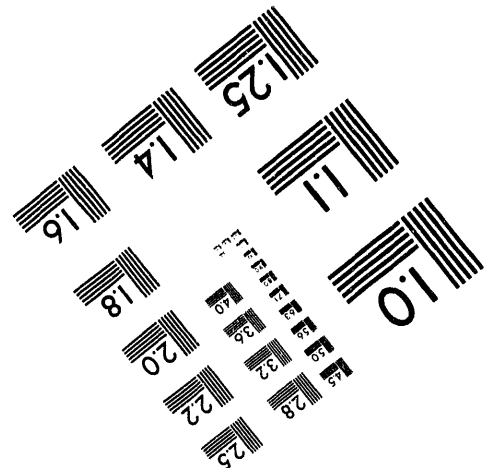
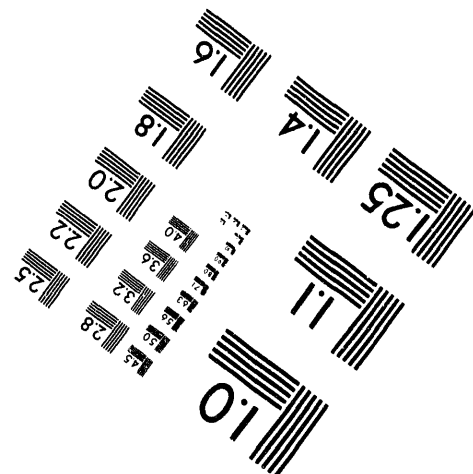
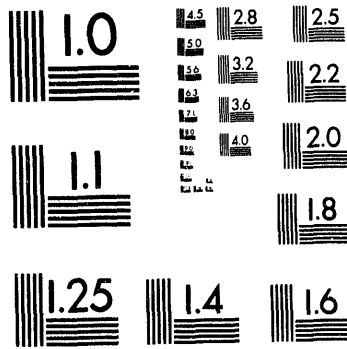
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**MONTHLY ACTIVITIES REPORT
FOR OCTOBER 1967**

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

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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
OCTOBER 1967

Division of Production
and
Hanford Plant Assistance
Programs

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F. W. Albaugh, Director

November 1967

PACIFIC NORTHWEST LABORATORY
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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

R. S. PAUL

PROJECT AEC SPONSORED PROGRAMS

Summary

Columbia River Studies

The results of a statistical study of Columbia River temperature trends were released on October 18 at the Sixth WSU Hydraulics Conference. The new John Day reservoir is under study for possible beneficial effects on flow regulation.

Mechanisms of Environmental Exposure

Measurements of radioactivity in 396 children were completed at Captain Gray elementary school, Pasco, Washington on October 19, 1967. Measurements began at the Emerson elementary school in Pasco on October 24, 1967, following lectures to the teaching staff and to each of the 18 classrooms in grades 1 through 6. Computer programming was started in October to determine the frequency distribution, mean, and median values of the environmental radiation dose values calculated last month from several thousand individual adult diet records. Response of game bird hunter-employees to a request for participation in a survey of game bird kills during the current season has been disappointing in spite of several articles in the project newspapers.

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DIRECT AEC SPONSORED PROGRAMS

Columbia River Studies

Effect of Reactor Effluent on the Quality of Columbia River
Water

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

Final computations in a statistical study of the temperature trends on the Columbia River were completed and, because of the timely opportunity, the results were presented at the Sixth Biennial Hydraulics Conference at Washington State University ahead of the formal reporting of the research. These results include the following conclusions:

- (a) The construction of dams on the Columbia River has had little effect on the annual average river temperature;
- (b) These programs have, however, had drastic effects on the timing of the temperature cycle tending to later and later dates as the reservoirs are developed;
- (c) Alterations of the natural temperatures by thermal additions persist for extended distances downstream and can be quantitatively assessed;
- (d) The Canadian dams now under construction may delay the peak temperatures of the impounded releases to the extent that lower temperature may result at the Hanford plant and on the lower Columbia as well.

These conclusions were also presented to the Columbia River Planning Committee at the annual research review October 19, 1967.

Computations related to the expected effect of the John Day Dam in partial dissipation of the plant heat release were started. The new reservoir will contain 2,450,000 acre feet and will delay transit of water through the system by about 14 days on the average. This additional transit time may have

interviews after it is analyzed through a statistically-designed computer program.

The response to the request for Hanford employee-hunters to participate in a season-long tally of game bird kills has been disappointingly low. Only one request for tally sheets was received in October. The total number of tally sheets distributed to date stands at 73. Environmental Monitoring personnel began collecting upland game birds adjacent to the Columbia River within the Hanford project for radiochemical analysis during October. Collection of road kills by employees of the Washington State Game Department is expected to begin in November.

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Summary

Mission 1

Standard N fuel (Fe and Si additions) after irradiation to 6500 MWd/T (0.75 at. % BU) is much more corrosion resistant in a 60-hour boiling water test than is the unirradiated fuel.

Six hundred forty-eight Mark IV fuel element components were measured and 626 were weighed.

A fast rise and fall pulser was developed which is capable of approximating a half cycle pulse of transducer frequency to as high as 120 MHz. This development eliminates the problems involved in evaluating high frequency, broad band, ultrasonic transducers.

An error in the N-Reactor computer simulation was found to be caused by the peak difference between plant and simulator gain. Pressure controller settings. Runs made after correcting the model show the simulated header pressure response to turbine trips to agree within 1% with actual

Mission 2

Three coprocessor driver elements that failed are being studied. Failure #39 appears to have occurred in the area of the inboard stop of the target locking hardware. Failure #40 appears to be associated with an inclusion through the clad at the contact area of the locked target support. Failure #41 is an end-associated type and a radial crack through the closure

The preliminary design of experimentally tested test sections to be used in this study was completed. A survey of inventories revealed that presently available materials will satisfy more than 75% of the requirements of this program. The remaining materials will be procured offsite; however, no delay in the program is expected. The detailed test section design is presently in progress, and construction will proceed with available materials as soon as possible.

Mission 8

Recommendations were made and forwarded to DGM on a comprehensive study to determine the capacity of a Gable Mountain Inland Lake to accommodate large volumes of reactor effluent. The 3-5 yr program would include flow system analysis, radio-nuclide and heat transport considerations.

Two fission product release experiments were completed using Zircaloy-2 clad inner elements of N-Reactor Mark II fuel assemblies. Rupture occurred at 1015 and 1050 with the extrusion of uranium foam, production of dark grey vapors and localized melting of the cladding. In both cases it was estimated that the amount of uranium within the cladding was sufficient to fuel the reactor for several hours after fuel.

Mass transport equations were derived for estimating release of gases from pools or basins of coolant following a reactor fuel meltdown. Initial application showed release of as much as 49% from the surface of the basin considered under conditions favoring release. Several conditions were

Mission 10

Termination of sodium dichromate addition to the deionized water cooling to two single pass tubes did not immediately affect the effluent concentrations of radioisotopes other than Cr^{51} . Three weeks later, the concentration of Co^{58} , an indicator of fuel jacket corrosion increased markedly. The data suggest that a one week addition of 1 ppm of sodium dichromate

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Examination of samples and corrosion probes installed in B-Reactor for 59 days showed small differences in aluminum and carbon steel corrosion between the side operating on 1/2 ppm dichromate and the side on 1 ppm dichromate.

Specimens of Zircaloy-2, Zircaloy-4 and Zr-2.5Nb alloys were exposed at in-flux and out-of-flux positions in an N-Reactor process tube. The fast fluence was 1.2×10^{20} , >1 MeV. Crud deposition was higher at the downstream out-of-flux position than at the in-flux position. Corrosion and hydrogen absorption were not markedly affected by the flux.

Laboratory evaluation of 22 polyelectrolytes used as filter bed conditioners showed that four of the 22 performed as well as Magnifloc 990.

A decontamination waste at 100N Area was shown to have no detrimental effects on cesium adsorption by soils.

All of the desired pre-test data on ground water quality, potentials and temperatures have been collected in the vicinity of the 100-NC trench disposal site. Reactor cooling water effluent discharge to the trench is scheduled to be started on October 27.

The Panametrics hygrometer probes installed at N-Reactor were removed and their present calibrations compared with the original calibration.

A portable neutron rem meter, for use in reactor areas, will include a 3-in. cadmium-loaded polyethylene sphere with a ^{252}Cf source. The light-weight electronics package will be powered by a six-volt battery with a unique 5-volt regulator.

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Mission 1 - Basic Production Mission

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

Open end fuel corrosion tests in boiling water of irradiated and unirradiated Mark I inner component wafer sections (Fe and Si bearing fuel) are being performed. The irradiated sections were obtained from the high exposure (~ 6500 MWd/T, 0.75 at.% BU), Mark I inner component that was rupture tested in the IRP facility with rather dramatic results. The August and September monthly reports contain a detailed account of this IRP test in which the defected test element blistered and ruptured after 10 hours of testing with a minimum amount of element damage and about 20 grams of fuel corroded. An unirradiated test element would have been expected to rupture in about 1 hr and a low exposure element (1000-2500 MWd/T range) even sooner. In the boiling water corrosion test 1/2 in. wafers of irradiated and unirradiated fuel sections were held in boiling water at atmospheric pressure in the same container. Weight losses were measured for two time intervals in the water, and the results indicated that irradiation has increased the corrosion resistance of the uranium. After 28 hr exposure to the boiling water test, the unirradiated control samples experienced 7.5 times more weight loss than the irradiated samples. During 32 additional hours of test the unirradiated samples experienced 4 times more weight loss than the irradiated samples. The irradiated fuel is significantly more corrosion resistant than the unirradiated fuel in spite of some cracks that unavoidably developed in the test surfaces during preparation of the irradiated test samples. The comparison corrosion test is being continued for an additional 30-hr period and will be followed by metallographic study of the test samples. It is believed that the improved corrosion resistance of the irradiated Fe and Si bearing uranium fuel is a consequence of the unique disposition of fission product atoms such as Zr and Mo. A composition of about 0.42 at.% or about 0.17 wt% Zr plus Mo content (~ 900 ppm Zr, 750 ppm Mo) has been calculated for the irradiated fuel in the test samples.

Vibration Test

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

The NRD fuel assembly vibration tester recently underwent final circuit modification, and is proving to be a reliable and stable piece of equipment. The need for a second tester is being reviewed.

Modification of 333 Building NDT Equipment

(T. J. Davis and E. F. Perrizo, NDT Systems Engineering Unit)

Design efforts are continuing to provide higher frequency bond testing for thin-clad fuels. A fast rise and fall pulser was developed which is capable of very closely approximating a half-cycle pulse of crystal frequency to as high as 120 MHz. This device, in conjunction with other recently developed circuitry, will compose a broad band, high-frequency, ultrasonic test unit for the thin-clad application. A vending source of PZT transducers with fundamental frequencies to 40 MHz was recently found, adding an option to the present stock of Li_2SO_4 broad band transducers.

Crack Tester

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

Capabilities of the bare-core deep crack tester were recently expanded to cover testing of CM1E and CM2E outers of the rod-in-tube configuration. This was primarily achieved through transducer modifications.

Zirconium Plating on Uranium

(R. W. Stromatt, Chemical Separations Unit)

Previous studies of a process for electroplating zirconium onto uranium metal from a molten fluoride salt plating bath have shown that the bath deteriorates in use to the extent that the quality of the zirconium plate changes for the worse during the course of a single run. It has also been found that the plating bath can be rejuvenated between plating runs by successive treatments with anhydrous HF and H_2 . It is believed that the bath deterioration problem could be avoided, and that

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that the two differ by less than 1% for this particular set of run conditions.

Statistical Assistance

(J. L. Jaech, Statistics Section)

A final report is in preparation summarizing the results of the analysis of the remeasured dimensional data from PT-A004. This test was designed to evaluate the effects of different heat processing treatments and concentrations of uranium core additives on dimensional distortion during irradiation.

A computer program is being written to facilitate analyses of fuel element failure data. The method of analysis is particularly adaptive to the situation where only a very small number of failures occur. Limits of uncertainty are also computed.

Graphs were prepared to aid in reaching decisions on when to terminate small scale testing of fuels from a failure viewpoint. Given k failures out of n fuels irradiated to E exposure, these graphs give the upper confidence limit on the failure rate at exposure CE , assuming that a Weibull failure model obtains.

A report was written describing the estimated equation for and efficiency of a calibration curve relating temperature gradient and power input. Guidance was given on the necessity for collecting additional data.

C-Basin profilometer data are being analyzed to determine the relative bias and precision of the instrument. The relative bias data consist of diameter measurements made on non-irradiated fuel elements at C-Basin by three operators each making three runs minus the "true" diameter measurements for each fuel element made at the 300 Area. This experiment is conducted routinely at the C-Basin.

A statistical analysis of data obtained for a group of irradiated fuel elements which had been subjected to severe self-support corrosion was started. Data include reactor tube characteristics, a set of height measurements for a random selection

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and analysis. Subsequent examinations have indicated the site of failure to be at the contact area of the locked stop of the target. An inclusion appears to be protruding through the driver clad at this location and metallographic study is being pursued to reveal its nature.

Failure #41 - Failure occurred to the upstream, locked end of the 3rd of 14 drivers in tube 2455 at a tube average exposure of 860 MWd/T. This failure was unique since it apparently ruptured following incident #40 while the reactor was down. Basin examination revealed a radial crack across the closure of the involved end. Disassembly and examination in Radiometallurgy indicates that the failure is an "end associated" type and that the area of greatest damage and the site of fuel corrosion is in the area of the cracked closure. Detailed metallographic examinations of a sector of the end closure centered on the radial crack are being pursued.

Mission 4 - Plutonium-238

Neptunium Impregnation Studies

(H. T. Fullam, Fission Product Chemistry Unit)

All work on the program has been completed.

Mission 5 - Other Isotopes

Code Development

ENDFB Evaluation Neutron Cross Section Library

(K. B. Stewart, Theoretical Physics Section)

A new ENDFB library was imported and is available for general use at Hanford. The new library tape contains the Category One isotopes (suitable for public release). Not all of the ENDFB isotopes are available on the tape; however, the remaining isotopes are expected soon and will be included on the Category One library tape as they become available.

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continued. Metallographic examination of 5 high power density fuel elements which operated near a maximum fuel temperature of 500°C is essentially complete. Optical photomicrographs and pictures taken with the electron microscope are being evaluated. It appears from a cursory look at the electron photomicrographs that all of the five fuel specimens have considerable porosity.

Aluminum Alloys

(R.R. Studer, Primary Fabrication Unit)

Two billets of X8001 aluminum alloy were cast to prove the operation of the direct chill casting machine. The direct chill casting machine is one in which molten aluminum is poured into a short chill mold in which a thin sheet of aluminum is withdrawn into a water spray where the billet solidifies. The result is a continuously cast aluminum ingot with a very fine dispersion of the second phase material. Previously, considerable difficulties were encountered in the operation of the casting machine. After modification to the mold and hydraulic drive mechanism, satisfactory operation was obtained. Metallographic examination of the cast ingots revealed many oxide inclusions. A temporary flux was made of equal parts by weight of NaCl and KCl pending receipt of a commercial aluminum cleaning flux. Fluxing the molten aluminum yielded clean castings, but some heats showed a visible evolution of hydrogen gas. A system for degassing with argon has been installed at the melt furnace and is ready for use on the 21 alloys to be cast. There has been a problem with the aluminum freezing in the transfer ladle thus resulting in short castings. Increasing the molten metal temperature lessens the effectiveness of degassing treatments. Hopefully, this problem has been alleviated by the construction of an electrically heated receiver which will maintain the alloys at their proper casting temperature during degassing, fluxing, and casting.

One of the X8001 cast billets was extruded into tubing. The product appears to have a satisfactory microstructure with no trace of the as cast eutectic network. Casting of the alloy billets for processing into tubing and sheet bar will proceed immediately.

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Detailed experimental run plans for this study are currently under preparation.

Mission 8 - Nuclear Safety

Reactor Effluent Disposal

(R. W. Nelson and J. R. Eliason, Geochemical and Geophysical Research Unit)

A study was made, and recommendations forwarded to DUN, to determine the hydrologic input data and analysis method requirements for evaluation of the flow aspects of discharging reactor cooling water effluents to a Gable Mountain Lake. The comprehensive program considered (3-5 year study) would require better field input data than are now available north of Gable Mountain and some work on methods development. Parameters to be evaluated would be: flow capacity of the Gable Mountain system, effects of such disposal on the Hanford project, and radionuclide and heat transport to the Columbia River.

The 100-BD disposal trench is completed and all of the necessary pre-test data on ground water temperatures, ground water potential and water quality have been collected. The test was scheduled to start October 30 at 9:00 a.m. The discharge rate will be increased daily until the maximum discharge rate is reached or until the trench infiltration rate limits the flow rate.

This test is designed to determine the long range effects of ground disposal of reactor effluent to trenches near the river. Field data will initially be collected on a weekly schedule and will later be reduced to a monthly schedule. Three riverbank spring sampling points have been established, and several wells near the test site are available for sampling.

Fission Product Release from Overheated Fuels

(D. L. Reid, Reactor Engineering Section)

Two fission product release experiments were completed in the laboratory using Zircaloy-2 clad inner elements of N-Reactor Mark II fuel assemblies. Induction heating of the fuel was

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Sections of the fuel showed about 40% extrusion of the uranium. Radiochemical analysis of the released material is in progress.

In both experiments it is estimated that the amount of uranium outside the cladding was sufficient to fill the annulus between the inner and outer fuel. For SR-2 the deeper Fiber Frax boat prevented overflow to the boat channel and diffusion through the boat. This created a sintered oxide mass on the upper half of the fuel that was about twice the original diameter of the fuel.

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

Needed improvement in the sensitivity of the "A" unit seismograph was obtained in a mocked up amplification scheme that increased the gain by 10. Operational amplifiers necessary to obtain this improved response are on order.

Remodeling of the Gable Mountain Seismic Station (623-B Bldg.) was completed.

Release of Fission Gases Under Reactor Accident Conditions
(A. K. Postma, Particulate & Gaseous Waste Research Unit)

Gaseous fission products dissolved or entrained in production reactor coolant following a serious meltdown of fuel would likely be released to the atmosphere to some degree and thus would constitute a radiation source. A possible way being considered to minimize consequences is to provide a large covered tank or basin to retain the effluent water during the period in which release of gaseous product would be greatest. Important considerations are to what degree noble gas isotopes of krypton and xenon would be released to the air from the liquid during the basin filling period and how can the release be minimized or controlled in the final disposal of the water. Several cases were considered and mass transfer equations derived to predict the rate of release under various circumstances. Assumptions were made regarding mixing and transfer coefficients which would favor the release from the water in the basin, since the initial design concept included some means, such as a

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Deionized Water Studies

(W. B. Silker, Water Reactor Corrosion and Chemistry Unit)

A delay in procurement of chemicals for reconstitution of process water from deionized water afforded the opportunity to conduct interim experiments with two single-pass process tubes. Addition of 1 ppm sodium dichromate to the deionized water was terminated in early August, and the effluent concentrations of several radioisotopes followed for about two months. Termination of inhibitor addition did not immediately influence the amounts of P^{32} , Sc^{46} , La^{140} , Zn^{65} , Np^{239} , or As^{76} by the effluent stream. Although perturbations were evident in the Cr^{51} data, one of which suggested that dichromate had been reintroduced to the system, a continued decrease in the effluent concentration of this isotope was noted. The decrease appeared to be exponential with time with a half life of about twenty days. The calculated release rate of inert chromium from the film was also exponential with time, with a release half-life of about twelve days. The effluent concentration of inert chromium shortly after cessation of dichromate addition was calculated to be about 3×10^{-10} g/ml, roughly three orders of magnitude less than the normal addition level.

The behavior of Co^{58} , an isotope formed by n,p reaction with the nickel in the fuel cladding, is of considerable interest as an index of fuel jacket corrosion. The concentration of this isotope remained at nominally low values for a period of three weeks subsequent to termination of dichromate addition. The concentration then increased and attained a new level which corresponded to a corrosion rate about ten times more rapid than before.

These data reinforce previous observations that a corrosion protection of fuel element surfaces might be afforded by occasional injection of the inhibitor into the coolant stream. The present results indicate that an injection period of one week inhibits corrosion for an additional three weeks. Although no spectacular relief would be obtained with regard to the amounts of Cr^{51} discharged to the river, implementation of such a practice would reduce the hexavalent chromium burden by 75%. It is

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column. It is speculated that the effectiveness of "Cat-Floc" would also be enhanced by presenting an uncorroded surface at the initiation of addition. It is recommended that investigations of the use of this additive be continued.

Film Studies

(W. B. Silker, Water Reactor Corrosion & Chemistry Unit)

To augment previous data, the films from four production run 03N fuel pieces were chemically removed and analyzed by activation analysis. The amounts of most elemental constituents were generally less than 10% of those found in films removed from exposed fuel. Gallium and nickel were both present in relatively large amounts. These results confirm the fact that most of the components present in the film on exposed fuel elements are not derived from impurities in the aluminum cladding material. The data will be incorporated into calculations currently being performed by BNW statisticians.

Process Technology

Be-Zr Braze Alloy Process Development

(R. G. Nelson, Primary Fabrication Unit)

A program has continued with the objective of producing high purity outer tube braze rings. This will be accomplished by starting with high purity elemental metals (Zr, Be, Sn, Fe, Ni, Cr) and consolidating and casting these materials using the vacuum consumable arc process.

During this reporting period, four skull heats were made. The melting results are as follows:

<u>Heat No.</u>	<u>Max. Amperes</u>	<u>Power on Time, Sec</u>	<u>Weight of Metal Poured, Pounds</u>
204	1800	45	0
204.5	2700	105	0
205	3500	145	9.75
206	3700	142	13.76

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4 in. from the downstream end, tapering to 4 mils thick at 60 in. Analysis of the hydrogen content of the metal with the hydride case removed (base metal) shows 80 ppm at 4 in., 129 ppm at 21 in., and 93 ppm at 60 in.

Tests using a platinum anode and Zircaloy-2 cathode immersed in specially prepared waters with a 3 volt applied potential at 95°C for ten days are continuing in an effort to find the particular ionic species in reactor process water that promote hydriding of Zircaloy cathodes. Using a base solution of NaHCO₃ pH adjusted to 6.6 with H₂SO₄ and specific resistivity of 6600 ohm cm at room temperature, various ions are tested by addition to the base solution. As previously reported, Na₂Cr₂O₇ increases the hydriding tendency of the water while sodium silicate tends to reduce the effect of the dichromate. Recent tests employing uranyl sulfate additions at 5 and 15 ppb concentrations indicate that it also enhances the hydriding tendency of the water. Stripping of the Zircaloy cathode after the test shows that less than 1% of the uranium passed by the cathode was precipitated on the cathode.

Graphite Oxidation Monitoring

(N. R. Gordon, Materials Compatibility Unit)

Channel 1880 KW, charged with monitors on March 18, 1967, was discharged September 14, 1967. The results indicated the presence of two locations of maximum oxidation rate, one at approximately 100 in. and another at approximately 200 in. from the front of the reactor. The first peak was 1.4%/KOD for graphite samples exposed directly to the gas atmosphere and 0.7%/KOD for those samples enclosed in graphite boats. The downstream peak was about 3.0%/KOD for the exposed graphite samples and 1.5%/KOD for the enclosed samples. TSGBF graphite samples were used exclusively. The first peak appears to be the result of an oxygen reaction with the graphite. The second peak is attributed to a CO₂ and/or H₂O reaction with the graphite.

Channel 1960C, charged February 12, 1967, was discharged September 8, 1967. The results indicated one sharp peak at the center of the reactor. KC graphite samples exposed to the gas

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mils deep; undercutting of the weld was found on the carbon steel side of the A212 Cs-304 SS weld samples. Heavy deposition up to 2700 mg/dm² was found on some of the carbon steel surfaces after removal from the system. Negligible deposition was found on the other alloys. The deposits were not analyzed but were assumed to be iron oxalate.

Half Plant Dichromate Test

(R. D. Weed, Water Reactor Corrosion and Chemistry Unit)

Sample holders exposed in the downstream end of two process tubes at B Reactor were examined for corrosion after 59 days exposure. One of the holders was exposed to process water containing 1 ppm Na₂Cr₂O₇.2H₂O and the other to 1/2 ppm Na₂Cr₂O₇.2H₂O. Data obtained from corrosion probes exposed in the rear nozzles of B Reactor were compared for the same time interval. Data are shown below.

<u>Alloy</u>	<u>Total Corrosion in 59 Days</u>			
	<u>1/2 ppm Dichromate</u>		<u>1 ppm Dichromate</u>	
	<u>Samples</u>	<u>Probes</u>	<u>Samples</u>	<u>Probes</u>
	<u>mils</u>	<u>mils</u>	<u>mils</u>	<u>mils</u>
8001 aluminum	1.07	1.37	1.09	1.0*
1100 aluminum	1.50	1.65	1.78	2.08
6063 aluminum	1.73	-	2.29	-
1020 carbon steel	-	0.74	-	0.78
A212 carbon steel	0.653	-	0.453	-

* For 15 days only

Although temperature and flow conditions were similar for the two tests, they were not identical and thus a comparison between low and normal dichromate conditions is only approximate.

Corrosion probes remaining in the test for 140 days in low dichromate water show corrosion of 8001 aluminum at 2.07 mils and carbon steel at 0.91 mil. Corrosion probes in the normal dichromate side failed before 140 days by water leakage. Additional sample holders were discharged recently for examination.

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Water Filtration Studies

(D. E. Olesen, Water & Wastewater Research Unit)

Laboratory evaluation of twenty-two polyelectrolytes used as filter bed conditioners was completed. Each polyelectrolyte was compared with Magnifloc 990 for resulting headloss and effluent turbidity on laboratory filters. Filters were operated at 8.0 gpm/ft². Alum was added at 18 ppm and polyelectrolytes at 0.03 ppm.

The following polyelectrolytes were found to perform as well as Magnifloc 990: Polyhall M-19; Purifloc N-11; Magnifloc 900N; and Mangifloc 985N.

Polyhall M-19 is listed at \$0.76/pound in truckload lots, F.O.B., Charlotte, N. C. Purifloc N-11 is listed at \$1.17/pound for lots greater than 2,000 pounds, F.O.B., Midland, Michigan. Price quotations have not been received for the Magnifloc products.

Each of the other eighteen polyelectrolytes allowed turbidity breakthrough to occur before the end of the filter run.

Soil Chemistry

(B. F. Hajek, Water & Wastewater Research Unit)

Previous studies have shown that a decontamination waste generated at 100-N Area could be disposed to the ground; however, the effects of this waste on cesium adsorption from subsequent waste disposal was not known. Soil column studies conducted this month have shown that this waste had no detrimental effect on cesium adsorption from a Cs¹³⁷ traced river-water solution.

Strontium and cesium equilibrium distribution coefficients were determined for soil adsorption from a cleaning solution containing a foam-aid and a detergent (Prime cleaner). The coefficients determined were 1047 ml/g for strontium and 682 ml/g for cesium. The cleaning solution was made up of 1 part Prime, 1/5 part foam-aid and 5 parts tap water. The strontium coefficient is about 50 times greater than for tap water alone. The cesium coefficient was about the same.

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

A DUN Research and Engineering authorized increase in the upper weight limit of the neutron rem meter permits use of a 9-in. cadmium-loaded polyethylene sphere in the instrument being developed. Design was started to incorporate a Hanford "BFQ" BF₃ tube and fabrication of the sphere was planned for the technical shops. Suitable cadmium foil for the assembly was ordered.

Completion of a voltage regulator design for the instrument promises to enable the electronics to be light weight, with all circuitry operating from a 6 volt battery or the 5 volt regulator. The latter will control the output voltage constant to within 4 millivolts at 20 milliamps for battery voltage variation between 5.2 to 7.5 volts. Output impedance of the regulator is approximately 0.1 ohm for a 0 to 50 mA load current variation.

Temperature test of this critical circuit indicated a total drift of only 2.7% for a temperature change of 70°F, equivalent to a temperature coefficient of 0.022%/°C. This should be quite adequate for the application at hand.

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...of the ... of the ...

In the study of electrodeless conductivity systems for ...
The fact that ... suspended in organic ...

... and ...

... decaying step ...

... technique can ...

... of the ...

Preliminary studies of the vaporization of UCl_3 were ...
continued in establishing techniques to be used for similar ...

The effect of polyethylene surfaces on the alpha radio-
lysis of plutonium nitrate solutions was examined and found ...

... of $NO(V)$...
... which has been found to catalyze the HNO_3-HNO_2 ...

... and methods of preparation of the nitropropane ...
... made of the radiation ...

MISSION 1

where the experiments will be conducted.

Plutonium entrained in air moving at 10 cm/sec over an evaporating plutonium nitrate solution represented about 10^{-7} wt%. During a subsequent drying period less than 10^{-7} wt% per hour was entrained.

Mission 9

Removal of uranium, thorium, and strontium from Purex solids with organic complexants at high pH.

The flame velocity in a 6-in. packed bed of molecular sieves was reduced over that in the gas space but the detonation was not prevented. Helium dilution increased the flame velocity.

Experiments with mass baffles indicate that the current design meets the needs.

Experiments are being carried out to obtain information to date indicate that the volume of waste should be a part of the program.

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Mission 1 - Basic Production Mission

Neptunium Chemistry

(J. L. Swanson, Chemical Separations Unit)

Studies continued of variations in the preparative steps used in converting 1-nitropropane into the material (of unknown composition) which accelerates the nitrous acid oxidation of neptunium(V). In studies of the effect of 1-nitropropane concentration upon the yield of the accelerator, lower yields result at higher concentrations; the concentration of accelerating material formed remained fairly constant on increasing the concentration of added nitropropane from 0.04 to 0.4M (at comparable acidities and temperatures).

Exposure of solutions containing rate-accelerating material obtained in different yields to a given dose of gamma radiation did not result in destruction of a given amount of the accelerating material. Rather, the decrease in concentration of accelerating material was found to depend on the concentration of the nitropropane used in the accelerator preparation procedure. At a given concentration of added nitropropane, a given radiation dose gave a constant fractional decrease in the concentration of accelerating material, regardless of the yield in which the accelerating material was obtained in the preparation employed. This suggests that the effect of the radiation is to destroy some material which is a precursor to the accelerating material in a sequence of reactions leading from the nitropropane to the accelerating material.

The radiation experiments indicated that after a dose of 10^5 R (comparable to that to which the aqueous phase is exposed in the extraction section of the HA column), no rate-accelerating material remained unless the original nitropropane concentration was greater than about 0.007M. The most highly concentrated solutions yet prepared (0.4M original nitropropane), after being diluted by factors of 40, 30, or 20 and subjected to a dose of 10^5 R, still contained sufficient rate-accelerat-

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(96-100%) and reproducibility are within the $\pm 2.0\%$ T range of accuracy currently inherent in the Perkin Elmer 521 IR spectrophotometer. The detailed procedure was written up for distribution to those concerned.

Alpha Radiolysis of Plutonium Nitrate Solutions

(J. C. Sheppard, Heavy Element Chemistry Unit)

Work continued on the alpha radiolysis of plutonium-nitric acid solutions. The table summarizes the new data obtained.

Gas Composition

Expt	HNO ₃ M	G _{Corr.}	%			CO ₂ or N ₂ O	Comments
			H ₂	O ₂	N ₂		
4	8.0	0.042	60	22	<5	8	
6	4.0	0.07	81	--	--	10	Polyethylene rings ^a
7	4.0	0.10	Not Analyzed				Polyethylene sheet ^b

- a) Area corresponds to 570 cm².
- b) Area corresponds to 504 cm².

The data indicate that no significant increase in the rate of radiolytic gas generation should be expected when polyethylene containers are used. The area of the polyethylene presented is 2-3 times that which would be presented by a polyethylene bottle. Assuming that radical reactions at the surface can be neglected, the surface to volume dose ratio is given by

$$\frac{R(\text{cm})}{D(\text{cm})} \approx \frac{4 \times 10^{-3}}{D(\text{cm})}$$

where R is the range of the alpha in water and D is the diameter of the container. If a G value of about 3 is assumed for polyethylene, then the ratio of gas generated at the surface to that in solution will be less than 0.1. This is consistent with the experimental observations.

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Thermodynamic Studies of the Actinide Halides
(J. D. Christian, Heavy Element Chemistry Unit)

Experimental work continued to measure some of the thermodynamic properties of certain actinide element halides, to provide a basis for the development of advanced, non-aqueous processes for the separation of plutonium and americium, americium and curium, etc. Recent experiments on the vaporization of UCl_3 indicated contamination of the UCl_3 sample with uranium oxide.

Some fresh UCl_3 was prepared and careful consideration given to cleaning the flowing argon. An improved temperature controlling system for the furnace was constructed.

Zirflex Dissolver Studies
(W. J. Van Slyke, Engineering Development Unit)

Two dissolutions were conducted and have indicated that the use of $7.5M NH_4F + 0.7M NH_4NO_3$, while not as effective as would have been predicted from the laboratory data, does provide an effective Zircaloy oxide penetration and multiple cladding penetration in four to five hours. This technique can, therefore, be used to expedite the Zircaloy decladding step. Visually, it appeared that the uranium was more severely attacked by the more concentrated NH_4F solution. These two runs completed all of the Zirflex dissolution tests which had been scheduled.

Effect of DBBP Contamination on 1C Column Performance
(G. L. Richardson, Engineering Development Unit)

The effect of DBBP (dibutylbutyl phosphonate) contamination in the Purex TBP solvent on the performance of a 1C column was reported in August. The major effect was a 50-fold increase in waste losses at a contamination level of 5% DBBP. Equilibrium data recently obtained explain this loss effect and can be used to set operating conditions that will restore the column's performance. In the absence of DBBP, the laboratory work showed that the maximum distribution ratio (E_a^0) was 0.80 at $50^\circ C$. The E_a^0 increased in a regular fashion with increased addition of 25% DBBP to 0.96 at 5% DBBP + 24% TBP. The aqueous-

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Ultrasonic Liquid Level

(O. H. Koski, Process Systems Development Unit)

An ultrasonic liquid level measurement system for Purex product tanks was under development as a substitute for capacitance devices which give uncertain results when exposed to foam.

The digital readout failed during laboratory testing and was repaired by the manufacturer. The repaired unit gave a 0.04 in. shift on warm-up and stabilized within ± 0.01 in. over a 72-hr period.

Dibutyl Butylphosphonate Extraction of Np and Pu from Purex LW

(W. W. Schulz, Materials and Process Chemistry Unit)

A plant test of this process is planned by ARHCO. In anticipation of this test various process items are being examined. For example, the initial plant test will most likely be done in existing tankage not particularly well suited for clean-cut separations of aqueous and organic phases. Consequently, a heel of aqueous phase from the previous contact will be carried forward to each succeeding extraction, scrub and strip contact. Laboratory results indicated that such heels will not affect plutonium and neptunium recovery operations. The presence of a heel of LW raffinate during the oxalate scrub step, however, will eliminate most of the $ZrNb^{95}$ DF potential of the process.

Additional batch equilibrium data for the distribution of uranium between TBP-DBBP-NPH solutions and 0.01M HNO_3 solutions were obtained in other work. The data are intended for use in McCabe-Thiele plots to determine conditions required for adequate stripping of uranium from such solvents in the Purex plant 1C column. Earlier results indicated that 1C column performance might be significantly affected if the mainline solvent were contaminated with DBBP.

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The recently completed gamma spectral study of eight process samples from the Waste Reclamation Facility using both Si(Li) and Ge(Li) high resolution diode detectors is being analyzed further. Ultimately these studies should lead to considerably improved process instrumentation for measuring low level concentrations of Pu²³⁹ and Am²⁴¹. Such in-line instrumentation systems are needed to provide control and possible measurement of plutonium concentrations over a range of 0.01 to 0.5 g/l in the CCW line and a range of 0.001 to 0.05 g/l in the CAW line.

A tentative design of a flexible advanced solid state dual channel spectrometer using integrated circuits was established and purchase orders were placed for the necessary electronic components.

This instrument represents a significant advance over the successful single channel model in that it will allow compensation of interference from Zr-Nb, Am²⁴¹, ruthenium, bremsstrahlung and other gamma radiation. This instrument should improve the present plutonium measuring range, typically 0.5 to 50 grams, to significantly lower values and should also provide advanced capability for work in new areas that may develop in connection with future Pu²³⁸, Cm²⁴⁴ and Np²³⁷ products.

Purex Simulation

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

A hybrid model of the Np loops in the Purex process was developed. Runs were then made in order to determine the effects of 4, 8, and 16 hour Np spikes. From these runs predictions can be made of the magnitude of the spike which would be required in order to measure the Np levels in the process tubes and tanks. The best sampling procedure required in order to follow the spike through the process and measure the effect of recycle can also be determined. Further work will be done upon completion of the actual spike test in the plant. The model will then be updated in order to match the experimental data and to gain a better understanding of the process.

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3) oxidation disintegration (i.e., mixed acid, basic dichromate, etc.). Although graphite is only slowly oxidized, the graphite matrix tends to fall apart in a strongly oxidizing medium and thus the mixed acid dissolution method could possibly be used to disintegrate the graphite prior to leaching.

A filtering device is being designed to facilitate separation of the graphite particles from the leaching solution.

Neptunium-Plutonium Solvent Extraction Studies

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

A program of tracer-level experiments has been carried out to scout the potential for recovery, partition, and purification of Np^{237} and Pu^{238} by solvent extraction techniques. Both DBBP and TBP have been evaluated. Results to date have been extremely promising. Coextraction (from dichromate oxidized solutions) is easily accomplished as is partition by reduction of the plutonium to Pu(III) with hydroxylamine nitrate.

Pulsed Current Migration

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

Five more pulsed current migration experiments were carried out to help evaluate the effect of various pulse forms on isotope separation. However, the results of the experiment were invalid (and somewhat confusing) due to operational difficulties experienced with the oscilloscope. Therefore, the experiments will be repeated.

Mission 8 - Nuclear Safety

Criticality Safety Evaluation of Proposed Shipping Cask for Irradiated Fuel Elements

(R. C. Lloyd, Critical Mass Physics Section)

Equipment was assembled and plans made for experiments that will determine criticality safety of a proposed shipping cask for irradiated fuel elements. Neutron multiplication measurements will be performed with the uranium in geometry similar to that of the fuel in the actual transfer cask. A

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Mission 9 - Waste Management

Purex Solids Studies

(F. P. Roberts, Fission Product Chemistry Unit)

Earlier it was reported that leaching of rare earths and strontium with organic complexants at high pH was only partially complete. Further investigation has shown that if the solids are treated with 0.1M DTPA (diethylene-triamine-pentaacetate) at pH 5.5 for 30 min at 90°C and the pH then raised to 12, not only do the rare earths and strontium dissolve but also most of the barium.

Under the same conditions NTA (nitrilotriacetic acid) and tartaric acid dissolves all of the rare earths but only 75% and 81% of the strontium.

This work was done with synthetic IWW. A full level test is under way using plant solutions in B-Cell of the High Level Radiochemistry Facility.

Hydrogen-Oxygen Safety

(W. E. Willingham, Engineering Analysis Unit)

The first series of runs with hydrogen-oxygen gas mixtures with a packed bed of Linde AW-500 Molecular Sieves, length 6 in., was completed. This length was not sufficient to prevent a detonation from taking place but the velocity over the packed section was apparently reduced. Additional runs were made using helium as a diluent; in each case there was some increase in velocity of the waves.

Organic Waste Disposal

(E. A. Coppinger, Engineering Analysis Unit)

Evaluation of the organic waste disposal problem continued. The problem was essentially defined in terms of volumes and activity. The information available to date indicates that in-plant improvements to reduce the volume discharged should be an integral part of the study. For example,

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than 70% of the neptunium extracted from the 0.1M H⁺ solution. These first experiments thus demonstrate process feasibility but optimum operating conditions remain to be defined.

Capacitance Probe Development

(O. H. Koski, Process Systems Development Unit)

The development of a radiation resistant capacitance probe for waste cask liquid level measurement has been undertaken. An electrodeless process for plating nickel and copper on the inside of alumina and mullite tubes was found to be successful. Nickel plating of six-foot probes is more difficult than short (30 in.) probes due to the unfavorable area to solution volume ratio combined with poor mixing. Copper plating was found to be simpler process-wise and to yield a better product. Nickel plating on copper improves soldering characteristics and provides an oxidation resistant surface. Probes plated include four 6-ft probes, two 4-ft probes, and a 30-in. probe.

Process Technology

Organic Layer in Tank 112 BY

(W. W. Schulz, Materials and Process Chemistry Unit)

Tank 112 BY contains approximately 700,000 gallons of neutralized Uranium Metal Recovery Plant waste. Eventual in-tank solidification of the contents of this tank is planned as part of the over-all Hanford Waste Management Program. Tank samples and photographs of the tank interior confirm that an organic material covers part of the aqueous surface. This material may complicate solidification operations because of possible unknown hazards. Determination of the composition, properties, and reactions of the organic material is under way to define the magnitude of such hazards.

Gas chromatographic analysis shows the material to contain about 83 vol% TBP diluted with degraded kerosene, probably Shell Spray Base. Differential thermal analysis confirms the gas chromatographic identification; endotherms (in air) were observed at about 260 to 270°C and at about 290°C. The latter probably corresponds to the boiling point of the TBP. The organic

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some Np and fission products. This was material from normal process fore- and tail-cuts, 234-5 Building calcination wastes, and C-Cell wastes.

Sampling System for Purex HAW

(J. Dunn, Process Systems Development Unit)

A new sampling system is being fabricated which incorporates a preliminary partitioning cycle to reduce the fission product content of the HAW stream to 1/10 prior to laboratory analysis. The prototype jumper will be cold tested in EDL-102 before being transferred to the separations plant for use.

Bank Tank Development Work

(J. Dunn, Process Systems Development Unit)

Installation of the pH monitor in the glass mock-up of the Plutonium Reclamation Facility bank tanks for continuously neutralizing the CAW stream is progressing. The neutralization control will be dual; one phase will produce a feed of pH 0.75 to the W-2 extraction column for plutonium and americium recovery and the second step will adjust the waste stream to pH 2.5 prior to final storage.

Soil Chemistry

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

Studies of the removal of Pu²³⁹ and Am²⁴¹ deposited on soil from CAW wastes by the passage of DBBP-CCl₄ (30-70%) through the soil were repeated. Gravity flow was used instead of pumping and the slower flow rate permitted infiltration of the high-salt, acidic waste neutralized to pH 2.65 with NaOH. The waste had been spiked with plutonium and americium. Only about 10% of the activity of the influent was removed by the soil. A solution of DBBP-CCl₄ (30-70%) was then allowed to flow through the soil by gravity. Two column volumes of DBBP-CCl₄ removed essentially 100% of the Am²⁴¹ and 85% of the Pu²³⁹ on the soil.

The removal of radionuclides from soil by partially neutralized hydroxyacetic acid at different dilutions was studied.

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A break in the well casing permitted waste to contaminate the inside of the well casing.

Land Release Studies

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

Simulation analysis of the irrigation of about 29 sections of land north of the 300 Area and 10 sections of land west of 200 West Area resulted in predicted rises of 23 ft beneath the 241-SX tank farm, 11 ft beneath 200 East Area and 4 ft beneath the 300 Area. Steady-state analysis indicates that a drainage system will be needed in the irrigated region north of 300 Area because of the close proximity of the land surface to the predicted water table caused by the 400-ft Ben Franklin Dam pool.

The location of the "irrigated" land in Upper Cold Creek Valley is outside the boundaries of the mathematical model that simulates the Hanford groundwater flow system, so effects of irrigating this land must be transferred to the model boundaries. The finite difference solution of the model predicted that surface flow would develop at this boundary. As the water flows across the land surface, increments of it would seep back into the ground until all of the water had seeped back into the ground a finite distance from the boundary. Since to simulate this effect in the model would mean expensive trial and error solution, and since results predicted from the first solution would be on the high side, the problem was not rerun. Subsequently, with the "irrigation" of the 200 West project, it was found necessary to approximately balance the surface and groundwater flow and rerun the Upper Cold Creek irrigation case. This resulted in a lowering of the original predictions. The new condition predicts rises of 38 ft beneath the 241-SX tank farm in 200 West Area, 7 ft beneath 200 East Area and 4 ft beneath the 300 Area.

The total predicted rises, resulting from the 400-ft Ben Franklin Dam pool and irrigation of Horn Rapids Triangle, Upper Cold Creek, 300 and 200 West irrigation projects, were 80 ft beneath the 241-SX tank farm, 40 ft beneath 200 East Area and 12 ft beneath the 300 Area.

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HWI-620

ENVIRONMENTAL ASSISTANCE AT THE HANFORD PLANT

Summary

Environmental Evaluation

Initial samples of upland game birds from on-plant confirm their potential significance as a source of P^{32} and Zn^{65} intake.

The concentration of aliphatic hydrocarbons in river water from the Hanford River, the Richland River, and Richland pumping plant, remains approximately identical. This indicates there is no significant contribution from the plant. It is estimated that approximately 12,000 gal of fuel oil were released to the river before the flow of oil from the river banks discontinued.

Personnel Dosimetry

Positive measurements were obtained using Whole Body Counter examination techniques to determine Am^{241} in an employee involved in a contamination incident. This case emphasized the importance of accurate monitoring for Am intake and the Am deposition.

Dosimetry Studies

Development work on a portable neutron survey instrument continued as did studies on a simple personnel dosimeter employing thermoluminescent material.



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BWL-620

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluation

(J. F. Honstead, Environmental Studies Section)

Routine on-plant collection of upland game birds has been added to the environmental surveillance program as a follow-up to work performed under the "Mechanisms of Exposure" R&D program indicating game birds as a potentially significant source of Zn⁶⁵ and P³² intake. Initial results from flesh of several pheasant and chukar partridge taken near the river showed both P³² and Zn⁶⁵ concentrations several-fold higher than in whitefish or ducks collected during the same period.

Routine river water sampling at 100-D Area was discontinued with the suspension of pump operation at 181-D. Richland is the first location downstream of the reactor where representative continuous water samples are now obtained.

Oil Contamination in Columbia River at 100-N

(J. F. Honstead, Environmental Studies Section)

Evaluation of problems associated with fuel oil entering the Columbia River at 100-D continued. Results of analyses of river water samples from four locations (Priest Rapids, 100-D, 100-F and Richland pumping plant) continue to indicate that little or no oil is reaching the Columbia. Two of the sampling sites, 100-D and Richland pumping plant, have been discontinued since the results indicate no variation in the concentrations found above the plant and below the plant. Assuming that the loss of fuel oil from the river banks averaged 50 gallons per day during the last eight months, approximately 12,000 gallons were released. Since the original loss from the fuel oil system was estimated to be at least 80,000 gallons, a sizable inventory may remain.

Dosimetry Studies

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

Thermoluminescent Personnel Dosimeter

Studies to develop a simple personnel dosimeter for use on employees who do not normally receive a significant radiation exposure continued with fabrication of several prototypes. The dosimeter uses two thermoluminescent-Teflon discs with Li^7F as the active material. One of the discs is shielded from sunlight by black tape, the other is shielded by 0.018 in. of stainless steel. The discs are held in place by a polyethylene holder. The prototype dosimeters were exposed in groups of five to 500 mR of filtered and fluorescent X rays of energies from 10 keV to 100 keV to determine the energy response of this filter system.

The energy response of the lightly shielded disc is quite flat. From slightly less than 20 keV to 100 keV the response varied no more than about 10%. In previous measurements with the discs, the response to 100 keV photons was found to be the same as the response of radium γ and Co^{60} radiation. It is reasonable to assume that the response is flat up to 1.25 MeV. The disc shielded with 0.018 in. of stainless steel showed no response to photon radiation below about 20 keV. Above 20 keV the response rises until it is equal to that of the unshielded disc at 100 keV. Below 80 keV the ratio of the readings of the two discs can be used to indicate the average energy of the incident photon radiation. For these exposures the dosimeter was placed behind the security credential which is nominally 0.025 in. thick.

Portable Neutron Survey Instrument

The tissue-equivalent proportional counter work continued. A comparison between the measured dose distribution in LET for 0.75 MeV neutrons compared very well the same quantity calculated for water. The peak in the D(L) curve comes at about 93 keV/ μ for water in the calculated curve and at about 100 keV/ μ in the measured curve for the tissue-equivalent gas.

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upstream. At least 80% of the salmon on both the Little Wenatchee and Okanogan were spawned out when sampled. The presence of large numbers of exposed fish which survive to spawn suggests that columnaris exposure and the development of high antibody titers may be protecting these fish from columnaris induced mortalities during migration.

Adult Chinook salmon (Jacks only) transported from Priest Rapids Dam are being tested to investigate their tolerance to sudden exposure to warm temperatures. Early results show that fish acclimated to 15°C respond to acute thermal challenges as follows:

- 30°C Dead in 12 minutes
- 28°C Dead in 30 minutes
- 26°C Dead in 24 hours
- 24°C At 48 hours the fish survive with no apparent stress

The section of the Columbia River from Richland to Priest Rapids Dam was surveyed from the air on September 28 and October 16. The Washington Department of Fisheries was represented on the first survey, hoping to find some early spawning in this section of the Columbia River to explain, at least partially, the apparent loss of salmon between McNary Dam and Ice Harbor-Priest Rapids Dam. No spawning activity was observed on either flight. Start of spawning in past years usually began between October 8 and 21.

Bioassay of liquid chemical wastes stored in tanks at 100-N for possible river discharge was conducted with juvenile rainbow trout. The wastes included a mixture of phosphoric acid, citric-oxalic acid, sodium hydroxide, potassium permanganate and oxides removed from pipe surfaces. Fish exposed for 48 hours to different dilutions ranging from 1:60 to 1:6000 suffered neither mortalities nor apparent stress.

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