AIM

Association for Information and Image Management

1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

Centimeter

Inches

MANUFACTURED TO AIM STANDARDS
BY APPLIED IMAGE, INC.

O

FOR JANUARY 1968
DIVISION OF PRODUCTION
AND
HANFORD PLANT ASSISTANCE
PROGRAMS

CLASSIFIED

DECLASSIFIED

Classification Canceled and Changed To
Classified

DECLASSIFIED

By Authority of CG PR 2 (PR 24),
JS Lewis, 2-10-94.
By Chris Mally, 2-15-94.
Verified By J.E. Savely 2-23-94

BNWL-696
C-65, Plutonium Production
(Special Distribution)

This document consists
of 89 pages. Copy No. 1
of 1 copies. Series 1

PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
JANUARY 1968

Division of Production
and
Hanford Plant Assistance
Programs

by the
Staff of Battelle-Northwest
F. W. Albaugh, Director

February 1968

Classification Canceled Change to

CONFIDENTIAL

By Authority

By

MASTER

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

DECLASSIFIED

This document contains Restricted Data as defined in the
Atomic Energy Act of 1954. Its transmission or the disclosure
of its contents in any manner to an unauthorized person is
prohibited.

Route To:	P.R. No.	Location	Route Date	Signature and Date
<u>20 - [illegible]</u>				

G [illegible]

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED ^{db}

DECLASSIFIED

-2-

BNWL-696

TABLE OF CONTENTS

	<u>Page</u>
Direct AEC Sponsored Programs - Summary	3
Direct AEC Sponsored Programs	4
Assistance to Douglas United Nuclear - Summary	7
Assistance to Douglas United Nuclear	12
Assistance to Atlantic Richfield Hanford Co. - Summary.	43
Assistance to Atlantic Richfield Hanford Co.	49
Technical Assistance to the Hanford Plant - Summary . .	76
Technical Assistance to the Hanford Plant	77

DECLASSIFIED

BNWL-696

PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

R. S. PAUL

DIRECT AEC SPONSORED PROGRAMS

Summary

Analysis of Columbia River temperature trends indicates conformity with those previously reported. Despite unusually high average temperatures, the temperatures of the upper and lower extremes continued to converge on the mean. The construction of the John Day Dam is expected to have little temperature effect other than a delay of ten days in the timing of the annual temperature peak.

Measurements of radioactivity in 465 children were completed at the third Pasco elementary school on January 12. Whole-body counting at the fourth school began on January 24, following lectures to the staff and 17 classrooms.

The whole-body counter was recalibrated for K^{40} and Zn^{65} in late January. Only small modifications in the calibration factors over those currently employed are expected.

Washington State Game Department personnel have supplied sufficient pheasant and quail samples shot near the Columbia River for comparison of radionuclide content between species and collection of these samples has been discontinued. The Department of Game continues to supply "road-kills" for radiochemical analysis in exchange for X-ray data on the number of birds which contain shot. A game-bird questionnaire for mailing to a statistical sample of Tri-City hunting license holders was drafted in January.

DECLASSIFIED

-4-

BM 100-100

DIRECT AEC SPONSORED PROGRAMS

Columbia River Studies

Effect of Reactor Effluent on the Quality of Columbia River Water

(R. T. Jaske, Water Quality Systems Analysis Unit)

The analysis of Columbia River temperature trends by the least squares fitting of sine curves to the annual data sets was continued with the close of the 1967 calendar year. The following have been observed:

1. The average temperatures for upstream dam projects continue to fall close together, in the vicinity of 11.3°C . This compares with the all time high in 1958 of 12.1°C at Rocky Reach.
2. The upper extremes of the fitted curves for the upstream projects show close grouping and a continuation of the decline previously reported. Typical upper extremes were 18.2°C . This can be compared with the average of the ten years 1938-1948 of 18.8°C .
3. The lower extremes of the fitted curves indicate a continuation of the trend to higher values. Upstream projects reached minimums of about 4.50°C , some 1.5° higher than in the 1938-1948 period.
4. Downstream projects reflect extremely high Snake River temperatures existing in 1967. Bonneville Dam averaged 0.3° higher than Richland, the first time since 1964, and higher than every preceding year except 1958 when the average reached 13.20°C . The Bonneville average for 1967 was 13.00°C .

The conclusions derived from the analysis of these trends indicate continued confirmation of trends previously reported. The extremes continue to converge on the mean, and the arrival of the peak temperatures throughout the entire system shows a high degree of correlation. Temporal increases from the operation

[REDACTED]

[REDACTED]

Hunting of three species of game-birds along the Columbia River by personnel of the Washington State Department of Game was discontinued at the request of Battelle at the end of January. Sufficient pheasant and quail samples have been obtained, while chukar samples apparently are too difficult to collect in a reasonably short period of time for intra-species comparisons of radionuclide content. The Department of Game continues to supply "road-kills" from eastern Washington for radiochemical analysis in exchange for X-ray data on the numbers of these birds which contain lead shot. To date, 51 such birds have been X rayed by personnel of the External Dose Evaluations Unit, with 14 found to contain shot. The results of the radiochemical analysis of the first batch of road-kills are not yet available. Preliminary indications are that the only radioactivity detected in the majority of the birds was naturally-occurring K^{40} .

A questionnaire to be mailed to a statistical sample of Tri-City license holders was drafted and approved by Public Relations personnel of Battelle-Northwest. It must still be submitted to the Washington State Department of Game for their approval. The questionnaire is designed to study the harvest and consumption of game birds from surrounding areas by local hunters.

DECLASSIFIED

[REDACTED]

[REDACTED]

DECLASSIFIED

ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Summary

Mission 1

Four Mark I outer components were examined in Radiometallurgy to identify unusual features observed in basin examination of discharged fuel. On two of the elements the anomalies were found to be associated with mechanical damage after discharge. On one element a blister of unknown origin was isolated for further study, and on the other element buckling from fuel swelling was observed on the inner surface.

Work was resumed on the study of the end-spider support concept for N-Reactor fuel, with the first objective to evaluate the existing end-spider design in a loop test of the Mark Ic assembly.

Experiments were performed to evaluate the effectiveness of a mixer spool of new design (H-3-14183). It was found that this mixer spool was less effective in reducing subchannel temperature disparities than the mixer currently in use (H-3-17493). The new design was found, however, to produce approximately 17% less pressure drop than the standard mixer.

Earlier, experiments were performed in the Thermal Hydraulics Low Pressure Facility to determine the thermal and hydraulic characteristics of a N-Reactor High Power Density (HPD) fuel column. These experiments were performed with an electrically heated test section which simulated the axial power distribution of a typical in-reactor fuel column.

In occluding silicon particles in electroplated nickel deposits, twice as much silicon can be occluded at a given pulsed-current density as can be occluded using conventional d.c. current plating.

A prototype weld cycle counter was completed and delivered to N-Fuels. This will enhance the control capability for the locking clip and support welder.

strips. There are 17 alloys included in this study. Work was also completed on the fabrication of duplex aluminum tubing (AlSi and 8001 Al) for use by Douglas United Nuclear in corrosion studies of hot die sized fuel.

Mission 8

Initial planning was carried out on an experimental approach to study the effectiveness of candidate charcoals for removing airborne iodine and organic iodides under conditions anticipated following serious reactor accidents.

Routine collection and analyses of meteorological data have continued and a problem in collection of data has been eliminated.

Laboratory studies showed that $\text{Al}_2(\text{SO}_4)_3$ was not as effective in reducing particulate migration through soils as Na_2SO_4 . The addition of 1000 ppm Na_2SO_4 gave the best results. Studies showed that the addition of Magnifloc 985 N effectively increased the permeability of solutions through soil columns; however, Magnifloc did not significantly reduce particulate migration.

Relatively high tritium concentrations in riverbank springs and well samples when correlated with waste discharge data confirmed previously calculated travel times for 1301-N crib waste to move from the crib to monitoring wells and springs.

Mission 10

Preliminary experimental results indicate that the streaming potential (caused by flow of a fluid past a surface) has a maximum value at high Reynold's numbers. Because the data are difficult to reproduce, this conclusion has not been substantiated.

Process Technology

A sample of TSX graphite, coated by the thermal decomposition of tetramethyl silane, appears to oxidize at a rate 1/3 to 1/4 that of an untreated sample. The protection afforded by

DECLASSIFIED

control tubes. The use of anodized spacers involves the last 11 to 12 months of a total 55-month exposure.

Completion of the prototype rem meter and battery charger prepared the instrument for neutron source calibration measurements and final adjustments.

DECLASSIFIED

[REDACTED]

DECLASSIFIED

[REDACTED]

-12-

BNWL-658

ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Mission 1 - Basic Production Mission

Examination of N-Reactor Elements
(J. W. Goffard, Metal Fuels Unit)

Four Mark I outer components were examined in Radiometallurgy because of unusual features noted during N-Reactor basin examination of discharged columns. Notes on each examination follow:

Element HAD-35-1, 23 in., 0.95 outer from tube 1850, appeared to have a blistered area near an end when viewed in the basin. In cell it was obvious that the blisters had resulted from water entry and fuel corrosion as a consequence of discharge or handling damage to the end closure. A large gouge (about 1/3 of closure cross-section) was noted and the adjacent clad had been pried away from the fuel so that water had access to the clad-fuel bond zone.

Element H-6769-12, 23 in., 0.95 outer from tube 2151, displayed localized inner bore buckling in one end. In-cell examination confirmed the existence of the buckling, apparently from fuel swelling, and revealed that it was most severe on the half of the inner bore opposite the locking hardware on the numbered end of the element. Incipient buckling was noted elsewhere on the element. There did not appear to be any reason for the localized nature of the most severe buckling such as surface crud or filming from hot-spots or narrowed annulus.

Element HAD-83-3A, 23 in., 0.95 outer from tube 1749, displayed a small blister on the inner bore very near to the end closure when examined in basin. In-cell examination revealed a small, intact blister on the inner surface opposite the locking hardware on the numbered end. The blister was quite close to the end cap but did not appear to be associated with it in any manner. The closure displayed no damage or defects in the zone of the blister. A sample was cut for further study.

[REDACTED]

[REDACTED]

[REDACTED]

N Fuel Measurements

(R. K. Marshall, Metal Fuels Unit)

The following fuel elements were measured and weighed:

	<u>Measured</u>	<u>Weighed</u>
26 in. Inner	24	24
26 " Outer	14	14
23 " Inner (Mark IV)	83	118
23 " Outer (Mark IV)	<u>24</u>	<u>63</u>
Total	145	219

Transient Heat Transfer Experiments for K-Reactor

(F. R. Zaloudek, Thermal Hydraulics Analysis Unit)

Methods are being developed to predict the rate of coolant ejection from a process channel following a loss-of-coolant flow accident and to define heat removal rates from the fuel during coolant ejection. Two specific incidents leading to the loss-of-coolant are being considered, upstream plugging and a front header break. Laboratory experiments performed before are being augmented by analytical work to enable the extrapolation of the experimental results to a wide variety of coolant conditions and accidents.

A document entitled "A Digital Program for the Prediction and Analysis of One-Dimensional, Two-Phase Flow Transients Following Sudden Loss of Coolant or Loss of Pressure" was prepared for publication. This document is an interim report describing a digital computer code developed to solve the hydrodynamic equations describing the two-phase flow transient following a sudden loss-of-coolant or loss of pressure in a coolant channel in a pressure-tube reactor. This code, written in Fortran IV, considers a separated, two-phase, slip flow in a constant cross-sectional flow channel with heat addition. A sample problem is presented which considers the events immediately following the rupture at the outlet of a straight, horizontal unheated duct. The results showed the characteristic depressurization and recompression cycles observed experimentally following pipe ruptures in pressure systems.

DECLASSIFIED

minimizing the cross-section temperature disparities at any axial position along the tube. Water mixer fuel pieces which mix the subchannel flow streams are used in the production to reduce these peripheral and hole-to-annulus coolant temperature disparities and thus reduce corrosion problems. Each mixer is approximately two inches long and is attached to a six inch fuel piece. The mixer is generally attached to the downstream end of the seventh fuel element from the end of the charge.

An experimental evaluation was performed to determine the effectiveness of two mixer spool designs. The first was a new design (H-3-24183) and the second, the standard design in current use (H-3-17493). Both of these designs exchange the hole flow stream for water from the annulus as it passes by the mixer. In performing the subchannel fluid interchange, the standard mixer utilizes round interchannels, whereas the new mixer utilizes larger slotted interchannels. The new mixer provides a much greater cross-sectional interchannel flow area than that provided by the standard mixer.

All experimentation was performed in the 189-D Hydraulics Laboratory. Each test assembly consisted of a mixer spool plus seven 03N fuel elements, four upstream and three downstream of the mixer in a standard B-Reactor ribbed process tube. The fuel charge was followed by five 8 in. perfs to complete the fuel train. In the mixing effectiveness study, the fuel charge was simulated using aluminum dummies which were welded to one another and to the mixer in the order mentioned above so that the hole and annulus flow channels were separated before and after the mixer. However, in the pressure drop study, typical single uranium fuel slugs were charged into the process tube in the above order with the two-inch mixer attached to the downstream end of the fourth fuel slug. Mixing effectiveness and pressure loss studies were performed separately for each mixer type. In order to properly compare the performance of the two mixer designs, the mixer spools were always charged into the ribbed process tube with the same orientation relative to the process tube.

DECLASSIFIED

fuel column from the measured pressure loss across a similar length including a mixer.

Pressure losses of the new mixer were found to be less than those of the standard mixer in current use. At a typical flow of 50 gpm, this difference amounted to 17.5% of the total pressure loss of the standard mixer. At higher flow rates, the difference increased. At typical flow rates, the pressure drop of the new mixer is approximately equal to that of a single uranium fuel element which amounts to 11.3 psi at 50 gpm.

In order to determine which mixer type is most applicable to reactor operations, an optimization which includes fabrication costs, pressure losses, and mixing effectiveness must be made. On the basis of pressure losses, the new mixer is best, whereas, on the basis of mixing effectiveness the standard mixer is better. As expected, neither mixer embodies the unattainable hydrodynamic combination of minimum pressure loss and maximum mixing effectiveness. Gains in mixing effectiveness are accompanied by increased pressure losses, whereas, decreases in pressure losses are accompanied by decreased mixing effectiveness. Therefore, the results of this study, the particular reactor operating improvement desired, and the relative fabrication costs of the mixers will determine the choice of the mixer type to be used.

Thermal-Hydraulic Behavior of K-Reactor High Power Density Fuel Column

(F. R. Zaloudek, Thermal Hydraulics Analysis Unit)

Earlier, experiments were performed in the Thermal Hydraulics Laboratory Low Pressure facility to determine the thermal and hydraulic characteristics of a K-Reactor High Power Density (HPD) fuel column.

In these experiments, an electrically heated model simulating a 38-element (HPD) fuel column was used. This model was 1.512 in. OD, 0.470 in. ID, and was fabricated from short lengths of several alloys having different electrical resistivities. This produced a stepped cosine axial power distribution approximating the power distribution for an in-reactor fuel column when

DECLASSIFIED

- With the standard rear hardware, flow instability would not result from upstream plugging in tubes operating at powers up to 1500 kW. With the open rear hardware, flow instability would not result from upstream plugging in tubes at powers up to about 2900 kW.
- For the standard rear hardware, protection against burnout from slow plugging should be provided by low Panellit pressure trips at tube powers up to about 1500 kW and by high Panellit pressure trips at tube powers above about 1200 kW. For the open rear hardware, Panellit trip protection against burnout from slow plugging is questionable. Such protection should be provided by low Panellit pressure trips at tube powers up to about 1500 kW and by high Panellit pressure trips at tube powers above about 2000 kW.
- Fast upstream plugging should produce low Panellit pressure trips for all cases in which instability could occur. Low trips should occur for sudden plugs sufficient to produce burnout for all powers investigated in these experiments and for both the standard and open hardware cases. High trips before burnout should be obtained also for tube powers above about 1200 kW with standard hardware and about 2000 kW with open hardware.
- For sudden upstream plugging, the response time of the safety circuits should be adequate to prevent fuel from reaching jacket melting temperatures for flow reductions up to 60% in a 3000 kW tube and more than 75% in a 2000 kW tube.
- The experimental boiling demand curves agree well with calculated curves based on saturation pressures in the annulus but are higher than calculated curves based on the bulk outlet saturation pressures. Trip limits determined from bulk outlet saturation curves would be erroneous in the non-conservative direction.

DECLASSIFIED

[REDACTED]

DECLASSIFIED

B [REDACTED]

1060 ma/cm². In both direct current plating and pulsed current plating, increasing the air flow from 35 to 150 ml/min produced a smoother deposit without reducing the silicon content; usually, increased agitation decreases the silicon content of the deposit.

Support Welder Cycle Counter

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

A prototype weld cycle counter was completed and delivered to N-Fuels. Fabrication of additional units is planned, so that continuous surveillance of each locking clip and support welder may be performed. The device automatically displays the number of half cycles of 60 Hz welding current per weld, which is indicative of weld quality, and must be precisely controlled.

UE-1 External Bond Chassis Modification

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

Design work was begun on a surface following RF gate, one of which will be installed in each UE-1 external bond chassis upon final approval. These additions will alleviate relative gate position drift caused by a combination of long water path with changing water temperatures.

Radiographic services to DUN this month include examination of six pipe welds in the upper crossheader at the X-1 level of 105 KE reactor. Several repairs due to faulty welding were necessary. Six welds in aluminum components were radiographed for DUN manufacturing section.

Statistical Assistance

(J. L. Jaech, Statistics Section)

A failure rate curve was estimated for hot die size fuel elements based on the limited experience available. Limits of uncertainty were also constructed. A computer program was developed recently which facilitates analyses of this type.

A large body of historical failure rate data from the old reactors is being used to construct a model relating failure

[REDACTED]

[REDACTED]

[REDACTED]

Compacts of fused MgO grain, sintered at various temperatures up to 1600°C (85-90% T.D.) reacted with water at 150°C at 170 psi. MgO was eliminated as a carrier when a fused single crystal specimen also failed during autoclave testing.

Preliminary autoclave tests indicate that PbO and ZnO may be suitable candidates. A literature study was started for alternative carrier materials.

Mission 5 - Other Isotopes

Preliminary Analysis of Am²⁴³ and Cm²⁴⁴ Data Obtained from Aluminum-Plutonium Fuel Irradiated in the PRTR (R. P. Matsen, Light Moderator Reactor Physics Unit)

Americium-243 and curium-244 data obtained from aluminum-plutonium fuel irradiated in the Plutonium Recycle Test Reactor (PRTR) were analyzed to obtain cross-section information about the Pu²⁴² → Am²⁴³ → Cm²⁴⁴ transmutation chain. Originally the fuel was 2.6 wt% plutonium in aluminum and contained 16.45 at.% Pu²⁴⁰. The fuel was irradiated in the PRTR during 1962 and 1963. Four samples of the fuel were destructively analyzed for americium and curium in 1967. These four samples plus an unexposed sample span a burn-up range of 0 to 50% in terms of the percent depletion of plutonium.

The Pu²³⁹, Pu²⁴², Am²⁴³, and Cm²⁴⁴ isotopic concentrations were determined as follows. The total plutonium concentration was determined by the isotopic dilution technique. The isotopic composition of a plutonium sample was determined by combining the results of a mass spectrometer analysis on an unspiked plutonium sample with the isotopic dilution results of the spiked sample. The amount of Pu fissions was determined by a Cs¹³⁷ analysis of the dissolved fuel sample. The americium and curium were chemically separated from a small portion of dissolved fuel sample. The Am and Cm came off together when they were separated. A total alpha count followed by a selective (by energy) alpha count on this separated sample yielded the isotopic concentrations of Am²⁴¹, Cm²⁴², and Cm²⁴⁴. The amount of Cm²⁴³ was assumed to be negligible. When the fuel is removed from the reactor, the Cm²⁴² alpha activity is large because of its short 163 day half-life (compared with 458 yr for Am²⁴¹ and 18 yr for Cm²⁴⁴). However, the four-year

DECLASSIFIED

DECLASSIFIED

-26-

BNW

The transmutation equations are fitted to the data with the least squares fitting program LIKELY. The iterative scheme used in LIKELY breaks down for these equations because of the complex way in which the cross section ratios appear in the transmutation equations. Thus, each equation was forced to pass through its first and last points by the use of hand calculations, to obtain good first estimates of the parameters and reduce the iteration problems. The cross section ratios obtained from the fit are listed in the next table. The calculated Pu^{242} curve fits the data quite well. For both the Am^{243} and Cm^{244} curves, the second data point is 15% low, the third data point is 30% high, and the fourth data point agrees with the curve. It is not too surprising to observe this correlation between the Am^{243} and Cm^{244} data since both of these numbers depend upon the total alpha count of the Am and Cm which are mixed together. If too high (low) an alpha count is measured, both of the Am^{243} and Cm^{244} will be high (low) by the same amount.

Ratios of Effective Cross Sections

$$\hat{\sigma}_c^{41}/\hat{\sigma}_a^{49} = 0.3155$$

$$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49} = 0.08779$$

$$\hat{\sigma}_a^{53}/\hat{\sigma}_a^{49} = 0.1665$$

$$\hat{\sigma}_a^{64}/\hat{\sigma}_a^{49} = 0.0 \text{ (assumed)}$$

The effect of applying the decay correction to the Cm^{244} data will be to increase the value of $\hat{\sigma}_a^{53}/\hat{\sigma}_a^{49}$ and will not significantly change the other ratios. In order to better establish the Am^{243} and Cm^{244} curves and also obtain more reliable values for the cross section ratios, several additional samples will be analyzed by the same method. The final analysis of all samples will include decay effects for Cm^{244} and a more accurate assessment of the uncertainties (including correlation effects) to be assigned to the experimental data.

reduction of the U^{236} reaction rate results from a depletion of the source of neutrons scattering into the energy range of the U^{236} resonance rather than from a direct competition of the two isotopes for the available neutrons.

Calculations were also made using the HAMMER code. The geometry and composition used for these calculations were the same as used for the Monte Carlo calculations; however, the cross sections were not obtained from the same library. The value obtained for the case where U^{238} is present is in good agreement with the value obtained using the RBU Monte Carlo code. This would be expected if the resonance shielding is caused by a depletion of the source. It is not understood why the results without U^{238} are in such large disagreement. Final results of this study are being obtained and will be reported in a topical report.

Calculated Values of the U^{236} Reaction
Rate per Source Neutron

$$\int_{V_{\text{fuel}}} dv \int_{5.04 \text{ eV}}^{6.47 \text{ eV}} \phi(E) \sigma_c(E) c(E) dE / \text{Source Neutron}$$

	Without U^{238} Column 2	Ratio Col 4/Col 2	With U^{238} Column 4
RBU Monte Carlo Code	81.40 ± 2.58	0.76	61.86 ± 1.52
HAMMER Code	71.69	0.91	65.18

DECLASSIFIED

DECLASSIFIED

-30-

BNV

Spectrographic Analysis of Aluminum Alloys

Alloying Elements, wt%

<u>Alloy No.</u>	<u>Ni</u>	<u>Fe</u>	<u>Si</u>	<u>Mg</u>
1	0.69	2.2	0.06	
2	1.4	1.75	0.07	
3	0.66	1.8	0.06	
4	1.0	0.78	0.08	
5	2.8	0.35	0.06	
6	1.85	1.5	0.06	
7	0.87	0.44	9.8	0.71
8	0.96	0.32	10.8	0.73
8a	0.72	0.22	8.2	0.57
9	1.3	0.56	0.15	
10		0.04	0.03	0.002
11	0.65	2.2	0.03	
12	1.2	2.0	0.02	
13	0.6	2.1	0.02	
14	1.0	0.69	0.02	
15	2.0	0.35	0.02	
16	1.4	1.7	0.02	

CMLE Overbore Fuel Element - PCTR Experiment

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

The amounts of copper necessary to achieve unit neutron multiplication in an experimental lattice of CMLE overbore fuel type inserted in a graphite lattice with 7.5 in. lattice pitch have been measured in the Physical Constants Test Reactor and are listed in the following table for various lattice conditions.

The gas mass flowmeter and thermal conductivity meter have been calibrated to determine the fraction of hydrogen in the gas stream using fission product release test apparatus. The initial calibration tests indicate a probable error of 5%. The equipment will be used to determine the metal-water reaction rate for both unirradiated and irradiated fuels heated in a steam atmosphere. In this way it will be possible to estimate the volume of hydrogen produced during a loss-of-coolant accident.

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

This program is intended to establish the possible local earthquake motions and forces and the capability of Hanford structures to resist these motions. Also sought is an indication of the coupling, during seismic activity, between the Ringold Basalt underlying the Hanford project and the reactor foundations constructed on overburden glacio-fluviatile deposits.

Natural seismic accelerations are being recorded at Gable Mountain and at reactor foundations (100-K). The Geo-Recon seismic detection system is being used. Both the "A" unit, installed on Gable Mountain and the "B" unit at 100-K are actuated by seismic accelerations of 5×10^{-4} g in the basalt.

Acceleration Response Spectra will be constructed from the recorded weak motion seismic data. These spectra will indicate coupling between the two sites and also the characteristic earthquake frequencies for Hanford. In addition, the vibration characteristics of reactor structures (natural period, damping and stiffness) will be obtained by experiment and/or analysis.

The "A" unit Geo-Recon weak motion accelerograph was installed on Gable Mountain January 4. The unit is operating complete with a 10X amplifier on all three channels, a Honeywell Brown Recorder, to show the time of any recorded event, and the temperature compensating circuit for the vertical channel. Operation has been satisfactory to date except for some slight gain in the vertical channel due to normal changes in room temperature. To reduce this effect minor changes are planned for the temperature

DECLASSIFIED

DECLASSIFIED

-34-

B-606

decoded and validity tests for the data were discussed. The groundwork was then laid for writing the program to summarize the telemetering system data.

Part of the program to determine trajectories of airflow over the Wahluke Slope includes the observation of balloons traveling at approximately a constant level. To eliminate the influence of winds near the surface, a mechanism was built to raise the balloon to the 300 ft level of the 100N tower before its release. This will eliminate problems associated with the effect of wind shear with height on the trajectory.

Particulate and Radionuclide Migration in N-Area Soils
(B. L. Carlile, Soil Science Unit)

Treatment of soil columns with various concentrations of Na_2SO_4 and $\text{Al}_2(\text{SO}_4)_3$ alone and in combination with a polyelectrolyte (Magnifloc 985N) were continued in an attempt to develop a procedure for reducing particulate migration in the N-Area emergency disposal facility.

Sodium sulfate continued to be most effective in reducing particle migration in soils which effectively reduce the breakthrough of Sr^{85} . Concentrations of 1000, 100, and 10 ppm of Na_2SO_4 in river water were studied and the higher concentration is much more effective. $\text{Al}_2(\text{SO}_4)_3$ at 20 and 100 ppm in river water was not so effective as Na_2SO_4 in soil flocculation but by the addition of 0.1 ppm Magnifloc with the $\text{Al}_2(\text{SO}_4)_3$, better results were obtained.

In leaching soil columns with weak electrolytes (e.g., tapwater), the soil will eventually disperse and the flow rate is severely reduced or stopped. By the addition of Magnifloc to these columns, some flow through the column can be regained although not at the original rate. Therefore, this treatment would be of some benefit in recovering the permeability in disposal facilities which become impermeable from waste disposal operations.

Corrosion and Water Quality Studies

(W. B. Silker, Radiological Chemistry Unit)

The use of filtered raw water, pH 6.6 and containing 1 ppm sodium dichromate, instead of process water, resulted in increased effluent concentrations of As^{76} and P^{32} . Concentrations of other effluent radionuclides were not drastically altered.

Cessation of sodium silicate addition and commencement of sodium dichromate addition to the deionized water provided significant decrease in the concentration of all effluent radioisotopes.

Process Technology

Coating Formation on Graphite

(R. C. Giberson, Materials Compatibility Unit)

Work continued on a method for depositing a silicon coating on graphite with the hope of reducing its oxidation rate. Deposition conditions which could be applied in a reactor system are chosen. The small sample of TSX graphite was oxidized to a total of 23% burnoff. This sample had been coated with 33.4 mg (4.2%) of the deposition product. Deposition conditions were ~1300 ppm of tetramethyl silane (TMS) in helium at 750°C . An infrared study of the thermal decomposition of TMS revealed only CH_4 , C_2H_4 and C_2H_2 . No ethane or higher hydrocarbons were formed. Hydrogen was also found by gas chromatography.

After ~350 hr of oxidation by water vapor (5400 ppm) at 900°C , the rate of reaction of the coated sample has remained constant in the range from 0.5×10^{-3} to 0.7×10^{-3} g/g-hr. It has now been oxidized for ~450 hr. An uncoated sample exposed to the same oxidation conditions showed a rate increase from 0.1×10^{-3} to 1.8×10^{-3} after 180 hr. The oxidation rate since that time has remained fairly constant in the range of 1.5×10^{-3} to 2.0×10^{-3} g/g-hr. The untreated sample has now been oxidized for a total of 250 hr. The long oxidation test was made to determine whether the coating was providing permanent protection or a temporary one. The data indicate a long-lived protection factor

DECLASSIFIED

DECLASSIFIED

-38-

possibly nitric oxide, were found, no ammonia was detected, and methane, if present, was obscured by nitrogen.

It appears that the predominant reaction of 350°C is simply the reaction of water vapor with graphite

The changes in yields of 650°C may indicate the increasing importance of secondary reactions involving products of the above reaction.

The nitrogen used in these runs contained small amounts of impurities which could not be removed in liquid-nitrogen cold traps. Further work on this system is awaiting the arrival of reagent-grade nitrogen.

Ammonia, in large quantities, comes through the gas chromatographic column with severe tailing. At concentrations of 100 ppm it is held back and not observed. "Porapak S" column material has been obtained to increase the accuracy of the ammonia analysis.

A gas recirculator was designed for use in the gamma facility and is being built. This will allow a graphite sample to be irradiated in a flowing gas system at elevated temperatures. Water and ammonia, but not CO₂, can be removed by adsorption on molecular sieve, and controlled amounts of water vapor can be added through a diffusion cell.

Corrosion of K Area Diesel Engine Powered Emergency Pumps (A. P. Larrick, Corrosion Research Unit)

A recent inspection of the K Area diesel engine-powered emergency backup coolant pumps showed extensive corrosion to the cast iron pump bowls and vanes. Up to 1/4 in. residual graphite was found remaining on the surface at some areas due to selective dissolution of the iron. Laboratory tests indicate that the probable cause of the corrosion is galvanic currents flowing between the brass impeller and the cast iron bowl. Increasing the pH from 6.6 to 11.0 and adding catalyzed sodium sulfite to either

not conducive to attack and four of the eight discharged tubes were equipped with this type of spacer during the last 11 to 12 months of their exposure.

To date, only the base metal hydrogen concentration of four tubes has been determined and this has been done at a position about 6 in. from the downstream end. Two of the four tubes were charged with anodized spacers during the latter portion of their exposure. Base metal concentrations of these tubes were 105 and 111 ppm. Base metal content of the remaining two tubes, charged with standard aluminum spacers throughout their exposure, was 89 and 128 ppm.

The base metal hydrogen concentration about 6 in. from the downstream end that was associated with the first set of eight tubes ranged from 74 to 115 ppm (average value, 97 ppm). All tubes of the first set, installed in-reactor for 42 to 44 months, were charged with standard spacers throughout their exposure.

Water Filtration Studies

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

Four polyelectrolytes were compared with Magnifloc 990N for use as filter conditioners. These were the Betz Poly-Floc Nos. 1100, 1110, 1120, and 1130.

From the standpoint of effluent turbidity readings during filtration, none of the four materials proved to be as effective as Magnifloc 990N. They were also found difficult to feed through flow-controlling rotameters because of the highly viscous nature of solutions at concentrations as low as 30 mg/l. Further testing of these materials is not recommended.

Additional equipment was installed on the laboratory water filtration pilot plant that will allow the addition of measured quantities of river sediment to the raw water. This will allow studies to be made on a wide range of fixed raw water turbidity levels to properly assess the effect of this variable on water treatment plant operation.

DECLASSIFIED

DECLASSIFIED

BNWL-696

N-Reactor Simulation

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

A series of analog simulation runs was made with the overall N-Reactor model involving setbacks in power initiated by excessive pressure in the main header during turbine trip transients. Several trip points and various values for the power setback ramp were used in an attempt to minimize the power rate of rise during simulated turbine trip transients, thus preventing a reactor scram. A second series of runs was made with turbine trips and reactor scrams at power levels of 20% above normal. A final set of runs was made with a simulated reactor operator in the loop during turbine trip transients. The simulated operators function was to initiate a power setback whenever the power rate of rise approached the scram level. Results of these runs are being studied and will be compared with actual plant observations in an attempt to obtain a suitable plant operator model.

Radiation Detection and Measurement Instrumentation

(E. M. Sheen, Instrumentation Section)

Completion of the prototype rem ratemeter and battery charger fabrication has prepared the instrument for checks with neutron sources. After initial discriminator adjustment and possible shifts in count rate and time constant ranges, the prototype instrument will be delivered to the External Dose Evaluation Unit for accuracy checks over a range of neutron energies.

DECLASSIFIED

-43-

6

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Mission 1

Complete leaching of plutonium from burned ZPPR fuels has not been obtained.

Batch contact solvent extraction of neptunium from Purex Acidified Sludge (PAS) with DBBP-NPH as the solvent appears to be feasible. Recoveries of greater than 90% were obtained in laboratory-scale experiments.

Erratic results in the preparation of Np(V) oxidation rate accelerating material were eliminated by increasing the nitrite to nitropropane ratio to 0.6.

Initial results indicate that a platinum catalyst will effect recombination of H₂ and O₂ formed by radiolytic generation in plutonium nitrate solutions.

The corrosion resistance of 304L stainless steel was determined in simulated sulfate-free Purex Waste solutions. The corrosivity of waste solutions free of Cr(VI) was essentially independent of the presence or absence of sulfate, whereas the corrosivity of waste solutions containing Cr(VI) was inhibited by the presence of sulfate.

Fabrication of the solid state, portable, dual-channel spectrometer for gamma measurements progressed satisfactorily with completion of the preamplifier, the single channel circuits, and the alarm unit; design of the remaining circuits moved forward.

Mission 4

Experiments intended to show the amount and form of iodine in irradiated pressed graphite and irradiated preformed-impregnated graphite neptunium target elements indicated that 15-20%

infinite cylinders, and finite spheres, and cubes. Dimensions are given for homogeneous PuO_2 -polystyrene mixtures, PuO_2 -water mixtures, and Pu-water mixtures. The results are in qualitative agreement with DTF calculations. The effect of the Pu^{240} (11.5 wt% Pu^{240}) is to increase the critical mass and volume by nearly 30% at the H/Pu ratio of 5.

Difficulty is being encountered in analyzing experiments with plutonium solutions in slab geometry. Agreement between measurement and calculation of critical thicknesses of infinite slabs of dilute solution is not satisfactory--although the same calculational technique (HFN code) reproduced the experimental data on plutonium solutions in spherical geometry reasonably well. Reasons for the discrepancy are being explored in detail.

Mission 9

A subroutine was written to use in calculating the non-linear equilibrium curve in correlating data for steady state modeling of a solvent extraction column.

Design data information was transmitted to ARHCO for a thermal conductivity probe for installation in underground waste tank systems.

Preliminary results from measurement of the thermal conductivity of block sodium nitrate indicates a value of 0.5 to 0.6 Btu/(hr)(ft²)(°F/ft).

Continuing design study based on heat transfer considerations only with candidate waste forms cesium chloride and strontium fluoride was completed.

New irradiation studies of purified D2EHPA at a level corresponding to about 10% destruction gave 5.7% monoester and 3.5% nonacidic material. Tests with plant grade solvent were in agreement. This represents less decomposition than previously reported. Nuclear Magnetic Resonance spectrometry was found to be too insensitive to distinguish components of the nonacidic fraction.

DECLASSIFIED

DECLASSIFIED

Process Technology

A pilot plant pulse column demonstration of americium recovery from neutralized CAW was completed. Low americium waste losses (ca. 0.5%) were obtained using feeds in the pH range 0.3-0.7.

Miniature mixer settler tests were made of a proposed flowsheet involving $\text{HNO}_3\text{-H}_2\text{SO}_4$ for stripping plutonium from solvent in the Purex 2B column. Satisfactory plutonium stripping could be obtained but the effect of the conditions used on uranium decontamination are yet to be determined.

Laboratory studies in support of B-plant startup on rare earth and strontium concentrates was completed. Flowsheet modifications were required to avoid lead sulfate precipitation.

The Am-Cm purification campaign was completed successfully. Slightly over 50 g Cm were obtained. About 760 g Am were recovered pure and another 200 g in end cuts.

Development of a continuous neutralization system for the Plutonium Reclamation Facility CAW stream was completed.

Four types of polyethylene plutonium storage bottles have been tested for resistance to stress cracking. These include the 10-liter bottles used at Hanford, the 3-liter and 10-liter bottles used by Nuclear Fuels Services, and bottles supplied by Monsanto. Density measurements show only slight differences in the polyethylene used in the various bottles.

Modifications to the prototype Purex HAW stream sampler are essentially complete. Operation of the device appears satisfactory.

Structural temperatures for two steady state and one transient case were estimated.

Assistance was provided in installing a thermocouple probe in the 101-SX tank sludge layer. The probe was impact-driven four feet into the 12 ft. thick hard sludge layer.

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Mission 1 - Basic Production Mission

Reprocessing ZPPR Fuel Scrap

(G. F. Schiefelbein, Separations Chemistry Unit)

Three 24-hour dissolution runs were made on burned ZPPR fuel (68% UO_2 , 28.6% PuO_2 , and 3.4% MoO_3). The initial nitric acid concentration was increased from 8.4 to 10.8M during subsequent trials while holding the fluoride and ferric ion concentration constant at 0.24 and 0.12M, respectively. After 24 hr reflux, the solutions still contained appreciable solids shown to be plutonium compounds by X-ray analysis. Apparently the burning process converts a part of the plutonium to a refractory compound. Increasing the nitric acid concentration has resulted in slightly increased plutonium dissolution but not stoichiometric increases.

DBBP Extraction of Neptunium from PAS Solution

(W. W. Schulz, Separations Chemistry Unit)

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

Experiments with synthetic PAS solutions have demonstrated the feasibility of the DBBP process. These experiments showed that the ease of adjustment of the neptunium valence state and subsequent DBBP extraction depends on the PAS acidity; satisfactory results are obtained with 0.5M acid PAS solutions but not with solutions which were 0.25M (or lower) in acidity. In

DECLASSIFIED

DECLASSIFIED

Another experiment was done to see what effect a small amount of butyl alcohol (decomposition product of TBP) would have on the rate of gas generation. In this experiment, the butyl alcohol concentration was 0.5 vol% in a 4.4M HNO₃-plutonium(IV) solution. A rapid initial reaction, lasting about a day, evolved NO₂ indicating that nitric acid was being reduced. The most likely net reaction is

After this, the rate of gas evolution corresponded to a G_T of 0.060 which is slightly lower than the 0.080 found for 4.5M nitric acid.

Some consideration was given to the effect that might be expected for aliphatic amines on the rate of radiolytic gas evolution. These amines are expected to behave like fragments from an anion exchange resin. Since nitrous acid is radiolytically formed in nitric acid, the following reaction is expected for any primary amines present

Secondary and tertiary amines should not evolve gaseous products.

An experiment is in progress to determine if the radiolytically generated O₂ and H₂ can be recombined using a platinum catalyst. The catalyst used in this experiment is that found in the Deoxo catalytic purifier. Over a period of a week, the pressure over a 4.5M nitric acid-plutonium(IV) solution remained constant after a small initial pressure increase (~20 mm Hg). After this, the catalyst was closed off from the system and the pressure built up at a rate corresponding to the expected G_T of 0.080 for 4.5M nitric acid. After the pressure has been built up to a higher value, the system will be exposed to the catalyst again to see if the pressure can be reduced to the initial value.

of the installed solid state electronics, thus insuring continued reliable operation.

Gamma spectral analysis of eight process samples from the Waste Reclamation Facility provided high resolution data directly applicable to the design of CCW and CAW in-line test cells and associated electronic components. Both Si(Li) and Ge(Li) high resolution, solid state detectors were used in the measurements from 6 to 600 keV. After detailed analysis and correlation, the reduced results provided the needed spectral information. When completed, the in-line measurement instruments will provide process control for plutonium concentrations over a 0.001 to 0.5 g/l range.

Fabrication of the advanced, compact, dual-channel spectrometer progressed well with completion of two single analyzers, an alarm circuit and the preamplifier. Delivery of all ordered electronic components allowed initiation of fabrication of the pulse amplifier and power supply. Anticipated remaining design work should be accomplished without impeding planned timing of fabrication. Compared to the previously developed single channel spectrometers, the dual-channel design provides a significantly improved capability for discrimination against background from Compton scattering. Background compensation for possible interference from Zr-Nb, Am²⁴¹, and ruthenium will enhance the present plutonium measuring capability, now typically 0.5 to 50 grams, in hoods and glove-box installations. Additionally, the improved discrimination provides a versatile instrument for investigation of new product areas in connection with possible Pu²³⁸, Am²⁴⁴ and Np²³⁷ processing.

Mission 4 - Plutonium-238

Chemical Processing of Graphite Matrix Targets

(R. E. Lerch, T. S. Soine and F. P. Roberts, Chemical Separations Unit)

Several hot cell experiments have been carried out to determine the relative amount and form of iodine in the irradiated graphite-neptunium oxide fuels. Three types of experiments were performed: 1) combustion, 2) acid leach, and 3) acid-Hg⁺⁺ leach. In each experiment the off-gas was passed through a train

DECLASSIFIED

DECLASSIFIED

-55-

BB

The much higher total iodine content in the pre-formed-impregnated graphite fuels (too high to be explained by the difference in the age of the samples) may simply be due to more effective trapping of the iodine within the solid graphite matrix; i.e., a portion of the iodine in the pressed targets may have been released when they were opened.

Hot cell experiments are also being performed on samples of irradiated aluminum-neptunium target materials. Both mercury-catalyzed HNO_3 dissolution and caustic dissolution (or disintegration) are being investigated from the standpoint of iodine behavior:

Mission 5 - Other Isotopes

Isotopic Separation - Pulsed Current Electromigration (R. E. Lerch, Separations Chemistry Unit)

Analytical results of the final pulsed current migration experiments substantiate previous results. In addition, the best lithium isotope enrichment found to date was achieved, namely, 14.1% (from 12.70 to 10.91 for the Li^7/Li^6 ratio), by migrating lithium for 12 hr using 60 ma direct current.

Extraction of Polonium from Bismuth with Fused NaOH (G. F. Schiefelbein, Separations Chemistry Unit)

Work continued on characterizing a separation scheme employing fused caustic for the separation of Po^{210} from irradiated bismuth.

The cause for the poor polonium transfer levels experienced previously was determined to be atmospheric contamination of the inert argon blanket. Polonium transfer levels below 30% are experienced if air is allowed to diffuse into the argon blanket during molten contact. It is theorized that the poor transfer levels in the presence of air are due to the formation of bismuth oxide at the melt interface which acts to prevent the migration of polonium to the caustic phase.

Mission 8 - Nuclear Safety

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

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

Further criticality experiments were performed with plutonium solution of 23.3 wt% Pu²⁴⁰ for determining critical thicknesses of infinite slabs. The solution contained 282 g Pu/l at a nitric acid concentration of 2.1M. The criticality vessel, a variable thickness slab tank with 42 in. dimensions, was water reflected. Criticality measurements over a 5°C temperature range about room temperature confirmed the temperature coefficient of reactivity to be negative for the solution.

Analysis of data from criticality experiments in the water-reflected slab shaped vessel has yielded a critical buckling of $12,700 \times 10^{-6} \text{ cm}^{-2}$ for the 282 g Pu/l solution. The clean water-reflected infinite slab thickness is estimated to be 13.3 cm. The computed value in the absence of Pu²⁴⁰ is 6.7 cm. Thus, the results of preliminary analysis indicate a very pronounced effect on criticality from the 23.3 wt% Pu²⁴⁰. For the water-reflected slab at a concentration of 282 g Pu/l, the critical thickness has been doubled as a consequence of the Pu²⁴⁰ content.

Criticality Studies with Plutonium Compounds in the Undermoderated Range; Effect of Pu²⁴⁰ on Criticality*

(S. R. Bierman, Critical Mass Physics Section)

Analysis and interpretation of criticality data on plutonium systems in the undermoderated range continued. Data from critical experiments previously performed with PuO₂-polystyrene compacts at an H/Pu of 5 (2.3 g Pu/cc) containing 11.5 wt% Pu²⁴⁰ have been used together with buckling conversions to derive critical dimensions for several basic geometries; results are given in the table.

* Funded in part by Division of Reactor Development and Technology under Applied Reactor Physics.

of data is also being examined.

Atmospheric Release of Radioactive Particles

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

Several cold trial runs were made with the apparatus to be used to measure the particle evolution from boiling plutonium nitrate solutions. The objective of these experiments will be to establish under a fixed set of conditions the rate of plutonium released as fine droplets and mist from solutions of plutonium nitrate. All particles normally airborne from the open vessel must be collected, desirably without materially changing the air flow conditions which exist in the absence of the collector. An effort was made to measure the velocity of the vertical convective currents over the boiling solution. The vertical currents proved to be less than the sensitivity of the precision vane anemometer, some 50 fpm. Tests in which air was swept through the annular filter surrounding the rim of the vessel and through the top center of the bell-shaped hood placed over the vessel, showed that a 20 fpm air flow would carry all evolved steam through the apparatus. A linear upward flow of about 2 fpm carried a large fraction of the steam across to the condenser, suggesting that normal convective currents might be in this few feet per minute range. Flow in actual experiments will be of this order. Plutonium nitrate will be used following additional shake-down runs of the apparatus.

Moisture Loading of Filters

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

Technical advice was provided on a special air sampling problem at the B Plant involving the moisture loading of high efficiency filters. Temperature measurements suggested by study of the problem showed that the likely source of water was condensate, rather than water carry-over originally suspected as the source of the problem.

DECLASSIFIED

DECLASSIFIED

-61-

BNWL

probes. Heat rates of about 15 watts/ft of heater minimum or 90 watts/ft maximum may be required depending upon the actual value of conductivity and the temperature difference required to get the desired accuracy in measurement.

Organic Waste Disposal

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

The major emphasis in the ARHCO low level waste program has been placed on the disposal of organic waste solvents by burning. A design study is under way on an organic burner off-gas cleaning train using components on site. If successful, this effort can save as much as six months lead time in design and procurement of engineering scale incinerator equipment.

Laboratory scale equipment is being set up to demonstrate the distillation of CCl_4 from TBP and DBBP with sufficient purity to allow recycle within the Z-Plant complex. The CCl_4 must be separated from these solvents prior to burning or else the off-gas system must be able to handle mixtures of chlorine, phosgene, and phosphoric and hydrochloric acids.

D2EHPA Decomposition

(M. R. Young, Organic and Natural Products Chemistry Unit)

Since plant grade D2EHPA contains iron, which interferes with NMR measurements, iron free D2EHPA was irradiated to 10^8 rad and titrated differentially in 1:1 methanol-water with 0.5M NaOH. The results showed lower decomposition than previous data. The previously irradiated plant grade sample was again titrated and the results agreed with those for the iron-free samples. To eliminate any time effects, another plant grade sample was irradiated and titrated. These results also agreed with the recent titrations and are shown in the following table.

time or providing environmental temperature control it is expected that the short term demonstration can be executed without delay.

Radiolytic and/or Thermal Destruction of Organic Materials in B-Plant Extraction Column Raffinate

(W. W. Schulz, Separations Chemistry Unit)

To prepare satisfactory B-Plant extraction column feed-stock from PAW solution, various organic reagents are added to provide proper pH control and/or to suppress extraction of various contaminants. These organic complexants and buffering agents remain in the aqueous raffinate from the extraction column. In normal B-Plant operation, the extraction column raffinate will be neutralized to pH 10 by addition of NaOH and stored at or near boiling temperature in underground tanks pending eventual ion exchange treatment to remove Cs^{137} . In some cases, rework of the raffinate through the extraction column will be required to achieve acceptable recovery of strontium and rare earths.

Experiments are in progress to determine the rates and consequences of radiolytic and/or thermal destruction of organic materials in both neutralized and unneutralized synthetic raffinates. As part of this program, portions of neutralized raffinates containing hydroxyacetate and either NTA, HEDTA, or citrate were digested at 85 to 90°C for 12 weeks. Another portion of the raffinate containing NTA was irradiated at 88°C over a 4-week period to a total exposure of 10^9 rad. The pH of each raffinate decreased only slightly during radiolysis and/or thermal digestion. More significantly, neither prolonged irradiation nor thermal treatment affected zeolite ion exchange take-up of Cs^{137} from the raffinate solutions. Continued irradiation of the NTA-raffinate is scheduled to evaluate radiolysis effects further.

Settling Rates of Water-Leached Purex Sludge

(W. W. Schulz, Separations Chemistry Unit)

As part of the over-all Hanford Waste Management Program, high pressure water jets will be used to remove sludge stored in

DECLASSIFIED

DECLASSIFIED

-65-

BN

product density.

2. The calcination approach for SrO and Cs_2O must be investigated.
3. The compatibility of the selected compounds and their decay products with the encapsulating materials must be evaluated.
4. The effects of radiolysis will have to be determined.
5. Based on temperature calculations, the optimum container size must be determined. Experimental tests with hot containers will be needed to verify the temperature calculations.
6. Container loading techniques must be evaluated. This is especially important if container diameters are less than one inch ID.

Burning of Mercury-Oxygen Mixtures in Closed Containers
(W. E. Willingham, Process Engineering Unit)

Work continued with a 36 in. bed of molecular sieves and so far produced no detonations within the system and there was no indication of a burning wave. Two experiments were repeated but a portion of the gas mixture was added above the bed. In this case there was a burning wave above the bed but no detonation.

Movement of Radionuclides through Hanford Soils
(D. B. Cearlock, R. W. Nelson and A. E. Reisenauer, Hydrogeology Unit)

Investigations on using finite difference techniques to deduce permeability distribution showed that crude approximations of the hydraulic potential gradient result and that the calculated permeabilities are too inaccurate to be useful in reliably modeling the project flow system. However, a somewhat different approach that looks promising circumvents the difficult and time-consuming step of surface fitting hydraulic potentials. This method involves graphical construction of

of eight curves showing the instantaneous sodium-to-cesium ratio in the effluent solution and the sodium-to-cesium ratio of the sodium and cesium remaining on the column vs. the column volumes of scrub throughput. These data enable the column operator to determine from the sodium-to-cesium ratio in the effluent exactly when to stop scrubbing operations in order to attain a given sodium-to-cesium ratio in the eluate.

Another aspect of column performance requiring additional study in this system is the probable precipitation of aluminate upon reloading of an ammonia-based column. A short study of aluminate behavior in the system would be of value and was planned for proposal.

Process Technology

Recovery of Am from Pu Reclamation Facility Wastes (G. L. Richardson, Process Engineering Unit)

The pilot plant pulse column demonstration of americium extraction from neutralized CAW was completed. Final results suggested strongly that high americium losses reported earlier in the WE-1 column were caused by operation at unfavorably high pH. Lowering the feed pH from 0.8-1 to 0.3-0.7 resulted in americium losses in the range of 0.4 to 0.5%, corresponding to HTU_{OR} values of 2.5 to 2.8 feet. Operation at still lower feed pH should be possible, since excess nitric acid can be efficiently extracted at aqueous/organic volume flow ratios in the vicinity of 1.5 or less. Failure to extract excess acid can lead to high americium losses because of the reduction in the E_a^0 . The loss in extraction efficiency at high pH is believed to be caused by formation of stable ferric hydroxide films that inhibit mass transfer.

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

The Hanford Purex Plant uses two TBP solvent extraction cycles to recover plutonium from nitric acid solutions of irradiated uranium and to free it from the bulk of the uranium and

DECLASSIFIED

DECLASSIFIED

-69-

BNV

In addition, cold feeds simulating the contents of each tank were tested for cerium and promethium extraction as a function of contact time, pH, amount and type of complexant, and temperature. The pH 1.5 shown on the plant flowsheet was found to give gross precipitation (Pb sulfate). The flowsheet was altered and actual tank samples were tested in the Analytical Laboratories hot-cells for comparison between synthetic feeds and actual behavior. The results compared favorably with the synthetic work and indicated that efficient recovery should be obtained in the plant. All requested work to evaluate processing of the rare earth concentrates (tanks 6-1, 6-2, and 7-1) has now been completed in the laboratory, analytical laboratory, and hot cell. Additional work is continuing to make similar studies on the strontium concentrates (tanks 8-2, 12-1, and 9-1). Also, back-up work was requested by the customer to define conditions for processing the lead sulfate sludges contained in each of these tanks.

Am-Cm Ion-Exchange Purification Runs

(E. J. Wheelwright and F. P. Roberts, Separations Chemistry Unit)

The "AMCM" ion-exchange purification campaign was completed successfully and on schedule early in the month. Feed for the January run was the combined Am-Cm product produced during the two DTPA runs performed in December. Nitrilotriacetic acid (NTA) was used in this final run to separate Am and Cm from each other.

Except for extremely severe gassing, which required frequent degassing of the columns and operation at as high a flow rate as practicable, performance of the NTA flowsheet was much as anticipated from the prior tracer-level laboratory work. Excellent separation and recovery were obtained. The curium product totaled slightly over 50 grams; 760 grams of americium, Am^{241} plus Am^{243} , were obtained pure. Another ca 200 grams of americium is available in various end cuts. All of the product fractions are in the form of aqueous solutions. The americium product solutions will be transferred to ARHCO for calcination and fabrication into reactor targets. Following clean-

Fe/liter of solids. The solids look brown-red and appear to be reformed resin. They dissolve in dilute nitric without evolving CO_2 .

The concentration step for the ion exchange eluate was also piloted with an off-gas train in place. The cold finger was operated with 60°C water and plugging appeared as the initial off-gas began coming off. The water was heated to 75°C without reducing the solids formation. All surfaces that were dry collected solids. All wetted surfaces were free of crystal growth. Identification of the solids will be attempted.

Continuous Neutralization of Plutonium Reclamation Facility CAW Stream

(J. Dunn, Process Engineering Unit)

The pH control system for the continuous neutralization of CAW in the Plutonium Reclamation Facility was satisfactorily tested. The control points of pH 0.3, 0.75, and 2.5 were attained precisely (within 0.1 pH unit) repeatedly without overshooting and without hesitation. A new fritted junction reference electrode has overcome the erratic results of the standard type palladium junction cell. In addition, the results of other investigation has lowered the pH required to 0.3 and thus precipitation of the ferric hydroxide (and possibly Al, Ca, and Mg hydroxides) has been avoided. The precipitation of these metals interferes with pumping, extraction and control.

Stress Cracking in Polyethylene Storage Bottles

(J. H. Kleinpeter, Materials Compatibility Research Unit)

Tests were continued in an attempt to understand more fully the factors affecting stress cracking in the polyethylene bottles used for storage and shipment of plutonium nitrate. Previously we have tested the 10-liter bottles used by ARHCO and the 3-liter and 10-liter bottles used by Nuclear Fuels Services.

During this reporting period, we have tested four bottles supplied by Monsanto Chemical Company. Two of these, tested at

DECLASSIFIED

DECLASSIFIED

BNV

-73-

completed and the system shown to be reasonably precise (within 5% reproducibility on volumetric delivery) and well within normal analytical limits. A second eductor was mounted separately to empty the treated liquid phases after sampling (extracted residue) and connected to a cell drain rather than back to the process.

Vessel Vent Chemistry

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

Arrangements were made for sampling several locations in the Purex vessel vent system for analysis early in March. Samples will be analyzed by mass spectrometer for nitrogen, oxygen and hydrogen; by spectrophotometry for NO_2 ; by oxidation followed by spectrophotometry for lower oxides of nitrogen; by gas chromatography for organic components including organic iodides; and by an as yet undefined absorption method for free iodine. Testing of sampling and analytical procedures will be done before actual samples are taken.

Circulation Patterns in Air Lift Agitated Storage Tanks

(W. E. Sillingham, Process Engineering Unit)

Measurements were completed on the baffled tank, quarter full and full, and circulation patterns were drawn. Velocity profiles were made and are being compared with the unbaffled tanks. Preliminary viewing indicates a shift in the velocity profiles from the center of the tank to the outside with the addition of the surface baffles.

Underground Tank Structural Temperatures

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

Structural temperatures were calculated for one new AY tank farm design and one set of operating conditions for Tank 108SX. Calculations of structural temperatures for Tank 108SX containing sludge and filled with sand were begun.

Transient temperatures in walls during tank startup were estimated for four typical startup conditions.

[REDACTED]

Soil Chemistry

(B. J. Hajek and J. W. Shade, Soil Science Unit and Water and Waste Management Unit)

Studies were initiated to determine the adsorption behavior of trace strontium in a partially-saturated transient flow system. The analogous field case is adsorption from wastewater invading dry soil. The results showed that adsorption of trace strontium from a 0.5N sodium solution is about one-fifth in this system of that predicted by use of adsorption constants determined by soil plug techniques or 50% activity breakthrough volumes. This initial low adsorption which increases with volume throughput seems to be the principal reason for lack of agreement between experimental and predicted radionuclide breakthrough concentrations in soil column effluents.

An investigation was started to determine the effect of dominant mineral constituents of the adsorption of trace radiostrontium and radiocesium from simulated low-level process wastes by a local Hanford soil. The soil was fractionated on the basis of size, density, and magnetic susceptibility. Specific minerals, characterized by a limited range in magnitude of these properties, could be concentrated into specific fractions.

Sand size particles composed about 90% of the soil, while silt and clay, which were not further separated, made up the remainder. Since past work has shown that the sand fraction adsorbs most of the radionuclides, it was further fractionated according to density and magnetic susceptibility. Optical examination of the light fraction (>2.9 sp g) consists largely of amphiboles, ore minerals (magnetite, hematite) plagioclase, biotitic micas, and both weathered and unweathered rock fragments mentioned above made up the magnetic portion. There were nearly equal amounts of magnetic and non-magnetic fractions. A portion of each of the above fractions was leached with 0.1N HCl to remove soluble soil materials in order to compare their behavior with the corresponding unleached portion in the column studies involving ion adsorption which are planned.

DECLASSIFIED

[REDACTED]

[REDACTED]

DECLASSIFIED

BNWL-696

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluation

Fresh radioactive fallout was detected in the Hanford environs during January. The thyroid dose increment to an infant as a result of fallout was estimated to be about 0.001 rad. An effort was made to correct a deficiency in the analytical procedure for measuring fuel oil in river water which has apparently resulted in the reporting of low results. A special program was initiated to determine the validity of results which seem to indicate that Hanford operations contribute Coliform organisms to the river.

Personnel Dosimetry

Efforts are being made to detect the low energy X rays, i.e., 0.1 to 0.2 Mev, emitted by very heavy metallic radionuclides as being emitted at the stationary whole body counter. An improved thermoluminescent finger ring dosimeter will soon be available for routine use at Hanford.

Dosimetry Studies

Development work continued on a long-term dosimeter employing thermoluminescent material, to be used by personnel

Radiation Detection and Measurement Instrumentation

Experiments with surface contoured diode radiation detectors and associated electronic circuits resulted in an acceptable tunnel diode amplifier-discriminator; similar experimental effort with germanium diodes, useful for radiation detection via internal charge multiplying effects, provided representative characteristics for the devices.

DECLASSIFIED

BNV

-77-

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluation

(J. F. Honstead, Environmental Studies Section)

Fallout in the Environment

Fallout attributed to a nuclear weapons test reached the Hanford environs in early January and again during the latter part of the month. The highest particulate concentrations found on weekly offsite air filters were 1.1 pCi/m³ at McNary Dam, of December 29, 1967, and 1.5 pCi/m³ at Yakima, of January 19, 1968. Gamma scan analyses of a few selected air filters showed that the expected short-lived nuclides were present. The highest concentrations of I¹³¹ in farm milk samples were found at Mesa, 13 pCi/l on January 3, 1968, 12 pCi/l on January 10, 1968, 18 pCi/l on January 24, 1968. The estimated thyroid dose increment for a small child drinking 1.0 liter of milk daily from the Mesa farm during January was about 4 mrem. Only small, localized increases were noted in other fallout-indicating media. The deposition, as evidenced by analyses of air filters and milk samples, was geographically uneven.

Oil Contamination in Columbia River at 100-N

Some difficulty was experienced with the analyses of river water samples for fuel oil. Apparently previous results are low as a result of incomplete extraction of the fuel oil from the sample. Efforts are under way to improve the solvent extraction-infrared procedure by incorporating part of the ASA standard method.

Coliform Measurements in Columbia River Water

Coliform organisms in samples of river water collected in the Hanford reach of the Columbia River continued to show an apparent increase over those collected above and below the site. With the small number of samples that have been collected to date and the problems associated with Coliform analyses,

An improved finger ring dosimeter designed by the Biophysical Sciences Section will soon be available for use for routine personnel dosimetry. This ring has several advantages over its predecessor. The thermoluminescent detector associated with this dosimeter is much less susceptible to atmosphere variables, e.g., humidity and heat. Also, the ring is designed to provide the maximum of comfort to the wearer.

There were 550 bioassay samples submitted for plutonium analysis during the month, including 35 from new employees who had either previously worked with plutonium or who were assigned to work with plutonium in their assignment at Hanford. In addition, 142 samples were submitted for uranium, 24 samples for promethium, 15 for americium and 6 for carbon analyses. There were 16 whole body counter or plutonium wound counter examinations conducted as a result of radiation occurrences and 5 re-examinations conducted on persons involved in previous incidents. Ten internal exposure evaluations for plutonium deposition (negative or $< 5\%$) were completed.

Special external exposure evaluations were performed for 19 Hanford contractor employees; however, no exposure over permissible limits was revealed. Five special occupational exposure summaries were prepared in response to requests from new employers of former Hanford employees.

Dosimetry Studies

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

Personnel Dosimeter System for Nonroutinely Exposed Workers

An iron-tantalum compromise position in the present badge with a TLD insert should allow the energy response of the TLD disc to be experimentally adjusted to the desired definition of penetrating radiation without modifying the badge filter system. A series of energy response measurements was made to determine the effects of iron and tantalum filters in the badge on the LiF discs to be used. When the discs are placed half over the iron and half over the tantalum filters the response to 16 keV X rays is about 35% of the response to radium

DECLASSIFIED

DECLASSIFIED

-81-

BNW-69

dose from radium gamma exposures than has been experienced from the Teflon TLD discs.

A program was initiated to further develop the activation foil system for measuring fast neutrons with TLD blocks. The initial data from PuF_4 exposures (22 to 220 mrad) was found to be random. A study was then made to observe its characteristic response to radium gamma. Again random data were obtained. Close inspection revealed that some TLD blocks had an imperfection on one surface. This could explain the random data from the fast neutron exposures. Currently a second set of fast neutron exposures is being prepared with block and Teflon TLD. Fast neutron exposures made at the Critical Mass Facility with Teflon discs in the activation foil dosimeter were encouraging.

A Conrad TLD reader modified by the addition of an analog to digital converter (ADC) and coupled typewriter was further improved. The original plan was to have independent determinations of the thermoluminescence by the original circuitry and by the additional digital hardware. However, one failure mode of the original circuit, high electrometer tube grid current, could and did cause both determinations to be erroneous. The original circuit was, therefore, removed to protect the validity of the data collected via the recently adapted digital hardware.

Because the original circuit had three ranges permitting readings up to 100 roentgens, the digital system had to be modified to provide readings to 100 roentgens without manual range changing. This was accomplished by the addition of a decade to the ADC. A decade from spare parts was used quite economically. This particular decade, however, does not have the binary coded electrical output required for the typewriter. The decade was, therefore, used in the least significant position corresponding to zero to nine milliroentgens and the typewriter coupler wired to cause a zero to be typed. Rounding off to the nearest tens of milliroentgens is considered to be acceptable in the light of the detection limit of the entire dosimeter-reader system of 15 to 30 milliroentgens. A simple sensitivity reduction by reducing photomultiplier tube gain was tried but ADC drift became too significant.

Organ Counter

A counter was designed that will simultaneously determine the amount of radioactive isotope in the pharynx, both lungs, liver and G.I. tract. The 6 in. diameter by 4 in. NaI detectors with low background phototubes were ordered. The detectors will be collimated and shielded such that each one will view only one organ. In addition to determining the amount of the isotope in each organ, the counter can be used to study the movement of an isotope in the body and should be helpful in assessing both the organ and whole body dose from the isotope.

Instrumentation Services

(W. G. Spear, Instrumentation Section)

Instrument Evaluation

Evaluation of a Ludlum Model 25 scaler-ratemeter, obtained on loan from Ludlum Measurements, Inc., continued. Initial tests revealed the ratemeter linearity and accuracy to be satisfactory for many Hanford applications. However, a major difficulty appeared during use with a NaI(Tl) scintillation probe. One turn of the single high voltage control changed the output voltage from 0 to 2500 V. Because of the rapid variation of phototube gain with voltage, it would be useful to modify the instrument by changing this control to a 10-turn unit and limiting the maximum output voltage to phototube ratings.

Wide Dynamic Range Radiation Monitoring with Surface Contoured Diode Detectors

Successful development of a wide range radiation monitoring system employing silicon surface-contoured diodes requires time and temperature stability for the amplifier-discriminator. Thus, the initial effort involves evaluation of candidate designs for sensitivity and stability. Tests on the circuit revealed the change in tunnel diode bias point voltage from free-running to stable-off to be 14 millivolts. This circuit utilized a F.E. TD251A tunnel diode and a 220 μ Hy inductor with a 20 ohm resistive load line. Changing the load line to 3 ohms resistive

DECLASSIFIED

DECLASSIFIED

just above 300 Area) which the geese do not use for some unknown reason. Locke Island had been selected for intensive studies to be conducted prior to the construction of the proposed Ben Franklin Dam.

Wintering waterfowl populations within the Hanford Reservation gradually declined during the month from about 190,000 birds to about 130,000. The major decline was due to large-scale movements of ducks out of this area as the food supply diminished. The Canada goose population fluctuated from 15,000-20,000, and there was considerable exchange between our main nesting habitat (near Richland) and the McNary Wildlife Refuge. Lack of precipitation during the preceding fall and winter months has delayed the new growth of cheatgrass which usually furnishes an important winter food for the geese. Lack of this normally available food has influenced the goose population to forage elsewhere. The first pairing of Canada geese was observed on January 3, about 1 month earlier than usual; this probably results from the unusually mild weather and predicts an early nesting season this year.

Chinook salmon eggs spawned in November and exposed to different experimental temperature regimes have completed hatching. Mortality through hatching showed no apparent relationship to the various experimental heat increments added above control water temperatures. Subsequent mortality at the critical feeding stage when the fish make the transition from absorbed yolk sac to active feeding will be of particular interest in this experiment since there has been no early mortality to complicate the interpretation of the later results.

Titers and release of columnaris organisms from control and immunized rainbow trout maintained in our hatchery have started to decrease slightly because of lack of challenge from the naturally occurring pathogen. Average titer for the immunized fish was 1-1100; for the controls, 1-300. Release of columnaris organisms from fish to trough water has decreased from two organisms per ml to zero. The drop in antibody production will be followed until columnaris season starts again in July.

DISTRIBUTION

Copy Number

USAEC, Washington-Division of Production

1-3 F. P. Baranowski

Richland Operations Office

4-6 H. E. Parker

7-8 C. L. Robinson

9 D. G. Williams

10-12 Division of Technical Information Extension

13 T. W. Ambrose

14 P. A. Carlson

15 R. Cooperstein

16 C. D. Corbit

17 R. G. Geier

18 A. E. Guay

19 C. Harrington

20 C. W. Kuhlman

21 M. Lewis

22 N. R. Miller

23 W. S. Nechodom

24 R. Nilson

25 G. F. Owsley

26 R. S. Peterson

27 R. W. Reid

28 D. L. Renberger

29 J. W. Riches

30 O. C. Schroeder

31 R. H. Shoemaker

32 J. R. Spink

33 H. F. Tew

69	G. A. Last
70	B. R. Leonard
71	J. E. Minor
72	R. L. Moore
73	C. E. Newton, Jr.
74	J. M. Nielsen
75	R. E. Nightingale
76	R. S. Paul
77	D. W. Pearce
78	A. M. Platt
79	W. D. Richmond
80	P. T. Santilli
81	L. C. Schmid
82	W. W. Schulz
83	W. G. Spear
84	G. L. Tingey
85	C. R. Tipton, Jr.
86	C. M. Unruh
87	E. E. Voiland
88	R. G. Wheeler
89	O. J. Wick
90	D. C. Worlton
91	H. H. Yoshikawa
92-93	Technical Information Files

DATE

FILMED

7 / 8 / 94

END

+