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BNWL-302

C-65, Plutonium Production  
(Special Distribution)

This document consists  
of 58 pages.

PACIFIC NORTHWEST LABORATORY  
MONTHLY ACTIVITIES REPORT  
JULY 1966

Classification Cancelled and Changed To

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Division  
of

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DS Views, 2-8-94,  
By J. M. Mally 3-11-94,  
Reviewed By J. E. Sauley 3-22-94

Production and Hanford Plant Assistance Program

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REPOSITORY PNL  
COLLECTION I-131 Atmosphere  
BOX No. D/A  
FOLDER D/A

August, 1966

NOTICE: PRELIMINARY REPORT

This report contains information of a preliminary nature prepared in the course of work under Atomic Energy Commission Contract AT(45-1)-183. This information is subject to correction or modification upon the collection and evaluation of additional data.

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RICHLAND, WASHINGTON

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

R. S. PAUL

DIRECT AEC SPONSORED PROGRAMS

SUMMARY

COLUMBIA RIVER STUDIES

A new mathematical simulation model to handle reactor effluent dispersion was started.

Final arrangements were concluded for the proposed test of the effectiveness of activated carbon for inorganic ion removal at the Richland Water Plant.

EXPOSURE MECHANISMS

The whole body counter used to measure the burdens of radionuclides in Richland school children is being calibrated for persons of small size and with different weight to height ratios. Controlled intake of oysters that have accumulated  $Zn^{65}$  is being used as one phase of this calibration.

Persons eating beef reared in the Riverview Irrigation District gained about 4 nanocuries of  $Zn^{65}$  after about four months and the consumption of 20 to 28 pounds of the beef.

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DIRECT AEC SPONSORED PROGRAMS

COLUMBIA RIVER STUDIES

Effect of Reactor Effluent on the Quality of Columbia River Water  
(Environmental Studies Section)

Some progress in organization of the test to determine the effectiveness of activated carbon for the removal of selected dissolved inorganic ions and total organic material was made despite strike difficulties. The experimental procedure was reviewed and agreed to by the City of Richland on a line by line basis. Radiation monitoring equipment is being assembled and all city sponsored modification of the plant is complete. Tests were planned for the months of August and September. Chemical blanks are being run to provide a valid comparison between city analytical procedures and plant procedures.

Initial steps were taken to prepare a new digital simulation model of the effluent dispersion of the Hanford reactors in a suitable format for potential use at other locations. The proposed program is being developed on the basis of solution of a matrix of difference equations with escape and entry probability adjustment among regions selected on a dimensionless equal energy distribution. The approach seeks to replace the use of field coefficients with a more general expression derived from theory.

EXPOSURE MECHANISMS

(Environmental Studies Section)

Whole body counting data that were obtained for about 1,000 school children in Richland require special calibration factors. These factors need to be adjusted for the physical size of each child. To do this, a calibration curve will be required for each radionuclide of interest, which relates calibration factor to physical dimensions. It is possible to construct such curves using phantoms of various sizes assembled from one pound packages of sugar. However, the validity of such curves is questionable unless verified with human data. We hope to obtain human calibration data for one radionuclide to verify the results from phantoms and thus establish a reliable phantom design for constructing other calibration curves. In particular, we will compare phantom and human data for calibrating the whole body counter for  $Zn^{65}$ .

Six volunteers were utilized to obtain  $Zn^{65}$  whole body counting data for calibration purposes. They were given several meals of oysters,

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totaling about 1,000 g for each person, and a careful record kept of the amount of  $Zn^{65}$  consumed. The oysters contained about 45 pCi  $Zn^{65}$  per g. The amount of  $Zn^{65}$  absorbed in each case was determined from excreta collection and analysis. The whole body counter response to each of these subjects was then used to determine the appropriate calibration factor for the individual. The individuals used in the experiment ranged from children having 1.0 lb body weight per in. of height to adults having 2.5 lb body weight per in. of height. The calculations needed to analyze these data are not yet completed.

An experiment to study the uptake of  $Zn^{65}$  from beef raised on a pasture irrigated with Columbia River water was completed except for continuing whole body measurements of  $Zn^{65}$  in the subjects who consumed the beef. The meat had a very low  $Zn^{65}$  concentration, which makes it difficult to detect the effect of three meals of beef per week on the subjects'  $Zn^{65}$  body burdens. After about four months of beef consumption, in which each subject consumed 20 to 28 lb of beef, they showed an average increase in  $Zn^{65}$  body burden of 4.1 nCi. This is of the same order as that normally encountered in Richland residents resulting from drinking water and normal diet. A more detailed analysis of the results will be required to interpret them and to calculate the average fractional absorption.

Work continued in the Statistics Section on the analysis of survey data obtained from sport fishermen on the Columbia River. This study is being conducted in cooperation with the Washington State Game Department.

#### ADVANCED CONCEPTS

##### Epimetheus

(Advanced Concepts and Analysis Section)

The computer runs from the economics chain which yield parameters for VESTA input were completed and the VESTA runs made. The output appears good, but has not yet been examined in detail. The data should complete the calculation of selected by-product isotopes from operation of FWR and HWGR types in the projected nuclear power industry. The code prepares a table of the availability of these isotopes with time over the next 60 years.

##### $Cm^{244}$ Production

(Advanced Concepts and Analysis Section)

A computer program designed to calculate the possible production of Cm isotopes as a function of time from both Richland and Savannah River reactors was developed by modifying the existing fuel element fabrication

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code. This program is now in the final debugging process. It makes available the Cm, Am, and Pu quantities and assays at any time along with the total program cost. The computer program uses a special burnup routine supplied by DUN, and a special separations routine supplied by Isochem. In the future Savannah River will supply similar routines describing their operations.

Isotopic Data Tape  
(Advanced Concepts and Analysis Section)

The monitor system problem which has prevented extensive use of this program is still present. When this program is run, no other programs can be executed on the UNIVAC 1107 until the Exec II system is reentered into the computer.

A document describing the data tape and its use was prepared and is being typed.

U<sup>235</sup> ANALYZER  
(Instrumentation Section)

Development of specific solid state circuits continued at a satisfactory pace for the portable gamma spectrometer being developed for use by the AEC-DIA in measuring the U<sup>235</sup> content of unirradiated reactor fuel elements.

Improvements incorporated in the gain stabilization portion of the spectrometer served to simplify initial adjustments needed to indicate gain in an accurate fashion. To achieve this, a differential counting rate circuit was employed to indicate the difference counting rate between two adjacent reference pulse height analysis channels. Initial gain adjustment is accomplished by nulling the difference rate. In addition, drafting work was initiated on the developed circuits and for the instrument panel, cabinet, and detector housing.

Specific solid state circuits developed and tested to date include the double-delay line pulse amplifier, the gain stabilizer, all of the counting rate circuits, and the high voltage supply.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

SUMMARY

MISSION 1\*

No measurable difference was observed in diffusion bonding of nickel-aluminum with either the direct plate or a zincate pretreatment.

MISSION 10

Experiments were made in the PCTR on two configurations of the 3/1 ratio supercell consisting of 3 enriched (1.25%) drivers and 1 thorium target. There is a small but detectable difference in the measured multiplication factor for the two configurations.

A new experiment was started in the PCTR with the same three enriched uranium drivers to one thorium target array as usual in the mixed lattice study. However, the diameter of the driver fuel was reduced slightly thus making a change in the amount of fuel and water coolant in the supercell.

A need for analyzing plutonium-thorium supercells has suggested the development of Kernel Superposition Theory, an extension of Small Source Theory and Heterogeneous Reactor Theory. This approach has successfully predicted reactivity measurements in the PCTR.

A program is being prepared to utilize the punched paper tape output of the new automatic foil counting equipment that is to be delivered in October.

An improvement of the thermal neutron flux intensity in the PCTR thermal column is being attempted by surrounding the thermal column with polyethylene.

MISSION 11

Research in code development section measurements is supported by both Douglas United Nuclear Incorporated and by General Electric N-Reactor Department. It is also funded under both Mission 11 and Mission 16. Monthly contributions appear under Mission 16 in "Assistance to General

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\* For Cross Section measurements partly sponsored by DUN, see page 21.

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Electric N-Reactor Department," see page 27.

MISSION 13

The second full-size, aluminum jacketed irradiated uranium fuel element was thermally stressed to failure. Physical changes during the heating cycle were similar to those observed in the earlier test.

Plutonium oxide particles released from ignition of a large ingot of plutonium were found to have a count mean diameter of  $1.2\mu$ .

MISSION 14

Radiochemical analyses of radioarsenic in the effluent from two tubes cooled with water containing added As provided data from which the kinetics of the sorption-desorption reactions can be evaluated under conditions of normal operation and reactor shutdown.

BNW computer program LEARN (a non-linear least squares program) will be used, with theoretical derivations for radionuclide formation in single pass reactor coolant tubes to correlate the data of the recently completed As addition to KE Reactor.

PROCESS TECHNOLOGY

Monitoring results indicate that the rate of graphite oxidation has increased at B, C, and D Reactors.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

MISSION 1 - BASIC PRODUCTION MISSION

Uranium Nickel Plating Studies  
(Chemical Separations Unit)

A study of some fundamental properties of systems for "pulsed" nickel plating was undertaken to establish values for process variables to be used in initial tests. Measurements of overvoltages and polarization times were made under various conditions. The overvoltages of sulfamate and Watts plating baths and of a 3M nickel perchlorate solution were compared and found to be highest in the sulfamate solution. Measured polarization times were greater in concentrated nickel perchlorate solution than in the Watts (dilute nickel) bath. Polarization times are also increased by increasing the length of time between pulses. The effects of agitation by moderate stirring and by ultrasonics were compared in the Watts bath. Measured overvoltages were about the same in the two cases, probably because the energy density transmitted by available ultrasonic transducers (0.3 w/cm<sup>2</sup> at 40 Kc) was not great enough.

Fuel Plating Studies  
(Water Reactor Corrosion and Chemistry Unit)

Adherent Ni coatings were consistently obtained by electrodeposition on a 8001 Al alloy tubing from a fluoborate nickel bath. Samples plated (1) directly on Al, (2) over a commercial zincate pretreatment, and (3) over a laboratory compounded zincate pretreatment were heat treated at 350 and 400°C. Metallurgical examination of the samples did not reveal any measurable differences between direct plate and zincate pretreatment in obtaining a diffusion bond.

MISSION 10 - HIGH POWER DENSITY FUEL

Mixed Lattice Experiments in the PCTR  
(Reactor Lattice Physics Section)

An enriched fuel and thoria target experiment was started in the PCTR when a 3:1 target ratio was loaded into a 5 x 5 square arrangement of graphite blocks with a 7.5 in. K lattice pitch.

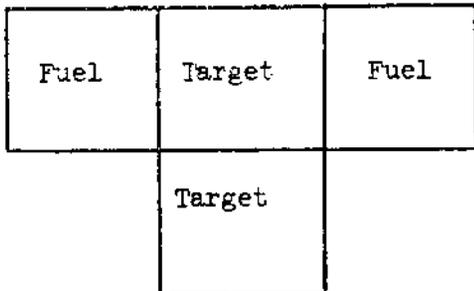
In order to derive the neutron multiplication factor of the super-cell, with and without coolant, the mass of Cu necessary to compensate the reactivity effect of each set of four cells was measured for two different

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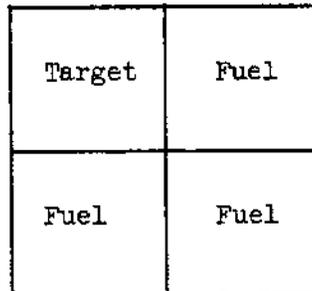
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fuel to target geometrical relationships. The two fuel-to-target geometries are illustrated in the following:



Removable Section for PCTR with 5 x 5 Test Core



Removable Section for PCTR with 6 x 6 Test Core

Due to the thoria being off-center in the square arrangement of the center cell, it was necessary to build a 6 x 6 graphite test core in the PCTR for the second set of measurements.

Preliminary results of these measurements indicate a 2.2% lower equilibrium neutron spectrum Cd ratio value in the 6 x 6 test core. The correct value of the mass of Cu necessary to poison the cell to a neutron multiplication of unity increased by 10.7% in the 6 x 6 test core (i.e., approximately 763 g vs. 689 g). Additional preliminary results are listed in the following table:

| <u>Test Condition</u> | <u>Cadmium Ratios</u>  |                        |
|-----------------------|------------------------|------------------------|
|                       | <u>5 x 5 Test Core</u> | <u>6 x 6 Test Core</u> |
| Wet-Poisoned          | 5.203                  | 4.910                  |
| Wet-Unpoisoned        | 5.155                  | Not Derived            |
| Dry-Poisoned          | 3.900                  | Not Derived            |
| Dry-Unpoisoned        | 4.059                  | Not Derived            |

|             | <u>Mass of Copper Necessary for Unit Multiplication</u> |                        |
|-------------|---|------------------------|
|             | <u>5 x 5 Test Core</u>                                  | <u>6 x 6 Test Core</u> |
| Wet (grams) | 689   | 763                    |
| Dry (grams) | 993   | Not Derived            |

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3 to 1 SUPERCELL EXPERIMENT in the PCTR  
(Reactor Lattice Physics Section)

Experimental supercell measurements were started in the PCTR using three 1.25% enriched drivers of a reduced O.D. to each thorium target in a 7½ in. pitch graphite lattice. These experiments are nearly identical to the "Mixed Lattice Experiments in the PCTR" (see preceding paragraphs), except that the three uranium columns in the test supercell have reduced outer diameters. Reactivity measurements are being made in the perturbed lattices which, along with the previous experiments, will give the infinite medium neutron multiplication factors  $k_{\infty}$  in the six configurations under study.

Measurements were completed in the first two configurations. The 5 mil Au Cd ratio and the correct mass of Cu to poison the test supercell to a  $k_{\infty}$  of unity were measured in the completely dry case. The Cu mass was also measured in the case with all tubes wet except three dry U columns in the test supercell.

Analysis of Plutonium-Thorium Supercells by Kernel Superposition Theory  
(Theoretical Physics Unit)

Kernel Superposition Theory, an extension of Small Source Theory and Heterogeneous Reactor Theory, is being used to analyze PCTR experiments on Pu-Th rod supercells. Basically, the theory starts with the Feinberg method of superposing production and loss kernels for the thermal flux. Previous workers have formulated the kernels on a pseudo-variational basis, but have lost the possible gains by taking a delta-function adjoint at the rod surface. A flat adjoint is now used, since it is applicable for moderate absorbers, and production and loss kernels are averaged over the volume of each rod. Such averaging tends to minimize the errors in reactivity due to errors in the flux, i.e., a pseudo-variational method. An eigenvalue matrix is solved for the static reactivity and absorption rates for each rod type. Specifically,  $k_{\infty}$  for a one-to-one cell ratio, infinite lattice experiment in the PCTR, including multi-sectional fuel and absorber rods has been correctly predicted by the Kernel Superposition Theory.

The flux shape between a Th and a Pu rod has not been well predicted as yet, possibly because the center-to-center distance is used for rod-to-rod kernels, while self-interaction is handled by evaluation at the rod surface. Thus, there may be a linear shifting of the flux coordinates. A too-flat thermal flux shape has been found, which is similar to the experience of Savannah River. The flux calculation is, however, actually independent of the reactivity and relative absorption rate calculations.

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Several other experiments are available which will be used to check out the theory.

Analysis of PCTR Experiments Using KVE Fuel  
(Reactor Lattice Physics Section)

Analysis of the experiments continued to determine the infinite medium multiplication factor and thermal utilization of wet and dry KVE fuel in a  $7\frac{1}{2}$  in. graphite lattice. Graphs were prepared for axial Au and Cu, and radial Au Cd ratio matching for wet and dry, poisoned and unpoisoned cases. It also includes plots of  $M_0$  vs.  $\frac{1}{\text{Cad Ratio} - 1}$  for wet and dry poisoned cases. Data were on hand to complete the two experimental  $\frac{1}{v}$  flux plots. Finally, computer solutions were obtained for the thermal flux distributions in the two unpoisoned cases, and only the two poisoned cases remain to be run before the latest graphs are completed. General calculations neared completion.

Data Conversion for the New Automatic Foil Counting System  
(Reactor Lattice Physics Section)

The amount and type of programming necessary to convert data from the new automatic foil counting system's punch paper tape to the usual deck of data cards for input to APDAC was investigated. The cards are desired in order to allow removal or correction of poor or incorrect individual counts. With the help of Computer Sciences Corporation's existing paper tape to tape program and the various options and blank routines available, there appears to be no insurmountable problem involved. A small addition to the original counting system was ordered which will allow the manual punching of additional data on the tape prior to the actual counting. Some problems still remain in fitting residual activity data into the program.

PCTR Thermal Column Study  
(Reactor Lattice Physics Section)

A thermal column study was undertaken on the PCTR to determine the flux change in the column when the sides and top are wrapped with polyethylene and Lucite sheets. At present a relatively weak flux is observed in the column, probably largely due to leakage of neutrons from the sides. Approximately 2 in. of reflector will be applied to the sides of the column, with a lesser amount on the top. Both horizontal and vertical traverses will be taken and Cd ratios will be calculated. Three measurements were planned: one with the column as is, one with reflector

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in place, and one with reflector in place and a void near the center of the column. The purpose of the void in the last measurement is to try to flatten the flux over the region in which foils are usually irradiated.

MISSION 13 - NUCLEAR SAFETY

Fission Product Release from Overheated Fuel  
(Particulate and Gaseous Waste Research Unit)

The second full-size, Al jacketed irradiated U fuel element was heated in a steam atmosphere. The 939 MWD/T material was programmed from 380 to 1010°C in 11 min and held at that temperature for 20 min. This test was similar to the first except that the heating period to goal temperature was shortened 10 min and the time at 1010°C extended 10 min for an equal heating period. The three fission product sampling periods were first to 665°C (about 6 min), second from 665 to 810°C (about 2 min), and third from 810°C to end of cooling period (about 45 min).

There were no significant visible physical differences between the two fuels during or after the heat cycle. The first molten Al appeared again at 620°C with no copious flow. Swelling (greatest in the regions of highest Al concentration) and rupture of the U-Al skull and fission product releases at the two phase changes occurred as in the first test. The amount of U oxide formed was greater as would be expected from the longer period at the goal temperature and the exothermic U-Al reaction was again sufficient to maintain the temperature increase through the phase changes.

Only the Xe and Kr data were completed for the samples of the first test. The release fractions were 3.6% for Xe<sup>131m</sup> and 1.2% for Kr<sup>85</sup>. The Xe release occurred as follows: 2.5% below 765°C, 8.5% between 765 and 1010°C, and 89% during the 10 min period at 1010°C plus the cooling period. Fractions of the Kr released for the same periods were 0.1%, 12%, and 88%.

Other than the fractional release of fission products, the most significant information obtained from the first two irradiated fuel tests was: (1) the absence of flowing Al, (2) the influence of the U-Al reaction on the rate of heating, and (3) the rupturing of the UAl<sub>4</sub> skull and resultant oxidation of the U. The first two relate to the calculated fuel temperature and suggest that the predicted maximum temperature might be low if not offset by other conservative assumptions. A higher maximum temperature and greater U oxidation point to a higher fission product release than that which might have been predicted from the tests with unirradiated fuels. The release of fission gases at the α-β and β-λ phase changes confirms the data collected by a few investigators during annealing or diffusion studies with small specimens.

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MISSION 14 - COLUMBIA RIVER STUDIES

Arsenic Addition

(Water Reactor Corrosion & Chemistry Unit)

For a period of 27 days, approximately 3 ppb of As was added to the process water cooling two tubes in the KE Reactor. The effluent radioarsenic concentrations from the two tubes were monitored both during this interval and for eleven days after the addition was discontinued. The data, and future analyses of the inert As concentrations existent in the effluent, will be applied to the interpretation of the sorption-desorption reactions occurring in the Al and Zr process tubes.

A reactor shutdown occurred during the addition portion of this experiment. The effluent radioarsenic concentration immediately decreased to about 10% of the pre-shutdown level, and attained a steady release rate approximately two days after shutdown. This behavior was different from that which occurred after the As addition was terminated, when a more gradual decrease in effluent As<sup>76</sup> concentration was measured. Comprehensive evaluation of the data is required, but these opposing patterns must reflect the influences of temperature and neutron flux on the sorption-desorption reactions.

Effluent Control Program

(Advanced Systems Corrosion Unit)

The theoretical derivation for the study of radionuclide formation and deposition in-reactor has been combined with the BNW program LEARN (BNWC-86B). Program LEARN will be used to fit the derivation to the data obtained from the recent As addition test in KE Reactor.

PROCESS TECHNOLOGY

Iodine Adsorber Bed Evaluation

(Particulate and Gaseous Waste Research Unit)

A tentative program was outlined to evaluate the performance of individual charcoal iodine collectors using an existing facility. Facility modifications were determined and necessary changes indicated to DUN. The objective will be to determine the unit collector efficiency for radioiodine under simulated reactor accident conditions of air moisture and temperature.

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Burnout Monitoring - B, C, and D Reactors  
(Ceramics and Graphite Research Section)

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| Channel | Charge | Dates              | Type of Graphite | Maximum Oxidation |   |
|---------|--------|--------------------|------------------|-------------------|---|
|         |        |                    |                  | Location/rate*    | Location/rate                                   |
| 3461-B  | 24     | 1-21-66 to 6-2-66  | KS               | 120"/7.5%         | 165"/7.5%                                       |
|         |        |                    | TSGBF            | -- below 0.5%     | --  |
| 3789-B  | 1      | 1-21-66 to 6-2-66  | KS               | 150"/0.5%         | below 0.55%                                     |
|         |        |                    | TSGBF            | -- scattered      | --  |
| 1889-C  | 12     | 2-10-66 to 6-13-66 | KS               | 165"/28%          | Half of the samples were lost during discharge. |
| 1960-C  | 21     | 2-10-66 to 6-13-66 | KS               | 130"/25%          | 165"/10%  |
|         |        |                    | TSGBF            | 120"/0.6%         | 165"/0.6%                                       |
| 1675-D  | 1      | 2-13-66 to 5-26-66 | KS               | 120"/4.0%         | 200"/39.6+%                                     |
|         |        |                    | TSGBF            | 120"/0.3%         | 175"/0.8+%                                      |
| 3478-D  | 24     | 2-13-66 to 5-26-66 | KS               | 120"/200%         | 165"/450%                                       |
|         |        |                    | TSGBF            | 130"/40%          | 165"/67%  |
| 1675-D  | 2      | 5-26-66 to 6-28-66 | KS               | 120"/1%           | 200"/28+%                                       |
|         |        |                    | TSGBF            | --                | 200"/1.5%                                       |
| 3478-D  | 25     | 5-26-66 to 6-28-66 | KS               | 120"/140%         | 165"/1190%                                      |

\* Rate is expressed as percent per thousand operating days.

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Radiation Detection and Measurement Instrumentation  
(Instrumentation Section)

Successful field tests on the BNW-modified, pocket-size, gamma detection instrument, which provides an aural signal when the preset gamma level is exceeded, have led to a request for similar modification of several more units.

The experimental pocket-size, meter-indicating gamma dose rate instrument was completed in breadboard form with chassis-mounted, encapsulated circuit modules. This unit, which provides an aural signal when the preset gamma level is exceeded, was delivered to the field for demonstration and testing. The final prototype instrument will be designed to incorporate any changes determined through the field tests.

Ultrasonic Inspection of Cooling Tubes  
(Nondestructive Testing Section)

Ultrasonic transducers, with improved resolution, were developed for measuring the thickness of stainless steel, graphite cooling tubes. A simple technique of applying a destructive lens to the front surface of a transducer was successfully employed. A spray applicator was used for depositing silver loaded epoxy on the front surface of a 5 MHz transducer, and the ringing immediately following the transmitter pulse spike was monitored. An improvement of 300 to 400% in the damping factor was noted after the sprayed-on lens technique had been optimized. This resulted in a transmitted pulse of only about 3 cycles of the transducer resonant frequency. It is expected that this ratio of cycles per pulse will remain constant for all frequencies, and higher frequency elements should provide proportionately higher thickness resolution. However, a compromise between frequency and surface finish will be necessary in practical applications. The resolution of the destructive lensed 5 MHz element was found to be quite ample for the current graphite cooling tube problem.

Thickness Measurements of Sulfuric Acid Facilities  
(Nondestructive Testing Section)

It was desired to survey the wall thickness of various tanks and pipe lines in the 183-KW sulfuric acid facilities. Measurements were taken on the acid head tank located on the roof of the 183 Building, on transfer lines leading to the storage tank area, and on one storage tank. The survey showed no evidence of any serious wall thinning on the transfer lines and storage tank; however, the acid head tank did show thinning of about 50% near the top of the tank.

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ADVANCED PLANNING

(Statistics Section and Mathematical Analysis Section)

A study was continued to approximate each of several reactor response variables with a polynomial in several input variables. This effort is part of a production cost analysis study.

Considerable progress was made on the development of an accept or reject decision method for strap-on type resistance temperature detectors (RTD's). The method depends upon the fitting of discrete data which have been recorded by an immersion type RTD. This fitting technique has been completed and a computer program written and satisfactorily tested.

The data for this fitting program will exist on a 7090 Fortran II binary tape since it is part of an automatic recording and data processing report in present use. There now remains the problem of converting the data to a form which is compatible with the Univac 1107.

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ASSISTANCE TO GENERAL ELECTRIC N-REACTOR DEPARTMENT

SUMMARY

MISSION 1

Analysis and theoretical interpretation continued on the existing scattering data for ice and water.

The task of assembling the results of four years of  $\sigma_t(E)$  measurements, prior to a massive publication endeavor, was continued.

Work continued on the compilation and evaluation of nuclear data on a dozen isotopes for the AEC Evaluated Nuclear Data File/B.

Examination of N-Fuel failures #18, 19 and 21 continued. Failures #18 and 19 are end associated, and end caps will be examined for porosity. Failure #21 is a fretting failure caused by the buggy spring supports on a co-product target element. Metallographic examination of a high exposure N-Fuel assembly is in progress.

Burst tests on two specimens near the ends of a discharged N-reactor process tube show a reduction in elongation but no significant change in ultimate strength as compared with as-installed material. The fracture in one of the specimens passed through two fret marks caused by the feet of the downstream dummies.

MISSION 2

Radiometallurgy examination continued on Al canned co-product targets containing  $\text{LiAlO}_2$  ceramic cores. Metallographic examination revealed no impairment of the integrity of the targets from bulging observed in the Al caps as a result of internal gas pressure. Free gas released from targets with lithium aluminate cores fabricated by high energy direct encapsulation and by vibrational compaction of bulk sintered powder was 344 and 980  $\text{cm}^3$  STP, respectively.

Alternate support designs for use on co-product fuels are being developed. An end spider in the form of a wheel with four curved spokes appears to be capable of meeting the design criteria for this type of support. Mechanical testing of supports of this geometry is underway.

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A special flowmeter was designed and constructed to allow the rapid measurement of flow splits between the inner and outer flow annuli in a column of N-Reactor coproduct elements. The flowmeter will be installed in the NPR-PCE loop and will be used to evaluate the effects of various coproduct fuel element supports on the coolant channel flow splits.

#### MISSIONS 1, 2, 4, 6, 10

Examination of Zircaloy-2 clad alloy fuel rods irradiated in NaK capsules continued. Density measurements on 79 rods and metallography on fuel rods of two compositions were completed. Uranium containing relatively high Fe and Al additions is the most resistant to fuel swelling of the 17 fuel compositions tested.

#### MISSION 16

An improved method for calculating resonance absorption cross sections in the HRG slowing down code yields effective cross sections 5 to 10% larger than the original treatment for a typical cell. The values are within 1% of those obtained from a relatively elaborate numerical integration.

The BNW Master Library System with its associated processing codes was modified to store resonance parameter tables with each isotope as well as at the beginning of the library tape. The system was also altered to correctly treat negative (bound state) resonances. The isotopes Fe<sup>56</sup>, C<sup>12</sup>, and Cu<sup>65</sup> were updated.

A standard Battelle version of the THERMOS code was created by introducing into the original code developments by various authors.

#### PROCESS TECHNOLOGY

Visual inspections of two Inconel tubes removed from steam generator 4A showed that no observable corrosion was present. The oxide appeared to be in excellent condition.

Pitting up to 1/16 in. diam was found in steel process tube nozzles.

Assistance continued in the preparation of LiAlO<sub>2</sub> pellets for co-product target elements and in the drawing of Zircaloy-2 tubing to finished dimensions for use as cladding material for the co-product target elements.

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ASSISTANCE TO GENERAL ELECTRIC N-REACTOR DEPARTMENT

MISSION 1 - BASIC PRODUCTION MISSION\*

Slow Neutron Scattering#  
(Neutron Physics Section)

Approximately 90% of the existing water and ice scattering data have been reduced to double-differential cross sections and the scattering law. Some progress has been made in the analysis and theoretical interpretations.

Fast Neutron Cross Sections#  
(Neutron Physics Section)

Effort continued on a paper which presents evidence for the non-existence of a bound state of the di-neutron. The evidence is in the form of absence of "Wigner Cusp Anomalies" in the n-d total cross section. These anomalies should have appeared at the energy threshold for di-neutron production, if the di-neutron existed. The paper is about 80% completed.

Work continued on the task of assembling the  $\sigma_t(E)$  results preparatory to the massive publication endeavor.

Cross Section Evaluation#  
(Neutron Physics Section)

Work continued on the evaluation of nuclear data on a dozen isotopes for the AEC Evaluated Nuclear Data File/B. Progress to date has been primarily in the accumulation of recent data and compilations. Some comparisons have been made of total cross sections in the MeV region. The experimental measurements of Foster and Glasgow have been found to depart significantly from the predictions given in Howerton's semi-empirical compilation in the rare earth region.

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- \* See also page 26 for additional reporting on this Mission.
  - # Partially supported by Douglas United Nuclear Incorporated.

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N-Reactor Fuel Failure Analyses  
(Metallurgy Design Unit)

The end cap from Failure #18 (inner component) is being examined to determine its role in the failure. Superficial examination has revealed nothing unusual or suspicious about the cap. The eutectic braze-weld closure is being examined in detail as the cap is being progressively sectioned in the transverse plane by grinding and metallographic preparation.

Failure #19, less its cap, was received for analysis. Its appearance is nearly identical to that of #18, and superficial examination revealed nothing unusual nor suggested any reason for failure of the inner component. Analysis of the failure is in abeyance pending location of the missing cap.

Failure #21 occurred in a co-product driver element and the rupture appears to be the result of a target element support fretting through the clad of the driver at the unclipped end of the target. The target associated with this rupture has been examined in Radiometallurgy and it was observed that two of the three supports on the unclipped end of the target had suffered severe fretting corrosion. The crown on one support was completely missing and the crown of the other support was thinned by the fretting attack. This element was a part of production test No. PT 66. It had been assembled by hand-sizing the target supports to position and fix the target component within the driver.

Surveillance of N Fuel Behavior  
(Metallurgy Design Unit)

An N fuel element assembly from the center of Tube 1757 (PTNR 4 Sup. A) is being examined in Radiometallurgy to characterize the behavior of N fuel (100-150 ppm Fe and Si additions). The calculated fuel exposures are 5200 MWD/T for the outer component and 4000 MWD/T for the inner. Postirradiation density measurements made on the fuel indicate 4.0 vol.% fuel swelling in the outer component and 3.0 vol.% in the inner. The theoretical minimum volume increase to accommodate solid fission products is 1.8% for the outer and 1.4% for the inner. The uranium microstructure in both the inner and outer elements was uniform across transverse sections. The structure was severely distorted and had no similarity to the initial microstructure. No grain boundary tearing or other porosity is resolved by optical microscopy. Metallographic surfaces are being replicated for electron microscopy.

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Pressure Tube Evaluation  
(Engineering Materials and Mechanics Section)

Two specimens from the recently discharged N-reactor pressure tube were burst-tested. One (#1) was from the extreme upstream end of the tube and the other (#2) near the downstream end. The #2 specimen contained a number of fret marks about 10 mils deep produced by the feet on the downstream dummies. The results are summarized in the following table:

|                          | <u>Ultimate<br/>Hoop Stress<br/>(psi)</u> | <u>Elongation<br/>%</u> |
|--------------------------|---|-------------------------|
| Upstream Specimen (#1)   | 111,300                                   | 5.3                     |
| Downstream Specimen (#2) | 108,500                                   | 10.2                    |
| As-installed Tubing      | 109,000                                   | 21                      |

These specimens have received little or no irradiation but have been subjected to the thermal and chemical effects of the coolant. The ultimate hoop stress has been virtually unaffected, but the elongation has been markedly reduced. The failure in the #2 specimen passed squarely through two of the fret marks that were in line axially and about 4 inches apart. These surface defects may have been a factor in determining the location of the tube failure, but they caused no significant reduction in strength.

MISSION 2 - COPRODUCT MISSION\*

Examination of Irradiated Coproduct Target Elements  
(Fuel Materials Development Unit)

Coproduct targets of lithium aluminate ( $\text{LiAlO}_2$ ) ceramic cores double-canned in Al and Zr-2 were irradiated to characterize their behavior and to provide irradiated core materials for product extraction tests at Savannah River Laboratory. Two 12 in. long by 1.25 in. OD Al canned targets are contained in each 24.4 in. long by 1.44 in. OD Zr-2 can. Gas volumes and samples from the annulus between the Zr-2 and the inner Al cans were obtained on two targets DE 033, 036 and SV 240, 247. These samplings were obtained to determine if any product gas had leaked or diffused out of the inner aluminum cans. Gas volumes obtained were, respectively, 9.6 and 16.3  $\text{cm}^3$  at STP. The tritium content of these samples was as follows:

\* See also page 26 for additional reporting on this Mission.

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|             | <u>T<sub>2</sub></u> | <u>HT</u> |
|-------------|----------------------|-----------|
| DE 033, 036 | <0.01                | <0.01     |
| SV 240, 247 | <0.01                | <0.03     |

These results show no significant leakage or diffusion of product through the Al cans.

Two Al canned targets, SP-169 and SP-152, were examined in the Radio-metallurgy Facility and found to have definite bulges in one end cap of each target. The bulging was attributed to internal pressure from T and He generated and released from the ceramic cores during irradiation. Metallographic examination of the weld areas of the caps showed no evidence of cracking or tearing which would lead to failure or leakage of the target in the weld. Two additional Al canned LiAlO<sub>2</sub> targets, DE 036 and SV 247, were drilled to measure and collect the contained gas. Each of these targets had a bulge in one end cap. Target DE 036 was fabricated using high energy impaction techniques to yield a compact approximately 88% of theoretical density ( $\rho$  theoretical 2.62 g/cc). The free gas volume measured in this element was 344 cm<sup>3</sup> at STP. Target SV 247 was fabricated by vibrational compaction of LiAlO<sub>2</sub> which had previously been bulk sintered, crushed, and screened. Overall density of the vibrationally compacted target was 68.7%. Sintered density of the particles was 82.8%. The free gas volume measured in this target was 980 cm<sup>3</sup> at STP.

Target Support Development  
(Metallurgy Design Unit)

Design criteria applicable to an alternate support for coproduct fuel were established to be as follows: (1) No rubbing of any Zr surface, (2) an endurance limit of 15 lbs, based on 10<sup>8</sup> cycles, and (3) an elastic axial deflection of 0.050 in. after an initial deflection of 0.150 in.

The first criterion is intended to eliminate the fretting problem which has been associated with spring-loaded support systems. This requirement practically dictates that supports for the target have welded attachments to both components. The second criterion is based in part on assumptions concerning service conditions and in part on comparative fatigue strengths of other successful fuels. The third criterion is based on estimates of maximum anticipated axial deflections resulting from growth, thermal expansion and manufacturing tolerances.

Various end-spider designs were evaluated with respect to the above criteria. One design, which would be fabricated from sheet material,

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appears to be capable of meeting the criteria and has manufacturing feasibility. This support is in the form of a wheel with four curved spokes. The hub is welded to the target and the rim to the driver. This type of spider, fabricated from 0.050 in. sheet and with spokes approximately 0.080 in. width, is the subject of current tests. As with any of the end-spider designs which have production feasibility, this concept more critically stresses Zircaloy-2 weld metal than do existing support systems, and fatigue tests of the welded assemblies provide the most important initial information.

The end support structures based on wire elements were found to have marginal fatigue resistance at the points of weld attachment. Further development of this concept has been deferred in favor of the sheet metal parts which allow more latitude in accomplishing the welded attachment.

Other contributions to the coproduct problem included a parametric study of the characteristics of uniform and tapered leaf-spring type supports, statistical testing of the properties of buggy-spring support configurations, and a preliminary analysis of an iron-Zircaloy bimetal support concept.

Flow Distribution Measurements for N-Reactor  
(Reactor Engineering Section)

A flowmeter was designed and constructed to allow the rapid measurement of the flow split between the inner and outer flow annuli in a column of N-Reactor coproduct elements. This flowmeter is to be installed at the downstream end of a normal coproduct element charge in the NPR-PCE loop. It consists of a solid cylinder centered in an outer tube. Over most of the 20.5 in. length of the assembly, these components have the same diameters as the target cylinder and driver tube in the coproduct element at normal in-reactor coolant temperatures. The OD's of both the cylinder and tube are expanded over a short length near the middle of the assembly to provide a reduced flow area. In effect, this assembly is an "annular venturi tube." Pressure taps in the inner cylinder allow measurement of the pressure differential between points in and upstream of the venturi throat in the annulus between the solid cylinder and the outer tube. The expansion of the outer tube surface was designed to balance flow resistances in the inner and outer annuli and, therefore, to prevent the flowmeter assembly from disturbing the normal column flow split.

In the flow distribution measurement experiments, the annular venturi will be used to measure flow in the inner annulus between target

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and driver. A calibrated N-Reactor venturi will be used to measure the total flow to the process tube and the flow in the outer annulus will be found by subtracting the two measured values.

The annular venturi was calibrated against a venturi of known characteristics in the Hydraulics Laboratory. Thirteen sets of flow and pressure difference measurements at a temperature of 60°F (15°C) showed the pressure differential to be proportional to the 1.954 power of the flow rate over a range of 20 to 145 gpm. This exponent differs slightly from the theoretical 2.0 because of the effects of friction. Because of the desired accuracy of the flow split measurements, it will be necessary to perform further hot calibration experiments before the flow split tests begin.

Tritium Extraction from Ceramic Targets  
(Chemical Separations Unit)

The residues from nine thermal extraction runs were analyzed for tritium. The starting materials were either 79 or 90% of theoretical density and included both lithium aluminate and petalite. The retention of T by the residues after a 850°C thermal extraction varied from 0.01% after a one hr extraction to 0.005% after a 4 hr extraction.

Nuclear Safety Specifications  
(Critical Mass Physics Section)

Nuclear safety specifications covering the shipment of 2.1 wt% U<sup>235</sup> enriched uranium to the National Lead Company of Ohio in Model 44 shipping containers were reviewed for N-Fuels Engineering.

ALTERNATE URANIUM COMPOSITION - MISSIONS 1, 2, 4, 6, 10  
(Fuel Materials Development Unit)

An irradiation test was conducted to evaluate the performance of seventeen U alloys containing small additions of Zr, Mo, Nb, Al, Si, Fe, P and C, in terms of composition, heat treatment, fabrication history, corrosion behavior, and irradiation history. Volume measurements were completed on 79 of the fuel samples irradiated in this test. The swelling rates for the β heat treated reference fuel in this test were determined for U volume average temperatures ranging from 430 to 580°C. Composition of the reference fuel, designated as Alloy 1, is U + 305 ppm Fe + 150 ppm Al + 185 ppm Si + 762 ppm C. The R values, where

$$R = \frac{\% \Delta \text{Vol.}}{\text{at.}\% \text{ BU}}$$

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show an increase from 430°C to a maximum of ca. 9 at 455°C followed by a decrease to ca. 6 at 555°C. From 555 to 580°C the data indicate an increasing swelling rate.

Optical and electron metallography was completed on three irradiated samples of Alloy 1. The respective volume mean fuel temperatures and burnups of these samples were: 468°C, 521°C, 569°C, and 0.38, 0.33, 0.39 at.%. Optical metallography does not show any significant grain boundary tears on these three specimens. High magnification optical metallography and electron microscopy show some spherical and non-spherical porosity in the grain boundaries and in twin interfaces.

Metallography was completed on five specimens of the alloy which showed the smallest volume increases of all the alloys tested. The composition of this alloy is U + 1053 ppm Fe + 895 ppm Al + 150 ppm Si + 470 ppm C. The photomicrographs from these specimens are being collected and analyzed to determine what influence the high iron content of this alloy had on improving swelling resistance.

#### MISSION 16 - OTHER ISOTOPES

Code Development#  
(Theoretical Physics Unit)

#### HRG

One of the distinguishing features of the GAM calculational scheme for determining the energy spectrum of epithermal neutrons is the method of treating resonance contributions of the principal resonance absorbers. The resonance integrals of individual resonances are found and these are converted into effective fine group cross sections by dividing by a fine energy group flux. GAM, and HRG until recently, has used a 1/E flux for this conversion. This procedure has not correctly allowed for spatial and energy self-shielding effects. A revision in HRG, recently reported, has modified this  $\sigma_{\text{eff}}$  calculation by using, instead of the 1/E flux, an approximation to the flux used in the resonance integral calculation itself.

Although the modified  $\sigma_{\text{eff}}$  represent an improvement, the use in the flux integral of the Wigner rational approximation to the collision probability and the neglect of Doppler broadening has resulted in some uncertainty as to the accuracy of the improvement. A measure of this improve-

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ment and its accuracy has recently been obtained by comparing numerical integrations of the functional form of the flux with the analytical evaluation used in the HRG revision. The reference numerical integrations (B-E) used Doppler-broadened cross sections and "exact" collision probabilities, while the analytical evaluation (U-R) used unbroadened cross sections and the rational approximation. The 1.056 eV Pu<sup>240</sup> resonance, at 8 wt% Pu<sup>240</sup> in PuO<sub>2</sub>-UO<sub>2</sub> rods of a representative range of diameters in an hexagonal water lattice at 300°K was used in the comparison. The revised  $\sigma_{eff}$  for the fuel rod itself is 25 to 50% larger than the unrevised value: this value would be lessened by 2 to 4% if the (B-E).

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- b. The transport kernel modifications for the treatment of void regions were added to the standard treatment.
- c. The cosine current treatment of the transport kernel was added and modified to allow for the input of an energy dependent right-hand albedo.
- d. The above calculations of the transport kernels in both cylindrical and rectangular geometry were placed at the user's option.
- e. A smear option was added which gives punched card output of a smeared cell which can be used for region input to a later THERMOS case. The cross sections are averaged as,

$$\Sigma(v) = \frac{\int_{\text{cell}(v,r)} \phi(v,r) dv}{\int \phi_{\text{cell}}(v,r) dv}$$

- f. The anisotropy correction to the scattering kernel was placed at the user's option.
- g. The relaxation routine internal to the flux iteration calculation was modified to switch the calculation to a standard power iteration in case of numeric difficulties (as in the use of option f).
- h. The cell editing routine was rewritten and expanded to include the calculation of a current as,

$$\phi_1(v,r) = \frac{\nabla \phi_0(v,r)}{\Sigma_{tr}(v,r)}$$

The subsequent cell editing calculations are listed in the table.

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NEW CELL EDITING CALCULATIONS IN THERMOS

Microscopic Values Per Isotope

| <u>Quantity</u>    | <u>Edit</u>  | <u>Calculation Output</u>   |
|--------------------|--------------|---|
| Density            | Region       | $N(\text{Region})$  |
| Partial<br>Density | Region       | $\frac{\int_{\text{reg}} N(r) dV}{\int_{\text{cell}} dV}$   |
|                    | Region Smear | $\frac{\int_{\text{cell}} N(r) \phi_0(v,r) dv dV}{\int_{\text{cell}} \phi_0(v,r) dv dV}$  |
| Cross Sections (a) | Cell Smear   | $\frac{\int_{\text{cell}} N(r) dV}{\int_{\text{cell}} dV}$  |
|                    | Region       | $\frac{\int_{\text{reg}} \sigma(v) \phi_i(v,r) dv dV}{\int_{\text{reg}} \phi_i(v,r) dv dV}$   |
|                    | Region Smear | $\frac{\int_{\text{cell}} \Sigma(v,r) \phi_i(v,r) dv dV}{\int_{\text{cell}} N(r) \phi_i(v,r) dv dV}$  |
|                    | Cell Smear   | $\frac{\int_{\text{cell}} \Sigma(v,r) \phi_i(v,r) dv dV}{\int_{\text{cell}} \phi_i(v,r) dv dV \left( \frac{\int_{\text{cell}} N(r) dV}{\int_{\text{cell}} dV} \right)}$ |
|                    | All types    | $1/3 \sigma_{tr}$   |

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Macroscopic Cell Values

| <u>Quantity</u>                                     | <u>Edit</u> | <u>Calculation Output</u>   |
|---|-------------|---|
| Radius  | Region      | R (region)  |
| Volume Fraction                                     | Region      | $\int_{reg} dV / \int_{cell} dV$  |
| Flux ( $\phi_0$ )<br>and<br>Current ( $\phi_1$ )    | Depression  | $\frac{\int_{reg} \phi_1(v,r) dv dV}{\int_{reg} dV} / \frac{\int_{cell} \phi_1(v,r) dv dV}{\int_{cell} dV}$ |
| Cross Sections (a)<br>and<br>Inverse Velocities (b) |             | Region  |
|   | Cell Smear  | $\frac{\int_{cell} \Sigma(v,r) \phi_1(v,r) dv dV}{\int_{cell} \phi_1(v,r) dv dV}$                           |
| D   | All Types   | $1/3 \Sigma_{tr}$   |

(a) Absorption, scattering and fission cross sections are flux ( $\phi_0$ ) weighted and the transport and first moment scattering cross sections are current ( $\phi_1$ ) weighted.

(b) The inverse velocities are flux ( $\phi_0$ ) weighted values.  
(NOTE: In this case  $\Sigma(v,r) = 1/v$ ).

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BNW Master Library

The keV to MeV region of Fe<sup>56</sup> and C<sup>12</sup> was updated to obtain a better fit to the total cross section data by setting the potential scattering term equal to zero for the resonances between 10 keV and 10 MeV.

The thermal (2200 m/sec) value for Cu<sup>65</sup> absorption was corrected to 2.2 barns from 2.0 barns. The change yields 3.79 barns for natural copper at 2200 m/sec.

Because of a lack of space, resonance parameter tables are now being stored in the even numbered records and designated by parameter table number 511. Any table with a different value will still be found in Record 2. The ability to use bound state ( $E_0 < 0$ ) resonances has been added. These changes will allow the use of resonance parameter tables for many of the less important but still necessary isotopes.

Program UPDATE

The computer program UPDATE has been modified to include the resonance parameter table for a particular isotope in its even numbered record. This change was necessary in order for the BNW Master Library to contain a resonance parameter table for any isotope when required to fit the data. Previously, all resonance parameter tables were stored together in Record Number 2; however, almost all of the available storage space in Record 2 is being used, thus prompting the change in the system.

APDAC-III

A new option has been added to the 1107 version of the foil processing code, APDAC, which allows for the input of a foil half life for use in the decay correction calculation. For this particular option it is necessary to specify a material type of 13. The half life (in hours) is then input in Columns 41-50 of the foil data control card.

PROCESS TECHNOLOGY

Inspection of Inconel Tubes

(Water Reactor Corrosion & Chemistry Unit)

Two Inconel tubes were removed from N Reactor steam generator 4A to determine their condition after one year of operation. They were visually examined and appeared to be in excellent condition. The ID was covered with a shiny, dark olive-colored oxide. The OD (secondary side) had a shiny, olive-colored oxide covered in most locations with a

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dull black deposit, presumably iron oxide. One area of the OD had a gritty appearance but this proved to be deposited material only.

The tubes were decontaminated and submitted for nondestructive and metallographic examinations. The oxalic-citric decontaminating process readily cleaned the tubing.

Oxide weights on the ID and on shiny areas of the OD were 17 to 23 mg/dm<sup>2</sup>. The oxide weight at a region where a deposit existed on the OD was 62 mg/dm<sup>2</sup>. In comparison, oxide weights on 304 SS tubing removed from other steam generators have ranged from 19 to 148 mg/dm<sup>2</sup>.

Inspection of Process Tube Nozzle  
(Water Reactor Corrosion & Chemistry Unit)

The front and rear forged steel nozzles removed from an N Reactor process tube after 1 1/2 yr of operation were examined. Oxides present were almost all dull and gray-black in color. Numerous pits up to approximately 1/16 in. diameter were observed. Most of the larger pits were associated with wear marks by the feet of fuel element spacers. Detailed metallographic examinations of the pitted areas and analyses of the oxides were scheduled.

Target Element Development  
(Fabrication Metallurgy Section)

Assistance in pellet fabrication was continued for the Coproduct Demonstration Loading.

A portion of the green pellet production of GE-HAFD continued to be sintered by BNW which also provided technical guidance in the areas of pelletizing and powder preparation. Possible improvements in the processing parameters for each step of the pelletizing process are being studied and their effects on pellet density are being cataloged.

A study of abnormal powder behavior (RL-GEN-1084) indicated that a small variation in the manufacture of the lithium aluminate powder was responsible. Another study is underway to characterize the current inventory of powder.

Target Element Cladding Development  
(Fabrication Metallurgy Section)

To date, 7,348 ft of Zr-2 tubing has been vacuum annealed, reduced in diameter by approximately 1/2 in., stress relief annealed and stretch

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straightened for use as target element sheathing.

All of the tubes processed prior to July 19, 1966, were stress relief annealed, by resistance heating, at 600 to 620°C in air and then stretch straightened at room temperature. Subsequent autoclaving of target assemblies by NRD resulted in excessive bowing due to the relaxing of residual stresses and the variations in wall thickness of the Zr-2 tubing.

In an attempt to correct this bowing problem, the stress relief annealing was omitted but the next 24 tubes were resistance heated to 300°C (the autoclaving temperature) during the stretch straightening operation. These 24 tubes showed less tendency to bow but the condition was not sufficiently improved. The remaining 92 tubes are being heated to 610°C and straightened at this temperature. The tube is allowed to cool under tension. The few tubes autoclaved after this latest procedure have shown still less tendency to bow.

#### Pre-Load Enrichment Tester

(Nondestructive Testing Systems Engineering Unit)

All equipment for the 100-N fuel enrichment tester, which measures the gamma emission from  $U^{235}$  in the fuel core, was received and assembled. Development of the timing and gating circuits was started. Design of the scintillation probe shield and housing was completed and fabrication started. Late delivery of the equipment by the vendor will delay completion of this project by about 1 1/2 mo.

#### Radiography of Fuel Element Weld Closures

(Nondestructive Testing Systems Engineering Unit)

Assistance was provided in continuing the radiographic inspection of lithium-aluminum clad fuel elements to monitor the quality of the end cap closure weld.

#### Transducer Development

(Nondestructive Testing Systems Engineering Unit)

Effort continued to develop ultrasonic transducers with improved resolution and sensitivity for various fuels testing applications. A new supply of lithium sulfate elements was procured and a different technique for obtaining thinner (i.e., higher frequency) elements was investigated with very promising results. Since lithium sulfate is soluble in water, an attempt was made to water etch a crystal by submerging a 18 mil thick element in methyl alcohol with a water content of one part per thousand. The crystal was removed from the solution at intervals and its thickness measured. It was found that the alcohol-water solution removed one mil

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per hour with both sides exposed. It is expected that by depositing a thin layer of quartz, or a metal electrode, on one side of the crystal, and dissolving the opposite side only, thicknesses down to one mil can be obtained. It was observed microscopically that the surface finish followed the original crystal surface and that the crystal dissolved in small microscopically thin flakes. Crystal thicknesses of one mil will result in resonant frequencies of about 75 MHz which is more than double the high frequency response of commercially available elements. A patent disclosure report was prepared which describes both the quartz sealant and the water solute etching techniques for fabricating ultrasonic transducers.

PZT-5 ferroelectric material was successfully deposited on an Al electrode but proper polarization could not be accomplished. Refinements in the deposition technique are planned and this experiment will be repeated using both PZT-5 and two types of barium titanate.

Downstream Spacer & Fuel Element Vibration  
(Nondestructive Testing Systems Engineering Unit)

Vibration studies continued on the interference-fitted N-Reactor fuel assemblies; however, the program goal of developing a method for detecting potential fretting corrosion was enlarged to include evaluation of the support design as a result of recent flow tests at 189-D. It is necessary to determine the characteristics of the vibration to permit design modifications to control the problem.

Longitudinal vibration of a fuel element target under simulated service conditions (pressurized water flow at elevated temperature) in the N-Reactor test loop at 189-D was successfully measured using a commercial accelerometer. The accelerometer was mounted in a recess at one end of the target and covered with epoxy to protect it from the water. The lead wire was brought out of the tube through a swage-lock pressure fitting.

Initial analysis of the data indicated that the target was vibrating at its longitudinal resonance frequency (ca. 230 cps) with a peak to peak amplitude of about 0.0005 in. There was also some evidence that the target element was vibrating laterally but this could not be measured directly with the arrangement used. A subsequent test was planned wherein vibration along all three axes will be monitored. Other studies, using a different sensing technique, also have shown several low frequency resonances in the buggy-spring mountings. The resonant frequencies are somewhat dependent on fuel assembly orientation; however, they all lie between 100 and 300 Hz. The driver fuel has apparent beam resonances at

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480 and 1200 Hz. The flow test results have not shown any reasons for the different resonance characteristics found among the fuel assemblies.

Vibration Studies

(Systems Analysis and Simulation Unit)

Spectral analysis of some recorded fuel element vibration data was received from N Reactor personnel. Attempts to replay the data showed the information to be clipped. The degree of clipping on the recorded data was observed to be a function of the recorder electronics used. Calibration of the electronics must be accomplished before the data are analyzed.

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ASSISTANCE TO ISOCEM

SUMMARY

MISSION 1

In studies of the dissolution of Zircaloy, hydrochloric acid only was satisfactory for use as an acidic anodic dissolution medium. Anodic dissolution is also possible in alkaline systems involving hydroxide and carbonate.

Tributyl phosphite (TBP(III)) was shown to reduce Pu in a typical Purex extraction system when the system was exposed to ultraviolet radiation. Within the limit of error of the measurement, all the Pu was reduced to the trivalent state.

MISSION 7

Subcritical neutron interaction experiments were performed with bare and Plexiglas reflected arrays of bottles containing aqueous solutions of U<sup>233</sup>. The critical numbers of bottles in arrays were determined for various spacings. The experiments provide data for nuclear safety guidance in handling, storage, and shipment of this material, and represent the first data on criticality of interacting arrays of U<sup>233</sup> solutions.

Adequate stripping of uranium from DSBPP was obtained in miniature mixer runs testing a study flowsheet for solvent extraction removal of U<sup>232</sup> decay products from product U<sup>233</sup>.

MISSION 9

Experiments were completed which showed that essentially no carbon monoxide disproportionation occurred on exposure of the gas to bismuth oxide when a silica glass system is used. If on the other hand stainless steel was involved in the system, significant disproportionation occurred. It is apparent that the choice of materials of construction will be important if this process is adopted.

MISSION 10

British alloys, candidate high power density fuels, dissolve in nitric acid at about the same rate as ingot uranium.

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MISSION 15

Work was started to define the conditions which control the amount of Sr and rare earths carried by solids in the Purex acid wastes.

Preliminary solvent extraction studies have indicated that Am and Cm behave similarly in DBBP and D2EHPA-TBP systems. The possibility of isolating a pure cut is good, but partitioning by pH control does not seem to be a practicable prospect.

The procedure previously defined for increasing the loading of STT casks with Cs<sup>137</sup> was verified by Isochem in recent Cs shipments.

Laboratory experiments conducted to determine potential Cs losses from cocurrent ion exchange loading and elution operations indicated acceptable losses at double elution volumes, compared with countercurrent loading and elution requirements.

A commercial catalyst and Linde 13-X zeolite are being evaluated for use in the catalytic oxidation of TBP-NPH wastes.

PROCESS TECHNOLOGY

Conditions were defined for the recovery of Pu values from 234-5 Building sump wastes. Although some fluoride exists, the presence of Al should be avoided to achieve acceptable distribution coefficients with Dowex 50 resin.

The installation of equipment required to conduct a series of Zirflex process studies in the 324 Building has been completed and the calibration of the equipment is in progress.

Differences observed in Zircaloy dissolution and 304-L corrosion in Zirflex decladding solution prepared from (1) Redox plant chemicals and (2) C. P. chemicals were not confirmed in tests with another sample of Redox chemicals.

The ISOSHLID program is being extended to include a provision for treatment of Bremsstrahlung spectra in shielding and energy deposition problems. The necessary physical data have been cast in formats suitable to the computational logic presently in flow-chart form; some coding was completed.

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ASSISTANCE TO ISOHEM

MISSION 1 - BASIC PRODUCTION MISSION

Zircaloy Decladding Studies  
(Chemical Separations Unit)

Anodic dissolution of Zircaloy was attempted in all the commonly available acids and combinations of acids and salts. Only hydrochloric acid gave satisfactory dissolution and stable final solutions.

Fluoride catalyzed dissolution systems containing low concentrations of fluoride are not practical because, as dissolution proceeds, F ion activity is decreased by Zr complexing and catalysis is inhibited.

According to the literature, strong caustic converts Zr fluoride to the hydrous oxide which in turn is soluble in ammonium or potassium carbonate. Zircaloy can be anodically dissolved in a mixture of KOH,  $\text{NH}_4\text{F}$  and  $(\text{NH}_4)_2\text{CO}_3$ . The extent of dissolution relative to the F content remains to be investigated.

Tributyl phosphite (TBP(III)) has been shown to serve as a reductant for U(VI) under the influence of UV light in a simulated LEXF solution. The stoichiometry and kinetics of this reaction are now under investigation with the final goal of determining the process compatibility of TBP(III) as an alternate reductant in PUREX-type processes.

The intent of recent experiments has been to determine the behavior of the Pu distribution ratio in extraction systems containing TBP(III). The addition of TBP(III) to a typical Purex extraction system containing U and Pu followed by exposure to UV radiation resulted in a change of distribution ratio from 3.1 to 0.07, typical values respectively for Pu(IV) and Pu(III) under the conditions of the experiment. Similarly, low distribution ratios were also found when an aqueous phase containing Pu(IV) was extracted with standard Purex extractant to which had been added about 15% of the TBP(III)-containing extractant used in the above experiment.

Evaluation of the data indicated that within the error of measurement, all the Pu had been reduced to the trivalent state in these experiments.

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Non-Metallic Materials

(Materials and Process Chemistry Unit)

A pressure sensitive polyethylene tape on a stainless steel substrate was tested in the liquid phase and in the vapor phase above several typical separations processes solutions. No observable change occurred during ten days exposure to 50% NaOH. During 10 days exposure to 60% HNO<sub>3</sub> the tape separated from the substrate over about 10% of the surface in both liquid and vapor phase. The tape swelled rapidly and failed completely during 24 hr exposure to both liquid and vapor phase of Purex HAX, Recuplex CAX and hexone.

Pump Test and Evaluation

(Process Systems Development Unit)

A new pump test stand was completed and placed in operation in the engineering development laboratory. Three new pumps have been installed; performance and life tests were initiated. These include a Weinman liquid ring pump which is of interest because of its ability to handle entrained air or vapors; and two canned motor pumps being evaluated as replacements for the standard plant canned pumps. All units are undergoing an initial 500-hour test using water; nitric acid at a temperature of approximately 90°C will be pumped for the remainder of the tests.

MISSION 7 - URANIUM-233 PROGRAM

Criticality of Interacting Arrays of U<sup>235</sup> Solution Containers

(Critical Mass Physics Section)

The first data on criticality of interacting arrays of aqueous U<sup>233</sup> solutions were obtained. Subcritical neutron multiplication experiments were performed with bare and Plexiglas reflected arrays of bottles containing uranyl nitrate solutions, UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub>, at a concentration of ca. 330 g U<sup>233</sup>/l. The experiments provide data for nuclear safety guidance in handling, storage, and shipment of this material. The data were also needed for checking theoretical methods of predicting criticality of arrays.

The polyethylene bottles were 18 in. in height with an OD of 4.74 in.; the wall thickness was 0.10 in. The bottles were partially filled with solution, with the average depth being about 12 in., and the uranium content ca. 960 g U<sup>233</sup> per bottle.

The arrays were assembled on the Remote Split-Table Machine, utilizing the inverse neutron multiplication technique to estimate the critical

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number of bottles; the critical spacings for fixed number square arrays were also determined. In the reflected arrays the Plexiglas was touching the outside surface of the exterior bottles comprising the array. A low density Al honeycomb was used to maintain separation between bottles. In the non-reflected arrays, stability of the outer bottles was maintained by an Al frame magnetically mounted on a 0.05 in. steel base plate. Al honeycomb was used to support the baseplate and reduce neutron reflection.

In addition to the experiments on bare and reflected arrays, Lucite sheet was positioned between the bottles in some cases to determine the effect of internal moderation, such as might be brought about by partial flooding or by materials used in the construction of bird cages, etc.

The results of the experiments completed are summarized in the following table.

A single row of nine bottles was found to be subcritical when unreflected; extrapolation of the inverse multiplication data indicated subcriticality for an infinitely long line of bottles. The critical number for a single line of reflected bottles in contact was estimated to be between two and three. It is noted that the critical spacing with four bottles was increased from 2.18 to 2.66 in. on placing a one-inch thick Lucite plate between the bottles, which for this case represents the maximum effect of internal moderation.

DSBPP Solvent Extraction Purification of U<sup>233</sup>  
(Materials and Process Chemistry Unit)

Miniature mixer settler runs testing the extraction-scrub column of a study flowsheet for removing U<sup>232</sup> decay daughters from U<sup>233</sup> product using di-sec-butyl phenylphosphorate (DSBPP) as solvent gave good U recovery and decontamination from T, P, and Pu. The study flowsheet strip column, with twelve mini-stages and a strip solution (1CX, 0.01M HNO<sub>3</sub>) volume equal to the organic feed (LAP) volume, gave U losses to the strip column waste (LCW) less than 0.1%. At a 1CX/LAP flow ratio of 0.75, U loss to the LCW was 0.2%. Difficulty was experienced in obtaining complete U stripping when as received DSBPP was used in preparing the LAP but this problem was eliminated by washing the DSBPP with 3% Na<sub>2</sub>CO<sub>3</sub>, 1M HNO<sub>3</sub> and water prior to LAP preparation.

Agitated Trough Calcination Studies  
(Engineering Development Unit)

Three denitration runs (3M Th(NO<sub>3</sub>)<sub>4</sub> feed solution) were made in the 4 in. diam agitated trough calciner. The calciner was also operated with water feed and at agitator speeds of 60, 85, and 120 rpm to obtain

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INTERACTION DATA ON CRITICALITY OF BOTTLES OF U<sup>233</sup> SOLUTION IN ARRAYS

| <u>Description</u>                   | <u>Config-<br/>uration</u> | <u>Critical Number</u>        | <u>Surface-to-Surface<br/>Spacing,<br/>Inches</u> |  |
|--------------------------------------|----------------------------|-------------------------------|---|--|
| Bare Single Row                      | 1 x 9                      | Subcritical                   | 0   |  |
| Bare Dbl. Row                        | 2 x 3                      | 6.1                           | 0   |  |
| Bare Lattice                         | 3 x 3                      | 9                             | 0.60  |  |
| Bare Lattice                         | 4 x 4                      | 16                            | 1.16  |  |
| Refl. Single Row                     | 1 x 2<br>1 x 3             | Subcritical<br>Supercritical* | 0<br>0  |  |
| Refl. Single Row                     | 1 x 3                      | 3                             | 0.60  |  |
| Refl. Lattice                        | 2 x 2                      | 4                             | 2.18  |  |
| Refl. Lattice/ $\frac{1}{2}$ " Mod.  | 2 x 2                      | 4                             | 2.48  | $\frac{1}{2}$ " Lucite Mod.<br>Between Bottles |
| Refl. Lattice/ $\frac{3}{4}$ " Mod.  | 2 x 2                      | 4                             | 2.58  | $\frac{3}{4}$ " " "                            |
| Refl. Lattice/1" Mod.                | 2 x 2                      | 4                             | 2.66  | 1" " "   |
| Refl. Lattice/ $1\frac{1}{2}$ " Mod. | 2 x 2                      | 4                             | 2.50  | $1\frac{1}{2}$ " " "                           |
| Refl. Lattice                        | 3 x 3                      | 9                             | 3.98  |  |
| Bare/1" Lucite                       | 2 x 3                      | 6.3                           | 1.00  | 1" Lucite Mod.<br>Between Bottles              |
| Bare/1" Lucite                       | 3 x 3                      | 9                             | 1.60  | " " "  |

\* From the neutron multiplication measurements it is apparent that 3 bottles would have been supercritical if assembled in contact.

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supplementary heat transfer data. An analysis of all the operating data was in progress.

MISSION 9 - POLONIUM-210

Bi/Bi<sub>2</sub>O<sub>3</sub> Catalysis of CO Disproportionation  
(Heavy Element Chemistry Unit)

The possibility of the disproportionation of carbon monoxide during the CO reduction of Bi<sub>2</sub>O<sub>3</sub> was investigated, since it is known that other metal oxides or their reduction products cause such disproportionation.

These experiments involved the passing of CO over known amounts of Bi<sub>2</sub>O<sub>3</sub> at temperatures ranging between 500 and 700°C. The gas flow rate was measured before and after passing over the Bi<sub>2</sub>O<sub>3</sub>. Subsequently the

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dissolution rates of these alloys unirradiated were comparable to or slightly lower than rates for unirradiated ingot uranium in 2-10 M  $\text{HNO}_3$ . Easily centrifuged white solids (probably silica) precipitated when the silicon-containing alloy was dissolved in 10.4M  $\text{HNO}_3$  to yield a 2.6M  $\text{U}(\text{NO}_3)_2$  solution. A Purex process feed solution (LAF) prepared from this solution (solids removed) showed normal disengaging behavior when contacted with 30% TBP-NPH solvent.

#### MISSION 13 - NUCLEAR SAFETY

##### Plutonium Aerosol Studies

(Particulate and Gaseous Waste Research Unit)

Plutonium oxide particles entrained by air passing over a 1770 g ingot of "as-cast" alpha Pu during oxidation above ignition temperatures were examined by electron microscopy and a size distribution was determined. The particles covered a range of sizes from submicron to as large as 18 $\mu$ ; the frequency distribution curve did not fit a linear plot on log-normal probability paper. The count median diameter was 1.2 $\mu$  with a MMD of 4.2 $\mu$ . The air velocity selected for sweeping the specimens during oxidation was relatively high; hence, larger particles were entrained. Very few, if any, particles were larger than inhalable particles.

##### Criticality Safety Orientation

(Critical Mass Physics Section)

A Criticality Safety Orientation course was presented to twenty new technical employees. The course consisted of five 1½ hr. lectures on the subject of nuclear safety and criticality control practices.

##### Computer Code Modifications

(Critical Mass Physics Section)

To facilitate nuclear safety and criticality calculations, the GAMEEC-II Code was modified to punch both macroscopic and microscopic cross section data for the DIF-IV Transport Theory Code. The code previously punched macroscopic data only. The DIF-IV Code was also modified to utilize correctly the  $\text{DB}^2$  leakage approximation for anisotropic scattering.

#### MISSION 15 - WASTE MANAGEMENT

##### Purex Acid Waste Studies

(Chemical Research Section)

Work has started which will define the conditions controlling the

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amount of Sr and rare earths carrying on the solids in Purex acid wastes. The effect of  $H^+$  and  $SO_4^{=}$  was studied on synthetic PAW and the following observations made.

- a) In the absence of  $SiO_2$ , Zr, Mo, Sn, and Ba a solid is formed on heating when the  $SO_4^{=}$  exceeds 0.5M and  $H^+$  is less than 1.0M.
- b) Little or no Ce or Sr is carried on the solid unless the  $SO_4^{=}$  is 1.0M or higher and the  $H^+$  is less than 0.6M.
- c) The amount of Ce and Sr carrying increases with the length of time of heating.
- d) The solid formed is quite insoluble in 1.0M acid.

The effect of the absent components will be studied on an individual basis. Experiments will also be undertaken using a 40 gallon sample of current plant PAW.

#### Trans-Plutonium Solvent Extraction (Chemical Research Section)

Work has begun to expand our knowledge of Cm behavior in the DBBP- $CCl_4$  and D2EHPA-TBP-NPH solvent extraction systems. The information sought includes relative extractability of Am and Cm, effect of complexants on Am and Cm distribution ratios, possible separation of Am and Cm in these systems, possible oxidation of Am by peroxysulfate and stripping characteristics of Am, Cm, Eu and Ce from the DBBP system.

Preliminary results show similar distribution ratios for Am and Cm in both of the DBBP and D2EHPA-TBP systems tried, thus providing conditions for recovering a clean mixture of the two. Experiments to date indicate that partition by pH control is not promising.

#### Cesium-Decalco Studies (Chemical Research Section)

Two successful loadings of STT casks with 60,000 Ci of  $Cs^{137}$  were made by Isochem, thus verifying the laboratory developed procedure. Current effort is aimed at reducing the dilution water from 100% of feed to 10%, the minimum believed necessary to prevent precipitation in the loading step. This should be possible if external cooling is used to control temperature (rather than water dilution). A test will be conducted using an available PNL cooler which is part of the High Level Radiochemistry Facility auxiliary equipment.

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In-Tank Solidification  
(Process Systems Development Unit)

Review of the program for in-tank solidification of stored wastes resulted in a decision to perform additional pilot studies using simulated wastes. The basic purpose will be to confirm results of earlier tests using new information on actual waste composition and viscosities. Pilot scale tests will be directed toward firming up circulation rates, solidification characteristics, and hot waste transfer feasibility.

In the Statistics Section a study was continued to formulate a model for assessing the cumulative error in the continuing waste tank inventory of radioactive isotopes.

Alkaline Waste Treatment  
(Water and Wastewater Research Unit)

Laboratory experiments were conducted to determine potential Cs waste losses when it is loaded on and eluted from a column of AW 500 zeolite in the same direction of flow. Downflow loading and elution may be necessary in the B Plant Cs recovery process if zeolite bed expansion is a problem with upflow elution. Fifty column volumes of simulated Purex alkaline supernatant solution were pumped downflow at 2 column volumes per hour through a column of AW 500. Cs breakthrough was 4% at the end of the loading cycle and the total waste loss was less than 1%. The column was eluted downflow with 8 column volumes of  $3M (NH_4)_2CO_3 + 2M NH_4OH$  at 55°C (specified in current flowsheet). The column was washed with 3 column volumes of water and loaded downflow on the second loading cycle to 50 column volumes at 2 column volumes per hour. Initial Cs breakthrough was 82% which decreased to 10% at the end of the cycle. The total waste loss was 21%. As expected, the results clearly show that the downflow elution volume must be increased to reduce waste losses to a tolerable level. A third elution cycle was made under the same conditions as the second except that the elution volume was doubled to 16 column volumes. Waste losses were then reduced to less than 1% during subsequent loading. The work will continue to more closely define the amount of elutant needed for this method of operation.

Catalytic Oxidation of Organic Wastes  
(Water and Waste Water Research Unit)

A catalyst supplied by W. R. Grace Company and Linde 13-X zeolite are being evaluated for use in the catalytic oxidation of organic wastes.

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Grace catalyst #908 at a temperature of 150°C starts to react with 30-70% TBP in NPH in two minutes. Reaction temperatures are normally kept at about 600°C, by controlling the air fuel ratio, but temperatures of 1000°C are obtainable. In a 6 hr run 190 ml of fuel was consumed on a 10g catalyst bed (2-0.5 mm diam particles). About 7% of the fuel remained as a black residue in the fuel vaporizer.

Ce or Na-based Linde 13-X initiates oxidation of the same fuel at 100°C and otherwise behaves similarly to the Grace catalyst.

#### PROCESS TECHNOLOGY

##### C-Cell Equipment Installation (Chemical Separations Unit)

The equipment installation in C-Cell for reprocessing irradiated Np was slowed by the lack of craft maintenance personnel. The column end plates and heat exchangers are to be machined off-site.

##### Neptunium Chemistry (Chemical Separations Unit)

Efforts to learn more about the nature of the material formed by dissolution of U (containing carbide impurity) which accelerates the oxidation of neptunyl(V) to (VI) have continued. Neither of the soluble organic materials (oxalic acid and mellitic acid), which have been identified at CRNL as products of the reaction of U carbides with nitric acid, was found to accelerate the oxidation of neptunyl(V). The rate accelerating material must be one of the reaction products mentioned but not identified by CRNL personnel.

A small "downdraft" dissolver will be used in studying the effect of U dissolution conditions on the oxidation of neptunyl(V) in the resulting dissolver solutions.

Hydrazine may be a suitable alternate to Fe(II) as a reductant for Np in high nitrate systems. Initial studies indicate that the mechanism of the reduction is quite complex.

##### Recovery of Plutonium from 234-5 Building Sump Water (Chemical Separations Unit)

The work on Pu recovery from 234-5 Building sump water was completed. The very satisfactory cation resin loading results obtained with synthetic plant sump water with Pu(III) in the absence of Al were confirmed using Pu<sup>238</sup> traced plant sump solutions. Six different batches of plant sump

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solutions were examined and distribution coefficients,  $D_v$ , on Dowex 50W X-8 of  $8.5 \times 10^3$  to  $2.7 \times 10^5$  were obtained using  $0.05M$   $NaHSO_3$  as reductant. These solutions ranged in fluoride concentration from  $<0.0005M$  to  $0.063M$   $F^-$ .

Corrosion by Zirflex Decladding Solution  
(Materials and Process Chemistry Unit)

The corrosion resistance of a sample of 304-L Stainless Steel containing one wt percent boron was determined in Zirflex dissolvent under initial, dissolving and terminal conditions. The overall corrosion rate was about fourfold higher than that for 304-L without boron. It was reported last month that Zircaloy dissolution rate and the amount of zirconium which would dissolve in a given volume of dissolvent were significantly lower for dissolvent prepared from Redox plant chemicals than for dissolvent prepared from reagent grade chemicals. Also that 304-L corrosion rates in the terminal decladding solution were higher when Redox plant chemicals were used. Since then, another sample from Redox plant has been evaluated with no differences in zirconium dissolution or 304-L corrosion between this dissolvent and dissolvent prepared from C. P. chemicals. No explanation has been found as yet for the anomalous behavior of the first sample of Redox chemicals.

Zirflex Process Studies  
(Engineering Development Unit)

The installation of equipment required to conduct a series of Zirflex process studies in the 324 Building has been completed and the calibration of the equipment is in progress. Several sections of 60 mil Zircaloy-2 tubing (0.625 inch OD and 1.75 inch OD sizes) have been autoclaved for 30 days at  $400^\circ C$ . These tubes will be used in the initial dissolutions to study the effect of oxide coating on the dissolution rate.

Modification of ISOSHLID to Incorporate Bremsstrahlung Calculations  
(Engineering Analysis Section)

Progress to date includes complete flow-charting of the ISOSHLID program logic; formulation of major aspects of the program modification; assembly of pertinent tables including beta and point energies, Bremsstrahlung source spectrum data, and (X-ray) mass absorption coefficients; coding of data handling routines for the source spectrum library; and organization of data-table interpolation logic. In addition, most major decisions have been made regarding what approximate numerical forms to use in computing within the limitations of the general multigroup methods to be employed here.

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Separations - Instrumentation  
(Process Instrumentation Unit)

Fabrication progressed on several portions of the developmental, compact, solid state gamma spectrometer instrument, which was developed for measurement of plutonium concentrations in hoods and glove boxes at the 234-5 Building. The spectrometer design incorporates refined electronic circuitry to assure maximum reliability and versatility.

The completed developmental Am<sup>241</sup> Breakthrough Monitor, to be used for the determination of abnormal concentrations of Am<sup>241</sup>, is in the field awaiting installation by Isochem technicians.

Wall Thickness Measurements of Waste Transfer Lines  
(NDT Customer Applications Unit)

Nondestructive Testing provided technical assistance to Isochem in determining the wall thickness of the 154-B diversion box radioactive waste transfer lines located in the 200 East Area. A ten-foot pole extension was attached to the ultrasonic probe, because of the radiation field, to permit the operator to test from the top of the trench. The test showed no evidence of gross corrosion. The five, 3-inch diameter stainless steel lines were found to have 0.119 inch maximum to 0.113 inch minimum wall thicknesses.

Soil Chemistry  
(Water and Wastewater Research Unit)

Characterization of soil-sorbed Pu beneath the 216-X-9 crib continued. Previous studies showed that groundwater would leach 0.2% of the Pu held on the soil. Current research, in which  $K_d$ 's were determined for soil and a Pu solution obtained from contaminated soil leachates, gave values greater than  $10^4$  ml/g. This high  $K_d$  indicates that the average movement of the 0.2% leached will be less than  $2 \times 10^{-3}$  of the transporting solution. A Pu soil diffusion coefficient also was determined ( $3 \times 10^{-12}$  cm<sup>2</sup>/sec) and indicates that movement by a diffusion mechanism (Pu concentration gradient) will be negligible during a 10 half-life period.

BEN FRANKLIN DAM

Waste Inventory of Ground  
(Geochemical and Geophysical Research Unit)

A waste crib exploratory well drilled at the 216-X-9 Redox process

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condensate crib encountered a 13 ft. thick saturated zone at a depth of 133 ft. (about 70 ft. above the regional water table). The zone is in sandy silt which overlies about 4 ft. of clay and caliche. A 5 gal. sample was obtained from the perched water and submitted for laboratory analysis. The liquid is essentially pure, decontaminated waste, and the analytical results are of particular interest for comparison with laboratory soil column results. A sharp decrease in activity level was noted in samples taken from below the clay and caliche layer.

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ASSISTANCE TO VITRO ENGINEERING COMPANY

(Mathematical Analysis Section)

Consultation services were provided on the hydrodynamical theory pertaining to the design of an elbow in a hydraulic line.

ASSISTANCE TO HANFORD OCCUPATIONAL HEALTH FOUNDATION

(Mathematics Department)

Work continued on a study to formulate a computer program to score and interpret batteries of personnel tests.

An additional series of correlation analyses was carried out on extensive personnel data from the Reliability Research Program on Safety.

Reactor Operator Certification data were summarized and several correlations were investigated.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

METEOROLOGICAL SERVICES  
(Atmospheric Sciences Section)

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

JULY 1966  
Forecasting

| <u>Type</u> | <u>No. Made</u> | <u>% Reliability</u> |
|-------------|-----------------|----------------------|
| Production  | 93              | 85.3                 |
| General     | 62              | 84.8                 |
| Special     | 100             | 90.8                 |

Number of calls processed by Code-A-Phone: 4,252

ENVIRONMENTAL EVALUATION  
(Environmental Studies Section)

Shutdown of Hanford production operations during the strike is expected to permit radiation levels in the environs, especially the river, to reach previously unobserved low levels. Direct measurements showed that gross gamma activity in river water at the 300 Area decreased to a nominal background level within 24 hr of the reactor shutdown. However, special river water analyses showed that Zn<sup>65</sup> transport at the 300 Area continued at one-fourth to one-third the normal rate, presumably associated with particulate material still being flushed downstream by high river flow. No other gamma-emitters were identified.

Continued radioiodine emissions from the Redox stack until mid-July and the normal lag in analytical results have prevented any summary to date of airborne activity reduction.

Special measurements have also been made of the temperature of the river while no heat is being added by the reactors. One set of traverses was made at each of seven stations from above the reactors to Richland, while another set was made at intervals during the day at the 300 Area and Richland. The increase in mean river temperature during the day was slightly more than 1°C in both cases, or about

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two-thirds of that shown by the temperature recorders established at the 300 Area and the Richland pumping plant. The temperature of the river as a whole is rising (typical of the summer months) and this rise averaged about 0.15°C per day during the last half of the month.

#### DOSIMETRY STUDIES

(Dosimetry Technology Section)

The Maushart lung counter was received and will be placed in operation as soon as the assembly and operating manuals are received. This device is expected to prove useful in the direct measurement of lung burdens of radionuclides, principally Pu in workers.

Installation of K-free phototubes on the 9 in. x 14 in. NaI crystal detector in the iron room was completed. Initial study indicated a reduction in K of 70% which should permit a significant improvement in detection capabilities.

The theoretical maximum Pu deposition obtained if a worker annually excreted Pu in urine at just less than the detection level for 50 yr of exposure was calculated. This and other calculations are a part of a development of a more rational basis for the Pu surveillance programs.

Study of the ICRP task force on Lung Dynamics continued. Recommendations on the practical application of the model to the evaluation of Hanford exposure cases are being developed.

The study was continued of U in workers as to the adequacy of present surveillance and evaluation practices.

Strontium-85 and creatinine urinary excretion data on two additional hospital patients were received from Swedish Hospital.

#### PERSONNEL DOSIMETRY

(Personnel Dosimetry Section)

There were 260 bioassay samples submitted for Pu analysis during the month including 9 from new employees who had either previously worked with Pu or who were assigned to work with it in their assignment at Hanford. No new deposition cases were confirmed by evaluation of bioassay data. Also, 295 samples were submitted for U analysis, 44 for T analysis, 67 for Sr analysis and 7 for analysis for other radioisotopes.

There were 170 whole body counter and thyroid examinations conducted, including 3 examinations requested as a result of radiation

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occurrences, 36 examinations of new hires, and 70 terminees. Based on evaluation of the results of these examinations one employee received a thyroid burden of 0.05  $\mu\text{Ci}$   $\text{I}^{125}$ . This is approximately 5% of the maximum permissible body burden of 1.1  $\mu\text{Ci}$ , thyroid as reference. Evaluation of the results of the rest of the examination does not indicate any internal deposition in excess of 1% of the applicable permissible limits.

Special external exposure evaluations were performed for 22 Hanford contractor employees; however, no exposures over permissible limits were revealed.

Special occupational exposure summaries were prepared in response to requests from new employers of 18 former Hanford employees under provision of the Atomic Energy Commission regulations 10 CFR 20 and AEC Manual Chapter 0502.

Records processing of 8504 employee beta-gamma film badge dosimeter results, 245 neutron results, 267 finger ring dosimeter results, and 378 visitor film dosimeter results was conducted. None of the results exceeded limits established in AEC Manual Chapter 0502.

AQUATIC BIOLOGY STUDIES  
(Biology Department)

Two races of chinook salmon (Puget Sound and local Columbia River), reared for four months in various concentrations of reactor effluent, were assayed for radionuclide content with results as summarized in the following table. The radioactivity observed is primarily due to direct uptake from water as the fish were reared on a nonradioactive diet. The direct relationship between concentration of the effluent and body burden is evident. The higher activity observed in the local race is most likely due to age difference rather than to racial characteristics, since the Puget Sound fish were about a month older at the start of the experiment.

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| Effluent Level, % | Race of Fish | Avg. wt., g | Average pCi/g fish |                 |                  |                  |                 |
|-------------------|--------------|-------------|--------------------|-----------------|------------------|------------------|-----------------|
|                   |              |             | Na <sup>24</sup>   | P <sup>32</sup> | Cr <sup>51</sup> | Zn <sup>65</sup> | K <sup>40</sup> |
| 0                 | Puget Sound  | 2.8         | 73                 | 44              | 26               | 8                | 4               |
| 0                 | Local        | 0.63        | 77                 | 32              | 19               | 7                | 3               |
| 2                 | Puget Sound  | 3.2         | 720                | 32              | 43               | 15               | 4               |
| 2                 | Local        | 0.77        | 750                | 58              | 38               | 20               | 5               |
| 4                 | Puget Sound  | 3.7         | 1400               | 50              | 48               | 19               | 5               |
| 4                 | Local        | 1.0         | 1400               | 69              | 53               | 36               | 7               |
| 6                 | Puget Sound  | 3.8         | 2000               | 49              | 77               | 33               | 6               |
| 6                 | Local        | 1.1         | 2200               | 100             | 65               | 45               | 9               |

Studies continued on artificially induced immunity of trout to columnaris organisms. As maximal titers are developed in the experimental animals, we shall determine the resistance to infection conferred by this immunity. A summer consultant is conducting studies on the effect of temperature on immune clearance of soluble and particulate materials from the circulating blood stream of fishes. These studies may contribute importantly to our overall interpretation of the seasonal fluctuations in incidence of columnaris disease in the Columbia River.

A total of 1,504 fledgling gulls (705 ring-billed and 799 California) were banded during two days operations at Ringold and Coyote Rapid colonies. The crowded conditions caused the birds to rest in "shifts" and a considerable number to rereest. The establishment of "satellite" colonies observed in 1965 was much reduced this year. The effect of population density on the nesting behavior of gulls is little understood; the gull populations at Hanford offer interesting possibilities for such behavioral studies.

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INSTRUMENTATION SERVICES

Laundry Monitoring System  
(Process Instrumentation Unit)

Initial testing of the contaminated clothing reject mechanism control chassis indicated that correct circuitry performance was being achieved. The background radiation monitoring probe, from which background subtraction information will be obtained for use in enhancing the system sensitivity, provided a gamma counting efficiency of about 15% during a series of tests; this is fully satisfactory for the intended application.

Assembly of the mechanical system moved forward with fabrication of the probe support hangers and with completion of all required rollers, shafts, and shaft bearings for the upper conveyor belt. General assembly of the system progressed rapidly.

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