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# DECLASSIFIED HANFORD LABORATORIES MONTHLY ACTIVITIES REPORT

MARCH, 1963

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HANFORD LABORATORIES  
MONTHLY ACTIVITIES REPORT  
MARCH 1963

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By Authority of CG-PR2  
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Compiled by  
Section Managers

April 15, 1963

HANFORD ATOMIC PRODUCTS OPERATION  
RICHLAND, WASHINGTON

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PRELIMINARY REPORT

This report was prepared only for use within General Electric Company in the course of work under Atomic Energy Commission Contract AT(45-1)-1350. Any views or opinions expressed in the report are those of the author only.

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TABLE OF CONTENTS

	Page
Force Report and Personnel Status Changes . . . . .	iv
General Summary	
Manager, H. M. Parker . . . . .	v through xxx
Reactor and Fuels Laboratory	
Manager, F. W. Albaugh . . . . .	A-1 through A-70
Physics and Instruments Laboratory	
Acting Manager, R. S. Paul . . . . .	B-1 through B-33
Chemical Laboratory	
Manager, W. H. Reas . . . . .	C-1 through C-23
Biology Laboratory	
Manager, H. A. Kornberg . . . . .	D-1 through D-11
Applied Mathematics Operation	
Manager, C. A. Bennett . . . . .	E-1 through E-7
Programming Operation	
Manager, W. K. Woods . . . . .	F-1 through F-15
Radiation Protection Operation	
Manager, A. R. Keene . . . . .	G-1 through G-9
Finance and Administration Operation	
Manager, W. Sale . . . . .	H-1 through H-11
Test Reactor and Auxiliaries Operation	
Manager, W. D. Richmond . . . . .	I-1 through I-6
Invention Report . . . . .	J-1

TABLE I - HANFORD LABORATORIES FORCE REPORT

DATE: March 31, 1963

	At Beginning of Month		At Close of Month		Total
	Exempt	Salaried	Exempt	Salaried	
Chemical Laboratory	132	131	133	132	265
Reactor & Fuels Laboratory	186	171	186	177	363
Physics & Instruments Laboratory	96	68	101	69	170
Biology Laboratory	39	61	40	61	101
Applied Mathematics Operation	18	4	18	4	22
Radiation Protection Operation	44	88	44	88	132
Finance & Administration Operation	113	113	108	112	220
Programming Operation	11	3	11	3	14
General	3	4	3	4	7
Test Reactor & Auxiliaries Oper.	56	298	55	298	353
TOTAL	698	941	699	948	1,647

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14

HW-77046

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## BUDGET AND COST SUMMARY

March operating costs totaled \$2,480,000, a decrease of \$200,000 from the previous month; fiscal year-to-date costs are \$21,156,000, or 69% of the \$30,857,000 control budget. The control budget includes an increase of \$525,000 received in a recent RLOO-AEC financial plan. However, a reduction of \$235,000 in the Gas-Cooled Power Reactor Program was not made because the AEC has indicated intent to restore a major portion of these funds. Hanford Laboratories' research and development costs for March compared with last month and the control budget are shown below:

(Dollars in thousands)	COST				
	Current Month	Previous Month	FY To Date	Budget	% Spent
HL Programs					
02 Program	\$ 70	\$ 83	\$ 659	\$ 1 069	62
03 Program	24	27	91	175	52
04 Program	1 026	1 155	8 755	12 718	69
05 Program	132	124	936	1 353	69
06 Program	241	275	2 313	3 154	73
08 Program	16	11	36	97	37
	1 509	1 675	12 790	18 566	69
NRD Sponsored	180	172	717	1 270	56
IPD Sponsored	59	73	696	932	75
CPD Sponsored	111	118	1 071	1 421	75
FPD Sponsored	0		493	493	100
Total	\$1 859	\$2 038	\$15 767	\$22 682	70%

## RESEARCH AND DEVELOPMENT

### 1. Reactor and Fuels

Radiometallurgical examination of prototype N-Reactor fuel elements irradiated to an average exposure of 1250 Mwd/ton continues to reveal no detrimental features.

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Two fluted single tube N-Reactor fuel elements successfully completed a second cycle of irradiation in the ETR. Exposure on these elements is now 400 Mwd/ton. Two irradiation tests of fluted N-inner components continued with the exposure in these two tests now being 1300 Mwd/ton.

Fabrication of two lots of "self brazed" closure N-inner fuel components has been completed. These elements are being evaluated for irradiation test purposes.

Analysis of N-Reactor fuel closures indicates a factor of three to four reduction in stress level is possible by proper shaping of the closure.

Tests simulating the wear of N-fuel supports on process tubes continued to show a minimal wear after four series of tests. Other charging tests of N-Reactor fuel elements were performed to evaluate the effect of the magazine-to-nozzle transition and the nozzle-to-pressure tube roll joint on the suitcase handle supports. Gouging noted on the shoes of two charges was attributed to the soft iron shims used between the supports and support shoes to provide the proper support height.

A program was initiated this month to study the irradiation performance of tritium target materials under N-Reactor conditions. Two series of double clad target rods are being fabricated with a 0.91 and 2.10 wt % lithium content. Billets have been prepared, fabricated, and sized into Al cans. Aluminum end caps must now be welded in place and the rods assembled into a second can of Zircaloy-4.

Phosphorus in quantities up to 400-500 ppm has been introduced into the enriched fuel stream for the N-fuel and for the present reactor I&E fuel as a result of scrap recovery process changes at NLO. Evaluation to date indicates that phosphorus forms a eutectic between UP and U. This behavior is quite unlike that of other nonmetallic impurities such as C, O<sub>2</sub>, and N<sub>2</sub>. Careful evaluation of effects of phosphorus throughout the fabrication process will be required to establish a meaningful specification.

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vii

HW-77046

Evaluation of  $\text{NH}_4\text{OH}$  as a substitute for  $\text{LiOH}$  to adjust pH in N-Reactor was delayed by operating difficulties of KER-1. While attempting to establish a satisfactory initial base operation before proceeding with the change from  $\text{LiOH}$  to  $\text{NH}_4\text{OH}$ , unexplained high crud levels were observed.

Corrosion penetration of aluminum coupon samples was less in quachrom glucosate inhibited water than in dichromate inhibited process water based on tests in the single pass KE tubes.

The distribution of radioisotopes deposited on fuel elements in a typical KE tube was determined for the first time. The activity distribution corresponded to the corrosion intensity.

A procedure to monitor sodium concentration and thus detect leakage of raw water into the N-Reactor secondary system has been evaluated and proved satisfactory.

Zircaloy-2 coupons with thick oxide films were found to hydride in 1 month in a  $\text{He-H}_2(3\%)-\text{CO}(2\%)-\text{H}_2\text{O}(0.4 \text{ mm})$  gas mixture at both 375 and 425 C, whereas thinly filmed samples did not hydride. This result indicates  $\text{H}_2\text{O}$  inhibition of gas phase hydriding of Zircaloy-2 process tubes may fail if extensive corrosion produces a thick film.

In studying the nickel plating of uranium, it has been discovered that gas pitting of uranium during the preplating electrolytic etch is reduced by ultrasonic agitation of the piece at 30 kc.

Pits, penetrating one-third of the pipe wall, have been observed radiographically in static sections of PRTR softened water and PRTR process water systems. No pitting is observed in sections of these systems where flow is maintained.

Intital measurements of the kinetics of the water gas shift reaction  $\text{CO} + \text{H}_2\text{O} \rightleftharpoons \text{CO}_2 + \text{H}_2$  indicate little reaction at N-Reactor temperatures.

An expression describing the burnout profile in D-Reactor was derived from rate expressions, temperature dependences, and reactor temperature profile.

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EGCR graphite samples have been irradiated to  $1.4 \times 10^{22}$  nvt ( $E > 0.18$  Mev). Examination is not yet complete.

Graphite samples compression-loaded to 300 psi were found to have contracted to a greater extent than unstressed samples; however, the change was less than in the first irradiation period.

The gamma induced water-graphite reaction rate was found to be proportional to dose rate.

The effect of vacuum degassing in lowering the initial rate of oxidation of graphite by  $\text{CO}_2$  was explained on the basis of removal of  $\text{CO}_2$  from the active sites and slow re-establishing of the steady state condition. Rate expressions applicable to the nonsteady state period were derived.

Calculation of the space-energy distribution of neutrons in the ETR disclosed steep flux gradients over irradiation positions that could lead to serious errors in interpretation if monitors are improperly used.

Flow characteristics were determined for aluminum support pieces to be placed in empty tubes at K-Reactor to decrease the flow rate and thus prevent cavitation in the inlet fittings.

The boiling burnout data from a 0.050-inch spaced 19-rod bundle which had small "warts" to maintain spacing between rods were compared with previous data obtained with a test section using wire wraps on 12 of the rods to maintain spacing. It was found that the burnout heat fluxes coincided very closely and indicated no effect of the wire wraps on boiling burnout.

Approximately 1990 two-phase, pressure-drop data points were obtained for typical reactor piping and fittings at flow rates between 500,000 and 4,000,000 lb/hr-ft<sup>2</sup> and at coolant conditions between all liquid and 25% steam by weight.

The General Electric Technological Hazards Council has endorsed increase of the PRTR heat transfer flux limit from 400,000 to 650,000 Btu/(hr)(ft<sup>2</sup>), and of the tube power limit from 1200 to 1800 kw (AEC approval

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required). The Council concluded that boiling convection cooling in PRTR is adequate in event of a total power failure provided the pressurizer relief valves open properly and correct operator action is taken at approximately the right times.

AEC approval has been received to operate the PRTR Fuel Element Rupture Testing Facility.

Design and fabrication of the second generation mechanical shim rod for PRTR continued with fabrication of the drive head approximately 70% complete. A first generation shim rod will be tested in a shim rod environmental test facility recently completed.

Experiments were continued to develop a procedure for monitoring zirconium concentrations in the PRTR coolant as an indication of fretting corrosion. Duplicate analyses of one sample analyzed at an interval of 4 days differed by a factor of 50, indicating that the zirconium tends to precipitate or adsorb on container surfaces. As a result, the validity of previous analyses is questioned.

A radiation resistant television camera has been considered for in-reactor visual inspection of the PRTR Zircaloy-2 pressure tubes. Measurement of electronic properties of a special radiation resistant Vidicon showed no change after  $1.1 \times 10^9$  R accumulated gamma irradiation. This substantiates previous results in which no degradation in visual television picture quality was noted after this irradiation. This is the highest known successful irradiation of a television Vidicon tube.

A small hole through the cladding of a vibrationally compacted  $UO_2$ - $PuO_2$  fuel rod was found in a PRTR Mk-I fuel element that was suspected to be a leaker on the basis of stagnant water tests. No evidence of waterlogging was seen, and slight fuel washout occurred. Other suspected leaky elements were examined without discovering cladding defects.

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Center void and columnar grain formation, found in incrementally loaded, swaged  $\text{UO}_2$ - $\text{PuO}_2$  fuel rods for the first time, was accompanied by normal cladding behavior. Other out-of-reactor experiments confirmed that low temperature irradiation sintering observed earlier in swaged  $\text{UO}_2$  PRTR fuel rods was not a gross thermal effect.

Destructive examination of a PRTR Mk-II (nested tubular) fuel element after irradiation to 1360 Mwd/ton<sub>U</sub> revealed that internal pressures of released fission gases and desorbed impurity gases were less than anticipated.

A PRTR fuel element equipped with modified 360° contact-type end brackets and exposed for 2 weeks under PRTR conditions in an out-of-reactor loop, produced negligible fretting corrosion on the pressure tube.

After 60 days of cooling, the rejuvenation test fuel element (GEA 4-81) is being returned to HL from NRTS for "recharging" with enriched material.

Equipment for remotely disassembling and overhauling damaged PRTR fuel elements was fabricated and successfully tested in a simulated storage basin. Operational testing of the new manipulator system in the remote fuel fabrication studies facility was nearly completed. Operation of the system is satisfactory.

Ideal particle size distribution for vibrational compaction of fuel in an annular test element was calculated from a model developed by the Applied Mathematics Operation. Very high density (93% TD) was achieved, indicating the applicability of the model to novel fuel geometries.

High densities (99.0% TD) were achieved in high energy rate compaction of  $\text{UO}_2$  sintered scrap using higher impact pressures than previously believed possible with tool steel punches.

A test program for prototype EBWR  $\text{UO}_2$ - $\text{PuO}_2$  fuel rods was prepared. The first irradiation test capsules containing impacted  $\text{UO}_2$ - $\text{PuO}_2$  were assembled and shipped to NRTS for irradiation.

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Tungsten cladding components were joined successfully by magnetic force welding without producing large recrystallized grains that normally appear in fusion zones. The first successful joining of W-UO<sub>2</sub> cermet material was achieved by magnetic force welding.

A technique was developed for fabricating 4-inch-diameter, solid cermet discs approximately 1 inch thick by impaction. W-UO<sub>2</sub> cermets completely encased in tungsten were formed by loading the cermet powders in the impaction container surrounded by a nonfueled layer of tungsten powder.

Both phases in UN-W cermets reacted appreciably with oxygen at 1000 C, but little interaction occurred between phases.

The melting point of UOS, prepared by deposition from molten salt, was determined as 1880 C.

In cooperation with Chemical Laboratory personnel, simulated fission product oxides were impacted in both 2.5- and 4-inch-diameter containers. Materials impacted included SrTiO<sub>3</sub>, SrO, CeO<sub>2</sub>, and Nd<sub>2</sub>O<sub>3</sub>.

Self-damage of PuO<sub>2</sub> and  $\beta$ -Pu<sub>2</sub>O<sub>3</sub> from alpha radiation caused lattice expansions of  $0.35 \times 10^{-3}$  and  $0.70 \times 10^{-3} \Delta a/a$ , respectively, in 100 days.  $\beta$ -Pu<sub>2</sub>O<sub>3</sub> expansion of the "c" axis was greater than of the "a" axis.

Preliminary measurements of thermal conductivity were made on single-crystal and polycrystalline UO<sub>2</sub> to 2000 C, greatly extending the temperature range of previous measurements. As predicted, the thermal conductivity of the single crystal oxide was greater than that of the polycrystalline material.

The thermoelectric potential of UO<sub>2</sub> was successfully measured in-reactor. The polarity of the thermoelectric effect reversed from p- to n-type as the temperature increased.

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  $UO_2$  microcores, as small as 0.015-inch diameter, were extracted from  $UO_2$  samples by ultrasonic drilling. This new technique permits greater accuracy in studies of fission product relocation.

Fabrication of 11 Zircaloy-2 clad thorium-uranium fuel elements is complete. These elements will be autoclaved in preparation for irradiation testing in the ETR.

An optical strain readout system has been developed which has no real limit to its range, a characteristic that will be extremely useful in measuring the large strains involved in stress rupture testing.

Metallography samples from the AISI 316 SS inner liner of helium-cooled DR-1 gas loop were examined. Samples from the high flux-high temperature region, 100-1200 F, showed a large amount of second phase precipitate at the grain boundaries. Samples from lower temperature, 800-1000 F, no flux region, were free from this second phase.

Oxidation tests of Hastelloy X, a heat resistant nickel-base alloy, in laboratory air and pure oxygen have been made at 1000 C on as-received and abraded sheet specimens. Weight gain curves for all tests deviate no more than 15% from one another for the 48-hour duration of the tests.

The occurrence of minimum hydrogen pickup for 10% cold-worked Zircaloy-2, in-reactor, has been further explored by x-ray diffraction studies which showed a highly oriented texture in annealed material with the basal planes predominantly parallel to the rolling plane and a reduced intensity of orientation in this mode.

Two thorium specimens irradiated to burnups of 0.18 and 0.92 at. % at < 300 C are being vacuum annealed at 750 C for 100 hours. Metallography, hardness, and density will be determined.

Notched beams of 23% cold-worked Zircaloy-2 containing a sharp fatigue crack were tested in bending at temperature intervals from -196 to 22 C. It was observed that a sharp transition from ductile to brittle fracture



occurred from -60 to -80 C, and that the range was the same for specimens cut in either the longitudinal or transverse direction of rolled sheet.

In an in-reactor creep test on 20% cold-worked Zircaloy-2 at 20,000 psi stress and 350 C, the creep rate during a 10-day down-period which occurred shortly after the capsule was charged did not show the sharp increase typical of tests run previously at 30,000 psi.

Tensile testing has been conducted on several samples of iron of varying impurity content to characterize the role of interstitial impurities in the deformation mechanisms of unirradiated iron. The yielding behavior of these samples showed marked differences in behavior which were dependent on the purity.

Procurement of materials for the coordinated Irradiation Effects on Reactor Structural Materials Program is continuing. Plates 4 inches thick representing 18,000 pounds each of A-212B and A-302B pressure vessel alloys were received on site. Strip and rod of AISI 406 SS have been ordered and bids have been received for sheet, plate, and rods of AISI 304 SS, AISI 348 SS, and AM 355 alloys.

X-ray lattice parameter measurements on irradiated and unirradiated single crystals of molybdenum containing 10 and 100 ppm carbon show a difference of about 0.0003 A between the outside edge and the center of the electron beam zone refined crystals. This was not expected, since an opposite effect has been measured in polycrystalline specimens.

A total of 20 molybdenum single crystals have been tested in tension, utilizing a time lapse photographic technique to record the onset of double necking and other forms of nonuniform deformation.

Defect structures have been observed in high purity, polycrystalline molybdenum which contains < 10 ppm carbon as well as > 100 ppm carbon after exposures of  $10^{19}$  nvt (fast). The interaction of moving dislocations with these defects has been investigated. Transmission electron microscopy

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has revealed that the defects formed during irradiation are eliminated by moving dislocations, and that the latter operate only in discrete channels. These observations afford direct physical verification of what constitutes radiation hardening, and why coarse slip is preferred over fine slip.

Plastic deformation by compression of the beta phase has a marked effect on the  $\beta \rightarrow \alpha$  transformation of plutonium. Reductions in thickness at 150 C, in the range 60-97%, affect the density of the beta phase by as much as  $\pm 0.4$  g/cc and increase the density of the alpha phase formed at 79 C by as much as 0.1 g/cc. The incubation time necessary for the start of  $\beta \rightarrow \alpha$  transformation at 79 C in nondeformed metal is increased by a factor of 10-15 by the deformations used. The decreased rate of transformation after plastic deformation is an indication that the  $\beta \rightarrow \alpha$  transformation may be a nucleation and shear transformation.

The feasibility of reducing an "as cast" plutonium plate 0.100-inch thick by rolling at 70 C to a final thickness of 0.012-inch has been demonstrated. No work hardening is apparent and further evaluation is under way.

The basic Fast Supercritical Pressure Power Reactor (FSPPR) core design was simplified by providing for one instead of two moderating regions separating fast-core regions, and thereby reducing the required fuel enrichment by 2% (from 13% to 11% fissile). With some redistribution of enrichment, an acceptable power distribution can be attained while still maintaining safe reactivity coefficients. Plant efficiency of FSPPR is calculated as about 43.8%, versus 43.0% for the thermal SPPR. Fuel element fabrication cost, using Rene-41 tubes and jackets, is estimated at  $\$300 \pm 50$ /kg U + Pu, which, at a burnup of 100,000 Mwd/metric ton, results in a fuel cycle cost of about 1 mill/kwh.

Work is proceeding on a 10 Mw(e) epithermal reactor concept, fueled with "Phoenix Fuel" plutonium isotopes, to provide power for a base on the moon. This reactor concept utilizes plutonium nitride fuel, yttrium

or zirconium hydrides as moderator, and is lithium cooled. Power is recovered through a potassium secondary coolant system. Reactor coolant temperatures will be in excess of 2000 F.

## 2. Physics and Instruments

Reactor physics work in support of N-Reactor continued with the re-analysis of experimental data on the lattice parameters. Small corrections have resulted in the value of two parameters.

Studies of the physics problems associated with re-tubing production reactors were done using an exponential mockup of C-Reactor. Buckling measurements were made in an overbored lattice fueled with standard uranium elements.

In computer code development, a program was written that will be useful in determining the maximum permissible dimensions for variables in a program that will not exceed the computer core memory space. The GROUSS code was made more versatile by the insertion of the multilevel Breit-Wigner formulation for describing neutron cross section resonances. System checkout of the RBU code with the improved thermalization model began during the latter part of the month. The complete CALX burnup code system consisting of programs GAM, TEMPEST, SIGMA-3C, CALX burnup, and two small control programs chain linked together has been successfully run as a unit with CALX doing a burnup calculation based upon the cross sections generated by GAM and TEMPEST and assembled by SIGMA-3C. The linking of the programs by the computer will greatly decrease the data handling by the program user.

A N-Reactor pressure-injection analog simulation investigating the transient effects of loss of power to one or more pumps, process tube diversion, reactor power level excursions, and scram conditions has been completed with highly satisfactory results. Some 200 separate cases were investigated in a 2-week, two-shift per day study.

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HW-77046

Criticality studies on plutonium nitrate solutions continued in an 11.5-inch diameter water reflected sphere. Plutonium concentrations were in the range 80-220 g Pu/l with an acid molarity of about 0.3. Repairs were required on the dump valve for this system. The valve had been leaking solution into the dump tank.

Very promising results were obtained from pulsed neutron experiments performed in the criticality sphere mentioned above. Using this technique a good prediction was made on the critical volume of a plutonium nitrate sphere. This technique gives promise of leading to the development of a "criticality meter."

Studies on the criticality properties of plutonium precipitates and polymers moved a step closer this month with the installation of a remotely operated criticality machine in the second hood at the Critical Mass Laboratory. Tests are being performed on the speed of action of the control and safety rods for this machine.

Calculations were made which indicated that the maximum buckling obtainable with tubes of 1.03% enriched uranium does not exceed the maximum buckling obtained for optimum size fuel rods of the same material.

Five Nuclear Safety Specifications were reviewed. Participation in the activities of the Recuplex Deactivation Committee was continued.

Work continued on the development of a method for analyzing partially filled spheres of fissile material. Improvements on the handling of neutron leakage from the volume is being developed. The code was used to solve the problem of a hemisphere, and was found to have a precision of 0.6%. The subcritical assembly interaction program INTERSET is being generalized to account for interaction between results of various concentrations and vessels with surrounding reflection.

A D-Reactor production test was approved to evaluate equivalent multivariable reactor models which are being studied as a means for developing optimum reactor control systems.

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Measurements were started on the inelastic scattering of slow neutrons from light water at 95 C. Measurements were completed for neutrons with two different initial energies and with a single energy after scattering.

Work continued on the development of apparatus to measure slow neutron inelastic scattering using time-of-flight techniques.

Data obtained on the interaction of fast neutrons with  $\text{Pm}^{147}\text{-O}_2$  are being processed to subtract the effect of oxygen from the data. Nine new samples of different elements are ready for total neutron cross section measurements.

A more exact mathematical expression is being used in deriving differential scattering cross sections from the modified Gaussian fit to the energy level structure of water. This work is being performed in conjunction with an effort to improve the neutron thermalization model of water.

The effect of neutron energy spectrum on the reactor physics properties of Pu-Al fuels is being measured in the PCTR. This month an 8-3/8-inch graphite lattice was studied using 19-rod clusters of Pu-Al fuel having an isotopic content of 20.6%  $\text{Pu}^{240}$ .

The PRCF achieved criticality on March 21 with a loading of 25  $\text{UO}_2$  and 30 Pu-Al fuel clusters. The moderator level coefficient was measured and found to be about one-half that of the PRTR.

Work continued in preparation for the conversion of the PRCF to light water moderation. Assistance was rendered in development of design criteria for the conversion. Calculations were made on the worth of the control and safety systems in the converted PRCF.

Equipment is being assembled to begin subcritical measurements on Pu-Al fuels in light water. The fuel will contain 2.0 wt% plutonium containing 16%  $\text{Pu}^{240}$ . Templates are being assembled for the lattice spacing which will be 66 inches center to center.

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HW-77046

Studies to compare PuN, PuO<sub>2</sub> vs UO<sub>2</sub> as fuel for compact, fast spectrum space reactors are continuing. The present calculations are being carried out with the transport theory S-XI code which handles neutron interactions in more detail than the previously used diffusion theory code. Phoenix fuel core life studies for a wide range of reactor types and fuel loadings are continuing at an accelerated pace. Thinner fuel plates and higher Pu<sup>240</sup> content fuel than previously assumed are indicated from these studies.

Destructive burnup analysis began on a Mark-II UO<sub>2</sub> fuel element. The first two of the instrumented high exposure mixed oxide fuel elements have been delivered to the PRTR for insertion into the reactor. Work continued on the development of an analytical model to be used in analyzing the data coming from the PRTR.

Cobalt, plutonium, and uranium samples have been charged in KE-Reactor for the purpose of determining neutron flux parameters needed for the development of regenerating neutron flux detectors. The samples have received their required exposure and are scheduled for discharge at the next reactor outage.

The usable parameter separation range of the prototype eddy-current multiparameter nondestructive test instrument has been extended by the use of four-frequency driving signals. The probe spacing parameter is now linearly measured to 0.030 inch.

Attempts to develop eddy current methods of inspecting Zircaloy-clad thermocouple wires intended for N-Reactor installation are progressing favorably. A differential test coil operating at 250 kc has revealed several defect indications on typical specimens which are presently being examined destructively.

A project proposal for design funds for the High Temperature Lattice Testing Reactor is being circulated for approval signatures. Work on the final design criteria is progressing.

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Mass spectrometer data should be improved with the development of a scintillation type ion detector which is currently under way. Final design of such a detector is now in progress.

Operation of the recharging pocket dose meter has been extended to 17,000 cycles of continuous duty by the use of a platinum fiber and an electrically conductive Teflon center rod. Each cycle corresponded to accumulation of a radiation dose of 100 mr.

Work on the analysis of atmospheric dispersion data collected over the past 3 years continued with emphasis on relating measurements of dispersion to meteorological variables. Precipitation scavenging studies progressed through successful completion of a difficult scavenging field trial during the month.

Radiological physics work continued on:  $P^{32}$  counting, shadow shield calibration, X-ray scintillation counting, cosmic ray measurements, neutron source studies, and calorimetry of  $Pm^{147}$ .

The precision long counter for neutron measurements that was sent to Mound and Argonne laboratories for comparison studies was returned to Hanford to determine if it had remained stable. It may not have; two tests have indicated small but detectable changes. The changes indicated, however, were in opposite directions.

### 3. Chemistry

In-reactor tests show that the concentrations of  $As^{76}$ ,  $Np^{239}$ ,  $P^{32}$ ,  $Mn^{56}$ ,  $Cu^{64}$ , and  $Na^{24}$  in effluent water are significantly reduced (relative to control systems utilizing aluminum components) when Zircaloy tubes are charged with Zircaloy fuel elements and cooled with deionized water.

In half-reactor tests no significant difference in the corrosion rate of aluminum reactor components is observed when coolant channels are fed with high-alum-treated vice low-alum-treated water.

In laboratory studies, two organic, water dispersible coating agents were found which effected a ten-fold reduction of  $As^{76}$  adsorption onto aluminum surfaces.

Analytical results of ground water samples from deep farm wells located on the Columbia Basin Irrigation Project just east of the Hanford Project give no indication that contaminants of Hanford origin have entered the wells.

All vertical monitoring wells in the 241-A tank farm were relogged during March and the data obtained were compared with earlier (February) results; little change was noted.

The CSREX solvent--used to coextract strontium, cesium and rare earths from Purex waste--was found to degrade when contacted with a 1M  $HNO_3$  solution. Degradation of the solvent was not observed with 0.5M  $HNO_2$  solutions. Solvent attack is probably initiated by nitrite ion.

In laboratory studies, the use of uranium(IV) vice ferrous sulfamate as a plutonium partitioning agent in the Purex 1BX column is found to increase significantly plutonium reflux.

Hot air drying of selected natural and synthetic zeolites does not cause significant dusting of the ion exchange material nor entrainment of fines in the off-gas stream.

The volatilization of fission product strontium and cesium from synthetic zeolites during hot air drying tests ranges from 0.01 to 0.1%.

Hot cell equipment was fabricated to test the absorption of fission product strontium by a synthetic zeolite and to evaluate operational problems which might be encountered during hot air drying of the bed or as a consequence of long-term storage.

Crude beet sugar and cane sugar molasses are found to be potential replacement reagents, vice formaldehyde or sucrose, for effecting the denitration of Purex waste.

DECLASSIFIED

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xxi

HW-77046

Crystalline UOS was prepared by the  $H_2S$  sparging of a molten chloride melt containing U(IV).

Evaluation of the vibratory decladding of  $UO_2$  fuels showed that the average  $UO_2$  loss to the cladding from seven half-section cold-swaged rods and three half-section vibrationally compacted rods was 0.072 and 0.014%, respectively.

Crushing of electrolytic  $UO_2$  by parallel plates mounted in a hydraulic press has produced products with a particle size distribution required for vibrational compaction.

Operation of the 18-inch radiant heat spray calciner with synthetic waste containing mercury compounds showed that mercury volatilizes from the calcine at temperatures above 450 C but redeposits on the calcine in the off-gas train at temperatures below 350 C.

In the radiant heat spray calcination of alkaline Purex waste, contamination of the off-gas apparently results from fission product entrainment rather than volatilization.

A method for the determination of  $Pm^{147}$  in urine was developed which is short, simple and has a detection limit of less than 10 dis/min for a 10-minute count.

Long-lived, organic free radicals were observed during the  $Co^{60}$  irradiation of oxygen-free, p-nitroaniline solutions.

High density compacts of strontium oxide, neodymium oxide (stand-in for  $Pm_2O_3$ ), and ceric oxide were prepared by high energy rate impaction techniques (Dynapaking). A new container design successfully eliminated weldment cracks.

1103005

#### 4. Biology

No difference was noted in mortality of young chinook salmon exposed for 10 weeks to 3 and 6% reactor effluent water containing either dichromate or Quachrome Glucosate. Growth depression among fish in the 6% effluent was noted, however.

It appears that some trout can develop resistance to the columnaris organism by prolonged exposure to the disease. Small and large salmon fingerlings of the same age were equally susceptible to acute death from columnaris.

A surprisingly large fraction (0.7) of  $Zn^{65}$  orally administered to fish is excreted through the gill tissue. It also appears that young trout retain more  $Zn^{65}$  than older trout.

The discrimination of bones in pigs against  $Sr^{90}$  relative to  $Ca^{45}$  was found to change with time. This corresponds to theory and confirms earlier work done with rats.

A female miniature pig whose bones had received about 100 rads per day for 6 weeks due to the ingestion of  $Sr^{90}$  gave birth to an apparently normal offspring.

The  $I^{127}$  being fed to two of the three cows was increased from 5 to 15 mg per day. This caused a 40% drop in thyroid  $I^{131}$  and a slight increase in milk  $I^{131}$ .

Female miniature goats fed  $I^{131}$  daily are showing  $I^{131}$  in thyroids of three to four times the daily dose (similar to that observed years ago in sheep). Nursing offspring have thyroid burdens of 1/4 to 1/2 that of the mothers.

TTHA was found to be only slightly more effective in removing Pu than DTPA when given orally. In chronic treatment studies, where the two agents were given for 9 days, neither agent was found effective.

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HW-77046

One dog died after being exposed to about 6  $\mu\text{c}$  of  $\text{Pu}^{239}\text{O}_2$  for 1200 days. Cause of death was cardiac failure.

By using  $\text{C}^{14}$ -labeled taurocholate and cholic acid, it was shown that irradiation of the GI tract does not decrease absorption of bile salts from the intestine.

Like the effect of  $\text{D}_2\text{O}$  on animal cells, yeast cells exposed to  $\text{D}_2\text{O}$  showed decreased cell division rate, but little change in total growth.

Antennal characteristics in Tribolium beetles exposed to radiation appear to provide a convenient measure of radiation damage to the beetles.

Barley seedlings were shown to take up  $\text{I}^{131}$  from nutrient solutions three times faster during the dark periods than during the light periods, with the uptake of water being reversed. Since uptake of  $\text{I}^{131}$  is nearly completely dissociated from the uptake of water, we may have definite evidence of a specific metabolically driven process in plants.

The uptake into plants of  $\text{W}^{185}$  increased with increase in pH of the soil over about an 8-fold range.

Terrestrial ecology included work on the relationship between loss of soil moisture and vegetation growing on it. At Rattlesnake Springs, population measurements are being made of aquatic plant and insect communities.

## 5. Programming

$\text{Cm}^{244}$  production capabilities in civilian power reactors are being studied under a variety of conditions of exposure, feed material composition, reactors, and reactor operating variables. The preliminary results show quite clearly that for exposures considered normal for today's power reactor conditions, reasonably pure  $\text{Cm}^{244}$  cannot be recovered at discharge unless essential pure  $\text{Pu}^{242}$  could be used as the target material. The decay of a substantial fraction of the  $\text{Pu}^{241}$  throughout the exposure and the accompanying

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conversion of the resulting  $\text{Am}^{241}$  produces large quantities (10 to 80%) of  $\text{Cm}^{242}$  in the curium product. If it is assumed that  $\text{Cm}^{244}$  at recovery must not contain more than 2%  $\text{Cm}^{242}$ , such quality cannot be obtained without aging for 2 to 4 years to reduce the  $\text{Cm}^{242}$  content to acceptable levels. However, such an aging would result in the formation and possible simple recovery of valuable and fairly pure  $\text{Pu}^{238}$  equivalent to the  $\text{Cm}^{242}$  which has decayed.

Although civilian power reactors have potential for the production of higher isotopes when burning plutonium, they should also be important sources of high exposure plutonium that will be valuable as feed to selected reactors in which prolonged plutonium burning will be emphasized. Thus, if curium has a high value, certainly the precursor isotopes also have appreciable value. In fact, with  $\text{Cm}^{244}$  value in the range of \$400 to \$800 per gram, common isotopes such as  $\text{Pu}^{242}$  necessary for the formation of  $\text{Cm}^{244}$  will also tend to have an equivalent high value. This concept of a significant value for this nonfissionable, nonfertile plutonium isotope could have more than moderate repercussions in the area of power reactor fuel cycle analysis and in power reactor operation to adopt conditions favorable for its (and higher isotope) formation rather than its burning.

HW-76323, "Radioisotopic Heat Sources," by C. A. Rohrmann was issued and distributed March 27, 1963. This document discusses the characteristics of all radioisotopes which have serious potential as heat sources, including both nuclear characteristics and economic considerations.

#### TECHNICAL AND OTHER SERVICES

There were no new plutonium deposition cases confirmed during the month. The total number of plutonium deposition cases that have occurred at Hanford is 316 of which 229 are currently employed.

The annual distribution to each employee of individual exposure record cards indicating exposure from whole body penetrating radiations was completed.

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xxv

HW-77046

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The approach to the N-Reactor reliability study has been broadened to include an application of the Monte Carlo method for determining percentage of time spent by the reactor in any of the modes of operation being considered. This provides a very faithful model of the reactor system and the operating rules which apply to it, while avoiding the sticky problem of writing and solving a large number of simultaneous differential equations.

A topical study was made of the measurement error structure for the nondestructive testers used in the evaluation of fuel quality. Primary emphasis was given an evaluation of the economic consequences of the existing measurement errors. It was concluded that immediate steps should be taken to eliminate biases, but that at present, there seems to be little incentive for improved precision. Concrete proposals for reduction of bias were presented.

In pursuance of the solution to the rail height specification problem for self-support K-Reactor fuels, an expression was found relating minimum annulus between the fuel element and the process tube wall as a function of tube and fuel element radii, rail height, and angle subtended at the fuel center by the rails. This expression, which takes into account the fact that both rails supporting the underside of the fuel are more than likely not equal in height, is being used in a computer simulation which gives the distributions of minimum annulus as functions of input distribution for the independent characteristics.

A rough analysis was made of rupture data for the past 2 years to estimate what proportion of ruptures are "atypical". This estimate was found by examining the distribution of "multiple-failure" charging dates, confounded with "multiple-failure" lots, and was checked by observing the proportion of ruptures occurring at exposures less than 50% of goal. Under the existing rupture model, about 3% of the "normal" ruptures will occur at these low exposures.

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A review is being made as requested of the current CPD practice of using monthly bias corrections for analytical results based on monthly analyses of standards.

Consulting assistance was provided in expressing minimum fuel cost as a function of price values assigned to the isotopes of plutonium, to Am<sup>243</sup> and to Cm<sup>244</sup> for each of several reactor cases and associated ground rules of operation. For the cases considered to date, very simple expressions were found.

An analysis of a series of tests of shear-spinning preformed metal blanks into selected shapes indicated the desirability of possessing a means of designing such blanks which would be rapid, yet flexible to alternative selections of design parameters. An EDPM program, based on a modified uniform shear theory, and which exhibits the desired flexibility, has been written and placed in service.

The EDPM program which generates the magnetic tape input to the experimental  $\delta$ - $\omega$  lathe, has been slightly modified in an attempt to prevent nonuniformly spaced bits from jamming the pulse motors. A complete 1251 exterior contour tape has been generated using the modified techniques and is presently under study on the lathe.

The EDPM program for obtaining numerical solutions to a simultaneous set of first order ordinary differential equations has been completed. A function comparison subroutine was written and added to the basic program. This program is now being used to study the N-Reactor stack gas chemical reactions which are vital to the Zirconium-Graphite compatibility problem.

An experimental model of a vibrationally compacted annular shaped fuel element was fabricated to a density of 93.3% of theoretical. This sets a new density record.

During March a new and unknown crystal was indexed from a powder pattern on an orthorhombic basis using the program being developed for

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this purpose. Subsequently, a single crystal of the compound was analyzed independently, which verified that the computer solution was correct. It is believed that this is the first time a comprehensive computer program has ever indexed a complicated crystal, such as an orthorhombic one, exclusively from powder pattern data, and subsequently to have had the indexing prove correct.

Many improvements were made and many errors corrected in the new IRA II system during March. These improvements have significantly reduced the running time on several passes. Several successful processings of large amounts of program data were completed. Work continues on the specification of improvements to the system.

#### SUPPORTING FUNCTIONS

The output of PRTR for March was 963 Mwd for an experimental time efficiency of 63% and a plant efficiency of 57.6%. There were six operating periods during the month, one of which was terminated by a scram as a result of pressurizer high pressure following malfunction of a pressure control valve. Two shutdowns were made because of high D<sub>2</sub>O collection in the recovery system, two were a result of personnel errors, and one was required when both high pressure helium compressors failed to function properly.

Fuel exposure history at month-end was:

Maximum UO <sub>2</sub> exposure/element	3030 Mwd/ton <sub>U</sub>
Average UO <sub>2</sub> exposure/element	2124 Mwd/ton <sub>U</sub>
Maximum Pu-Al exposure/element	85.8 Mwd
Average Pu-Al exposure/element	69.8 Mwd
Maximum Moxtyl exposure/element	49.1 Mwd (~982 Mwd/ton <sub>U</sub> )
Average Moxtyl exposure/element	32.0 Mwd (~640 Mwd/ton <sub>U</sub> )

A total of 5.9 kg of plutonium was recovered at the Redox plant, from 32 irradiated Pu-Al fuel elements. Composition of the final Redox product was as follows:

Pu <sup>239</sup>	74.678
Pu <sup>240</sup>	21.468
Pu <sup>241</sup>	3.418
Pu <sup>242</sup>	0.440

A total of 104 outage hours was charged to repair work. The majority, 62 hours, was required for high pressure helium compressor repairs, 16 hours were required for gasket replacement, and 18 hours were required for helium valve repairs to eliminate in-line leaks. Compressor difficulties were traced to severe wearing of piston rings. Four tube-to-nozzle and 7 jumper-to-tube gaskets were replaced.

Nineteen uninterrupted days of beneficial use were realized in the Plutonium Recycle Critical Facility. The initial loading was established. The initial criticality milestone occurred on March 21.

Mechanical design testing of the Fuel Element Rupture Test Facility was 90% complete. A 24-hour-test at 2000 psig and 600 F was completed on March 24, 1963.

Project status of the Gas-Cooled Loop Facility remains at 94% complete. Blower delivery was again delayed due to a second thrust plate failure. The vendor was advised to ship these blowers by mid-April.

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Technical Shops Operation's total productive time for the period was 23,194 hours. Distribution of time was as follows:

	<u>Man-Hours</u>	<u>% of Total</u>
N-Reactor Department	3 845	16.58
Irradiation Processing Department	3 964	17.09
Chemical Processing Department	617	2.66
Hanford Laboratories	14 768	63.67
Construction Engineering and Utilities	0	0

Total productive time realized for Laboratory Maintenance Operation was 18,100 hours of a possible 19,100 hours potentially available. Manpower utilization for March is summarized as follows:

	<u>Hours</u>	<u>Hours</u>
A. Shop Work		3200
B. Maintenance		7600
1. Preventive Maintenance	1900	
2. Emergency or Unscheduled Maintenance	1900	
3. Normal Scheduled Maintenance	3800	
4. Overtime	500	
C. R&D Assistance		7300

Heavy water physical inventory at the end of March indicated a loss of 1185 pounds valued at \$16,365. This loss represents 1165 pounds applicable to PRTR and 20 pound to PRCF. Scrap generated during the month amounted to 1644 pounds (\$1400).

A review by Accounting Operations of the zirconium inventory records maintained by Contract and Accounting disclosed that the zirconium inventory account was overstated \$67,166 at December 31, 1962 and that costs have been understated by an equal amount. The overstatement represents almost entirely testing and inspection costs that were not charged to operating costs as the material was withdrawn from the inventory for use. An adjustment was made in March to correct the records.

The Capital Equipment control budget was decreased \$93,000 to agree with the most recent financial plan issued by RLOO-AEC.

Activity in the area of visitor presentations is summarized below:

	(Number Participating)			
	<u>Visitors Center</u>		<u>Plant Tours</u>	
	<u>Number</u>	<u>People</u>	<u>Number</u>	<u>People</u>
March: Groups	6	220*	16	329
Individuals	-	1 533 (*included)		329
Cumulative Since 6-13-62: Individuals	-	40 025	--	---

Professional recruiting activity this month follows:

	<u>Plant Visits</u>	<u>Offers Extended</u>	<u>Acceptances Received</u>	<u>Rejections Received</u>	<u>Open Offers at Month-End</u>
Ph. D.	9	5	1	-	7
BS/MS (Direct Placement)		9	3	1	12
BS/MS (Program)		61	22	16	162

Five technical graduates were placed on permanent assignment. Two additions to the program were made and two terminations occurred; its current strength is 41.

Authorized funds for eight active projects total \$6,300,500. The total estimated cost of these projects is \$7,745,000 of which \$650,000 had been spent through February 28, 1963.

  
 Manager, Hanford Laboratories

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REACTOR AND FUELS LABORATORY MONTHLY REPORTMARCH 1963TECHNICAL ACTIVITIESA. FISSIONABLE MATERIALS - O2 PROGRAM1. Metallic Fuel Development

Fuel Irradiations. Detailed post-irradiation examination of the second "N" outer fuel element component (NOE) to be irradiated at prototypic conditions has been completed. No unusual features or shortcomings were observed in the Be-Zr eutectic brazed closure, the fuel, clad, or the fuel-to-clad bond. A slight build-up of corrosion product has been observed in a tight crevice between a spot-welded support and the fuel clad. The nature of the deposit in the crevice, however, suggests that the corrosion occurred during autoclave testing and not during reactor irradiation. A third NOE has been received at the Radiometallurgy facility for additional post-irradiation examination.

Two fluted single-tube N-Reactor size fuel elements completed the second cycle of irradiation in the ETR M-3 pressurized loop facility. An exposure of approximately 400 MWD/T has been accumulated on these test elements which will remain in place for another cycle before removal for an interim examination.

Two other irradiation tests of fluted elements have both reached an exposure of about 1300 MWD/T. In one test, three N-inner size elements are being irradiated in cold water in the KE Reactor. In the other test an N-inner size fluted element has completed five cycles of irradiation in the pressurized high temperature P-7 loop of the ETR.

MTR Irradiation of I & E Fuel Elements. Recent irradiations of thick walled uranium tubes by SRL have resulted in volume increases considerably in excess of those that would be predicted solely from fission gas swelling. Examinations have shown that extensive grain boundary tearing has occurred in the uranium resulting in large numbers of irregular shaped voids. Four Hanford I & E fuel elements are being irradiated in the GEH-4 loop facility in the MTR to investigate the possible occurrence of large volume increases in these elements at intermediate temperatures. The average maximum metal fuel temperature during the last cycle was 380 C. The fuel elements,

which are now at an exposure of 500 MWD/T, have been temporarily removed from the reactor to make space available for a short term experiment. Goal exposure of the I & E elements is approximately 2000 MWD/T.

Fuel Measurements Programs. Modifications of the measurement data analysis program have been made which give card outputs suitable for "mercy" evaluation of measurement errors. A program has been initiated to statistically determine the measurement errors which exist between the pre- and post-irradiation measuring equipment and three conservative measurement runs each on 10 inner and 10 outer N-Reactor type fuel elements have been completed on the pre-irradiation machines. New reeds have been installed in the post-irradiation measurement machine and the machine is being checked to determine whether this modification has eliminated a two- to three-mil drift which had previously developed.

In-Reactor Fuel Deformation Studies. An in-reactor strain-cycling capsule and its associated instrumentation have been shipped to the ETR for irradiation testing. Both the temperature of the uranium specimen and the deformation rate for given load conditions will be measured while the specimen is being irradiated, thus providing data on the effects of fission on the plastic properties of uranium.

"Self-Brazed" Closure Development. Steps are being taken to get two lots of "self-brazed" closure fuel elements irradiated under a production test. This will include three specimens beta-heat treated after closure, and three beta-heat treated before making the closures. The latter procedure offers advantages in time, economy and surface quality of the finished element, but the compatibility of this procedure with irradiation requirements has not yet been evaluated.

Specimens being given long-term corrosion testing in the TF-7 high temperature loop in K-East, including one whose weld bead had been removed to lay bare the braze annulus between cap and clad, appear unattacked after 84 days of exposure.

Lithium-Aluminum Target Element. An investigation to establish the irradiation performance of tritium target materials under N-Reactor conditions was initiated this month. A design for the proposed irradiation test has been completed. The assembly consists of an Al-Li alloy cylindrical target core (0.250" ID x 0.625" OD x 7.50" in length) doubly canned, unbonded, in 0.030" C-64 Al alloy and 0.035" Zr-4 alloy. The final closures of each can are to be made in helium in order to minimize heat transfer resistances in the finished element. The void volume of the target core is calculated conservatively,

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and is designed to accommodate any swelling which would result from precipitation and agglomeration of helium and tritium produced by the  $\text{Li}^6 (n, \alpha)\text{H}^3$  reaction. The primary objectives of the planned irradiation test series are to determine under N-Reactor conditions: (1) the rate of swelling of the over-all target assembly and the target core; (2) the effect of irradiation on mass transport of Li and tritium in the target assembly; and (3) the effect of surface treatments of Al on tritium transport. The fabrication of the target assemblies has been started. Two Al-Li target alloys containing 0.91 and 2.10 w/o Li, both with the same  $\text{Li}^6$  concentration (0.39 w/o), were melted and cast. Seven billets of each composition, preheated to 200 C, were extruded at an extrusion ratio of 12 to 1 and dry machined to final dimensions. Twenty-five of the Al-0.91 w/o Li target alloy cores were subsequently canned in C-64 Al alloy, the 2S-Al alloy end caps were inserted and the resulting assemblies die sized. The final fabrication steps are: welding of the Al-end caps, canning in Zr-4 and welding of supports to the Zr-4 can.

A series of annealing studies in the range of 300-600 C results in precipitation of a second phase (presumably Al-Li compound) in the 2 w/o Li alloy. At 600 C for the same alloy, large grain growth was observed with the deposition of material (presumably oxide inclusions) at the grain boundaries. The 1 w/o Li alloys were apparently stable in the temperature and time range of the experiments.

Cold pressure welding is being used for joining 2-S aluminum to aluminum-1½% lithium. The samples are used for diffusion studies and require a narrow, straight interface. One-inch diameter by 1-5/8 inch pieces are placed in holding dies and then pressed together (end to end) with a 44-ton force. After the upset is removed the piece is again pressed. Thirty-five percent upset on the first pressing and 50% on the second. Double pressing reduced the amount of trapped oxide in the joint. Fully annealed Al-Li against as-extruded 2-S produced a narrow, even interface.

Analysis of Fuel Element End Caps. The difference in thermal expansions between the fuel and end cap material in N-Reactor fuels is a potential cause of fuel element failure. A numerical method of stress analysis for end closures with thickened or reinforced cladding near the cap has been run on a number of shapes. Reductions of stress by a factor of from three to four seems possible by the proper shaping of the closure.

N-Reactor Support Development. Fabrication of N-fuel supports continued as required by NRD. The following quantities of supports

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were produced during March: shoes, 24,000; inner supports, 22,000; outer supports, 9,000.

Fuel Supports - Process Tube Wear. Laboratory tests to study possible damage to N-Reactor process tubes by charging N-fuel elements are proceeding. A series of tests are being conducted to determine to what extent the reactor process tube is damaged by the repeated corrosion of the bare Zr-2 that is exposed by scratching by the fuel element supports. The test cycle consists of passing steel support shoes 40 times over a single path on the process tube, then autoclaving the tube in 400 C, 1500 psi steam for 14 days to give a corrosion product build-up on the bare Zr-2 equivalent to that expected to form in-reactor between the charge and discharge of the fuel elements. After the third autoclave cycle, despite the fact that the elements were passing over a previously scratched area of the tube, a total of 50 passes with four different shoes were made with no signs of scratching the autoclaved film. Steel was deposited on the autoclave film from the shoes, each of which experienced considerable wear. Scratching was then intentionally initiated, with considerable difficulty, and 40 passes made over the same single path. Four series of 40 passes, with autoclaving between each series, have now been completed on the same region of the process tube section. The scratch depth along the tube varies from 0.0002 to 0.0014 inch. The average depth over the entire length of the tube increased from 0.0008 inch to 0.0009 inch. Further tests will be run to determine the relationship of additional corrosion and charging cycles to the scratch depth.

Cladding Deformation Studies. Thirty-six NaK capsules containing a total of 94 Zr-2 clad uranium rods are being irradiated in F-Reactor to provide data on the effects of cladding thickness uniformity, temperature, and exposure on the strain capabilities of Zr-2 fuel cladding. Four of the capsules have thermocouples to measure the central uranium temperatures. One thermocouple developed a high resistance after approximately eight days of irradiation. Efforts to repair the thermocouple have been unsuccessful. The other three thermocouples are recording satisfactorily, and agreement between measured and calculated central uranium temperatures is good. The latest temperature and power data indicate that the irradiation is continuing to operate with the cladding temperatures very close to those desired. Exposure has now reached approximately 1100 MWD/T. Goal exposure for nine of these capsules has thus been reached and these will be discharged at the next reactor shutdown. The fuel samples in these nine capsules will be examined as quickly as possible and the information obtained from them will be used to establish discharge exposures for the remaining 27 capsules.

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Heat Treatment of Zircaloy Clad Fuel. N-outer tube stock with the core material containing 640 ppm Al and 400 ppm Fe is being studied to determine the effects of heat treatment on the size and dispersion of  $UAl_2$  and  $U_6Fe$  compounds. Specimens quenched from the beta and gamma phases were examined after 100, 200, and 500 hours at 400, 450, and 500 C. Growth of the  $U_6Fe$  phases was observed in longer times at 450 and 500 C, indicating that the iron addition does not form a stable dispersion at fuel operating conditions. The  $UAl_2$  particle size appears stable after these treatments. The dispersion of  $UAl_2$  is random for material precipitated after beta phase treatment, but is preferentially aligned in a sub-boundary network in material precipitated after gamma phase treatment. Additional etching procedures were used for electron microscope study of these materials in attempts to arrive at a standard procedure yielding true structure. This work is continuing. Additional samples of the material have been gamma phase treated with the cooling rate varied in order to study the effect of cooling rate on sub-boundary structure development.

The mechanisms discussed give rise to a basic difference in structure between as-cast and beta heat treated material, and wrought and beta heat treated material. In the as-cast case the original sub-boundary network is still present after beta treatment, whereas in the wrought material the sub-boundary structure has been broken up by working the metal. The effect of this difference in structure on swelling and grain boundary tearing should be determined.

Phosphorus Impurity in Uranium. Phosphorus, in quantities up to 400-500 ppm, has been introduced into the 0.94% enriched uranium ingot for the N-fuel and I & E fuel material as a result of scrap recovery process changes at the AEC Feed Materials Production Center, National Lead Company. The effect of this impurity on uranium structure, fabricability, and properties and fuel element quality is not known. The uranium-phosphorus phase diagram has not been determined except for the identification of the compounds UP,  $U_2P_4$ , and  $UP_2$ . UP (11.52 w/o P) is reported to have a melting point of approximately 2200 C. The evaluation to date has included a section of derby metal containing about 430 ppm P and sections of cast and beta heat treated NIE (150 ppm P) and NOE (300 ppm P) ingots.

Metallographic examination of the derby metal as reduced as gamma heat treated, and as remelted has indicated that, unlike C, O<sub>2</sub> and N<sub>2</sub> impurities, the phosphorus forms a eutectic between UP and uranium, believed to be at 1000-1500 ppm P. There appears to be little, if any, solubility of phosphorus even in the gamma uranium

phase. Comparative melting point determinations have shown approximately 10 C depression due to the eutectic. The eutectic structure was observed also in the top, middle and bottom sections of cast and heat treated N-fuel ingots. Chemistry distribution was determined as shown in the table below. No pronounced segregation of phosphorus is evident. Complete spectrographic analysis has not shown any other impurity at unusual concentration except aluminum at approximately 100 ppm in Ingot 36421

Phosphorus, Iron, and Silicon Distribution  
in N-Ingots (in ppm)

<u>Ingot 36421 (NOE)</u>	<u>Top Edge</u>	<u>Top Center</u>	<u>Middle Edge</u>	<u>Middle Center</u>	<u>Bottom Edge</u>	<u>Bottom Center</u>
Phosphorus						
HAPO	370	322	385	322	349	340
NLO	298		293		262	
Iron (HAPO)	150	160	160	160	170	220
Silicon (HAPO)	50	60	48	58	65	60
Carbon (NLO)	640					
Oxygen (NLO)	7					
Nitrogen (NLO)	8					
Density (g/cc)	18.75					
<u>Ingot 36407 (NIE)</u>						
Phosphorus						
HAPO	163	187	168	168	169	165
NLO	141		135		128	
Iron (HAPO)	180	200	180	230	200	170
Silicon (HAPO)	65	58	73	73	78	58
Carbon (NLO)	576					
Oxygen (NLO)	5					
Nitrogen (NLO)	8					
Density (g/cc)	18.89					

Sections examined from the center of these ingots which were heat treated hollow did not show any cracking or grain boundary tearing due to the quenching stresses in heat treating. A section of the ingot containing approximately 300 ppm P was hot rolled at 600-625 C to about 70% reduction in area. No unusual behavior was observed and the structure after this fabrication showed only a compression and extension of the eutectic network. These ingots will be primary extruded at HAPO and billet sections prepared for co-extrusion for further evaluation of fabricability, properties and effect on fuel element quality.

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## 2. Corrosion and Water Quality Studies

Effects of Velocity on Corrosion of Aluminum - Rotating Disk Method. Studies of the effect of velocity on the corrosion of aluminum are being conducted in a rotating disk apparatus. A rotational speed of 4150 rpm is used, giving maximum peripheral speeds of 72 fps on four-inch diameter disks and 54 fps on three-inch diameter disks. Preliminary tests have been conducted at 80 C and 92 C in 300 Area water and reactor process water.

The most striking evidence of erosion occurred on a four-inch disk rotated in 300 Area water at 90-95 C. At the outer edge complete film removal occurred; the erosive attack appeared to extend into the metal, giving rise to a phenomenon similar to ledging attack observed on aluminum process tubes. Within a band about 0.25-inch wide, the film was highly eroded, resulting in an area of "plateaus and valleys." The erosive attack was essentially uniform around the periphery of the disk, but tapered off quickly toward the center and was almost absent within 0.5 inch of the periphery.

At 80 C in 300 Area water, much less film formed on the four-inch disk than in the test at 90-95 C. At the lower temperature the erosive effect was much less pronounced, although areas were observed near the outer edge of the disk where sections of oxide had apparently lifted out of the film.

Weight losses (averaged over the entire sample) in tap water for 72-74-hour exposures were 2.5 mg/cm<sup>2</sup> and 2.8 mg/cm<sup>2</sup> on three-inch and four-inch samples, respectively, at 90 C. At 80 C the weight loss was 0.55 mg/cm<sup>2</sup> on a four-inch disk.

On a four-inch disk rotated in reactor process water for 72 hours at 90-92 C, the weight loss was 2.8 mg/cm<sup>2</sup>, equal to that observed in tap water under similar conditions. However, the erosive attack observed in tap water was almost completely absent on the sample rotated in process water. Process water taken from the tank where the disks are rotated showed a pH of 8.4, compared to an inlet pH of 6.8; the increase in pH was undoubtedly due to leaching of boiler scale deposited during prior runs in tap water. On five samples rotated in tap water, the upper surface was dark, while the lower surface had large light-colored areas. In process water, both surfaces were uniformly dark.

Process Assistance to N-Fuel Fabrication. Process assistance to N-fuel fabrication during the past month has been divided into two phases of related work. The first phase consists of categorizing and grading N-fuel elements with various amounts and types of white

oxide. The second phase of the work consists in determining causes of these various white oxides along with possible methods of elimination.

The two most questionable categories are grey to white oxide on the end welds and spot weld defects. The grey to white oxide on the end welds is probably caused by uranium contamination in the weld metal. Although long term corrosion tests in 360 degree pH-10 water on this type of weld have not shown an excessive amount of corrosion penetration, considerable oxide sloughing has been observed. Such sloughing could conceivably cause high radiation levels in the reactor and complicate contact maintenance. A joint testing program with N-Fuels Product Engineering and CMO personnel is being conducted to verify uranium contamination as cause of this sloughing.

The spot weld defects on the Zircaloy-2 cladding are caused by arcing between the fuel element holder (copper or Zircaloy-2) and the Zircaloy-2 cladding. Visual inspection does not indicate the extent of damage caused by localized overheating or introduction of copper as a contaminant. Several specimens are presently being sectioned for a metallographic study to establish what damage may be expected.

The remaining categories of off-standard surface appearance are less serious. They consist mostly of localized grey to white oxide on either the Zircaloy-2 clad or attached hardware. In general, these defects are caused by improper rinsing which results in acid staining. Although trivial in itself, acid staining can mask serious corrosion associated with metal quality.

Nickel Plating. The properties of two nickel sulfate plating baths were investigated at room temperature. One bath containing 200 g/l of nickel sulfate, 40 g/l sodium citrate and 5 g/l ammonium chloride had good throwing power at 15 amp/ft<sup>2</sup>. Ultrasonic agitation of the plating solution at 33 Kcs resulted in less gas pitting, higher cathode efficiency and a brighter plate. The plate was no longer bright at 72 amp/ft<sup>2</sup> even with ultrasonic agitation. The other bath containing about half the nickel sulfate concentration had less throwing power.

A number of two-inch sections of standard uranium fuel material were anodically etched at 100 amp/ft<sup>2</sup> in 10% nitric acid, and nickel plated. Delay in transfer between solutions probably led to the poor adhesion observed in some plates. Equipment is being fabricated to eliminate this problem.

Use of  $\text{NH}_4\text{OH}$  to Adjust pH in Pressurized Water Systems - KER Testing. Present experiments in KER-1 are designed to determine effects on water quality of changing pH and substituting  $\text{NH}_4\text{OH}$  for  $\text{LiOH}$ . The initial phase included operating with  $\text{LiOH}$  at pH 10 until equilibrium conditions were attained and then operating with  $\text{NH}_4\text{OH}$  at pH 10 to determine crud levels, crud deposition, and operating problems.

The desired initial operating conditions were never obtained due to the erratic operation of the reactor and the loop during this period. On four occasions operation with  $\text{LiOH}$  at high temperature was initiated; however, in each case operation was terminated by loop leaks or reactor scrams before normal coolant conditions were established. Since the required conditions were not achieved,  $\text{NH}_4\text{OH}$  addition was not initiated. These studies have been rescheduled.

On two occasions the loop was operated in-reactor at low temperature (70-80 C) due to severe leakage at the front-face fitting. The first period under these conditions lasted 8 days. During this time the oxygen and hydrogen peroxide concentrations increased drastically due to radiolytic decomposition of the coolant even though the loop was operated at the maximum feed-and-bleed and degas rates. These materials increased the oxidative characteristics of the coolant to such an extent that appreciable amounts of magnetite were converted to ferric oxide and released to the coolant. Extremely high turbidities were encountered for several days following resumption of high temperature operation.

The second period of low-temperature, in-reactor operation lasted 3 days. During this period all necessary preparations were made to perform a hydrazine injection test to determine whether this material can satisfactorily reduce the oxidative characteristics of the coolant. However, a reactor scram occurred about 25 minutes after hydrazine injection was initiated, and it was necessary to discontinue the test. Considerable ferric oxide formation was also observed during this period, and crud levels remained high for 2-3 days after high temperature operation was resumed.

The reason for the high crud levels observed following routine shutdown-startup operations during the past two months is not fully understood. It is not unusual to observe crud levels above normal following these operations; however, the levels encountered recently have been extremely high by comparison. It is probable that the crud problem has been aggravated considerably by the frequent cleaning operations in this loop and by exposure to coolant with large amounts of oxidizing materials present. It is also possible that residual chemicals remained in the loop after chemical cleaning was completed. Both possibilities are being investigated.

Stainless Steel Clad Thermocouple Elements. One of the primary variables to be evaluated during the coming in-reactor test in KER-1 is the effect of  $\text{NH}_4\text{OH}$  on rates of formation and deposition of films. The rate of deposition of films (crud) on the fuel elements can best be followed with a thermocouple fuel element. Special S/S clad,  $\text{UO}_2$  thermocouple elements were designed and the first test assemblies charged in to the TF-loops during this past month for pre-irradiation testing. The thermocouple elements are of two types, designated as Mark II, and differ with respect to placement of the thermocouple. Both types are encased in a Zr-2 sleeve without endcaps.

Two Mark I elements were charged into TF-9 and dynamically tested at 300 C. After the first three inspection shutdowns at 108, 285, and 423 hours, the elements are in excellent condition and no visual defects have been detected. One of the elements had a tight Zr-2 jacket (shrunk on) and the other was slip fitted. However, at the end of 108 hours, the tight Zr-2 jacket had expanded and had about the same clearance as the slip-fit element. It was originally thought this would be important; however, the amount of corrosion of the S/S under the jackets appears to be the same, and jacket tightness does not appear to be of major concern. These two elements have had three 300-20 C thermocycles - the thermocycling experienced from normal startup and shutdown.

One Mark I element was charged into Manifold No. 3 of TF-7 for thermocycling. The element has completed 55 cycles and appears to be in excellent condition also. Two Mark II elements were charged into TF-9 and the third element will be charged into TF-7 during the next shutdown.

Continuous Analyses of Sodium in N-Reactor Secondary Water. Evaluation of the sodium magnesium versenate procedure for water hardness measurement was completed. The procedure has been adapted for use with a continuous wet chemical analyzer. Laboratory studies have confirmed that the procedure has satisfactory reproducibility and that hardness concentrations as low as 10 parts per billion can be detected without difficulty. This procedure will be used to continuously monitor the N-Reactor secondary coolant for raw water in-leakage.

Corrosion of Carbon Steel at Low Temperatures. A small, low-pressure, recirculation facility was designed and constructed to measure the corrosion rate of carbon steel in room temperature water. Studies are scheduled with deionized, deoxygenated water and with filtered water. The system will also be used to study the effects of various

filtered water additives. These studies will have direct application for emergency cooling conditions at N-Reactor. The system is designed to utilize corrosometer probes for corrosion measurements.

Alternate Inhibitor Studies. Corrosion penetration of aluminum coupon samples was less in quachrom glucosate inhibited water than in process (dichromate-inhibited) water, but corrosion of carbon steel during the first exposure period was greater in the quachrom glucosate water. First exposure of coupon samples and K<sub>4</sub>N test elements was completed in 0.20 ppm quachrom glucosate water in 1706 Facilities SP-5 and SP-6, with control exposure in SP-2, SP-X, and SP-Y. Until additional results are obtained, firm conclusions cannot be made.

Decontamination of Low Temperature Reactors. The most effective safe procedure for decontaminating the low-temperature reactors is one utilizing the proprietary sulfanic acid, Turco 4306-C. Some tests were completed during the month to determine the optimum time and concentration. Perforated aluminum spacers were used as samples for a test of Turco 4306-C solutions, with contact times of 10, 12.5, and 15 minutes. The chemical concentration of 3 oz/gal was effective in reducing both gamma and beta radiation to near background levels. At a chemical concentration of only 1 oz/gal, a minimum of 12 minutes contact was required for significant reduction of gamma activity, but neither 1 or 2 oz/gal concentrations were sufficient to reduce beta radiation to background levels. Apparently, 3 oz/gal is the minimum effective concentration.

Determination of Activities Deposited on Fuel Elements in Present Reactors. The radioactivity in the effluent of the present reactors results principally from the adsorption of ionic or colloidal components in the water onto the fuel element surfaces. These adsorbed species are activated and later released. To obtain a better understanding of the adsorption and activation processes, some studies were made of activity distributions on a typical fuel charge in the KE-Reactor.

The radiochemical data from the second KE fuel element column have been resolved into equations for activity distribution as a function of column length. Computer curve fitting by the technique of least mean squares has given curves of the form  $\ln y = a + bx + cx^2$  for Sc-46, Cr-51, Fe-59, Co-58, Zn-65 and Sb-124, where y represents activity/foot and x the length variable. These equations are as follows:

- (a)  $\ln y = 4.708 + 0.741x - 0.023x^2$  for chromium-51
- (b)  $\ln y = -0.831 + 0.408x - 0.012x^2$  for antimony-124
- (c)  $\ln y = 2.474 + 0.513x - 0.014x^2$  for zinc-65
- (d)  $\ln y = 2.996 + 0.577x - 0.019x^2$  for cobalt-58
- (e)  $\ln y = 0.362 + 0.791x - 0.023x^2$  for scandium-46
- (f)  $\ln y = 1.751 + 0.618x - 0.019x^2$  for iron-59.

Integration gives the total amount of each isotope held in the films of this KE fuel column. These amounts are as follows:

- (a) 0.534 curies of chromium-51
- (b) 0.244 millicuries of antimony-124
- (c) 0.0204 curies of zinc-65
- (d) 0.0192 curies of cobalt-58
- (e) 0.0171 curies of scandium-46
- (f) 0.0103 curies of iron-59.

The amount of activity from the deposited crud was related both to water temperature and flux pattern. From the preliminary figures cited above, it appeared that the maximum activity occurred at the point of maximum corrosion.

Removal of Excess VPI from N-Reactor Primary System. During layup of the carbon steel piping at N-Reactor, vapor phase inhibitor (VPI) was added to prevent rusting and the pipes were sealed from the atmosphere. The amount of VPI added was excessive and complicated removal. Laboratory tests demonstrated that the undecomposed VPI could be removed with hot water. However, areas of thin black scale remained in portions of the piping which had been heated during welding. This layer is believed to be carbon. Tests are continuing to identify the material and evaluate corrosion effects which may occur if this material is not removed.

Removal of Oxides and Mill Scale From N-Reactor Secondary System. Two commercial procedures for descaling N-Reactor secondary system are being evaluated in laboratory and loop (TF-14) tests. A one-

step procedure (Vertan 675) successfully removed all oxides and mill scale from TF-14 in  $3\frac{1}{2}$  hours at  $\sim 200$  F. The pipe walls were left in a passive condition. Another procedure (Turco 4306 D followed by Turco 4517) will be evaluated next month.

In another dynamic test (TF-17), the ammonium citrate process was tested and proved very successful in descaling the loop of rust and mill scale remaining from fabrication. The one-step process consisted of recirculating 5% ammonium citrate for four hours, adding an additional 0.5% ammonium citrate (a small amount of concentrate was added) to prevent precipitation of iron citrates, adjusting the pH to 9 with ammonium hydroxide, and the addition of 0.1% sodium nitrite for passivation. The passivation step was for a one-hour period and included bubbling air into the solution. The surfaces were all metallic gray and surface post-rusting did not occur. A small amount of grease in the bottom of one test section was not removed during the process.

### 3. Gas Atmosphere Studies

Hydriding of Zircaloy-2 with Thick Oxide Films. In a previous long term exposure of Zircaloy-2 to a helium atmosphere contaminated with 3% H<sub>2</sub>, 2% CO, and 0.05% H<sub>2</sub>O vapor, it was found that gas phase hydriding began after 125 days at 425 C but was not observed even after 330 days at 375 C. The shift from protective to non-protective films at 425 C was attributed to extensive cracking and thick film growth which resulted from the high corrosion rates at 425 C.

To test this hypothesis additional Zircaloy-2 samples were pre-filmed in oxygen for 56 days at 500 C which resulted in a 250 mg/dm film. The samples were then transferred to the helium experiment along with coupons with a three-day autoclave (400 C, 150 psi steam)film. After 37 days in the contaminated helium the thick film samples had hydrided, absorbing 300 ppm hydrogen at 425 C and 131 ppm at 375 C compared to 5 ppm at 425 C and 0 ppm at 375 C for the coupons with a thin three-day autoclave film.

It appears that the thick adherent oxide film is acting as a semi-permeable membrane in which the light H<sub>2</sub> molecule diffuses more readily than the heavier H<sub>2</sub>O molecule resulting in local depletion of oxidant at the metal surface. The fact that weight gains for the thick samples were only half that of the thin autoclave samples also suggests local depletion of water.

The films formed by accelerated oxidation may not be entirely typical of low temperature filming. The initial results indicate that the

effect of thick films must be carefully investigated because there may be an upper weight gain limit at which a given water concentration is an effective inhibitor to gas phase hydriding.

Continuation of the exposure of Zircaloy-2 coupons to dry mixtures of 1% CO<sub>2</sub> and 2% H<sub>2</sub> in helium for 28 days has resulted in hydriding of only the vapor blasted samples, whereas etched or preautoclaved samples have not hydrided. The results also suggest the hydriding rate of the vapor blasted samples is decreasing. This experiment is continuing.

Resistance Measurement Capsule. Preliminary laboratory testing has been completed on a capsule designed to measure the electrical resistance of zirconium oxide films. The results will be compared with data obtained from in-reactor testing of the capsule. The capsule was scheduled for in-reactor charging about March 27, 1963.

Graphite Burnout Monitoring. New burnout monitors were charged in channel 3461 at B-Reactor. Results for the monitoring period from October 28, 1962 to February 21, 1963, showed a maximum burnout rate of 4% per 1000 operating days. The profile was characterized by a sharp peak appearing at the usual distance of 80 inches into the graphite stack.

Channels 0373 and 1366, which were assigned temporarily to the burnout monitoring program, were not recharged.

It has been postulated that some of the high burnout rates observed on small monitoring samples in the Hanford reactors is the result of air leakage into the reactor stack. The location of peak burnout rates forward of the region of the stack having the highest temperature suggests a depletion of the oxygen species in traversing the burnout channel.

A theoretical model based on a combination of radiation and thermal activation with oxygen depletion has been developed and data reported for D-Reactor (HW-71751) have been used to derive constants for the model. A good fit to the experimental data was obtained by proper selection of the constants. The inlet side of the burnout peak yielded an activation energy of 15.8 kcal/mole for the radiation-activated rate. A thermal-rate expression derived previously was used (see HW-75072). The experimental data indicate only a very slight decrease in oxygen pressure in going through the first few feet of graphite where the thermal reaction is relatively unimportant. At temperatures where the thermal reaction becomes important this model indicates that the pressure of the oxygen decreases

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according to the expression:

$$\log p_{O_2} = 3.91 - 1.21x \quad (6 \leq x \leq 14)$$

where  $p_{O_2}$  is expressed in mm and  $x$  is given in feet.

The complete expression for the rate in units of percent per 1000 operating days is:

$$R = (5.26 \times 10^7 e^{-15,800/RT} + 1.34 \times 10^{17} e^{-50,000/RT}) p_{O_2}^{\frac{1}{2}}$$

The reported average inlet oxygen concentration during this particular burnout test was 0.09%; a value of 0.1% was used in fitting the data.

This equation will be tested on other burnout data as information is accumulated.

#### 4. Process Tube Development

Stress-Strain Tests of N-Reactor Pressure Tubes. Room temperature and 300 C stress-strain curves have been plotted for sections of 30% cold worked N-Reactor tubing pressurized to produce a state of bi-axial stress. Strain measurements were made independently with the optical read-out extensometer and with an electrical resistance strain gage. The former instrument measures the elongation of the entire circumference of the specimen while the gage length of the latter is limited to  $\frac{1}{4}$  to one inch. The two systems agreed within less than 1% on both proportional limit and 0.2% yield strength at room temperature. The elevated temperature strain gage data have not yet been analyzed so no comparison of the two systems can be made for that condition.

Readings were taken with the optical extensometer to extensions as high as 0.070 in/in. This is five times the usable range of the electric strain gage. The optical device has no real limit to its range and, therefore, will be extremely useful in measuring the large strains inherent in stress rupture tests.

Proportional limit, 0.2% yield strength, maximum stress, and strain at maximum stress were obtained at room temperature and 300 C on N-Reactor process tubing for the first time.

Project CAH-922 Irradiated Burst Test Facility. Construction was started on this project by the CPFF contractor (J. A. Jones) on

1103029

February 27, 1963. The excavations for the building, sanitary sewer, contaminated sewer and the new load lugger ramp have been made. The building foundation has been opened for the transfer mechanism penetration.

#### 5. Thermal Hydraulic Studies

Present Reactor Studies. Flow characteristics were determined for aluminum support pieces to be placed in empty tubes at K-Reactor to decrease the flow rate and thus prevent cavitation in the inlet fittings. Two charges were considered; one consisted of eight solid aluminum and two thin wall perfs (front to rear) and the other of 12 solid aluminum and two thin wall perfs. The process tube and hydraulic fittings were standard for a K-Reactor process channel.

It was found that flow rates would be 100 and 94.2 gpm for the eight- and 12-piece charges at a front header pressure of 500 psig. At a front header pressure of 400 psig, the flow rates would be 87 and 82.5 gpm for the eight- and 12-piece charges. Since the flow rates at which cavitation could be expected are 107 and 96 gpm at respective front header pressures of 500 and 400 psig, it was concluded that either support charge would be of sufficient resistance to lower the flow rate in an empty tube below the cavitation point.

N-Reactor Studies. A revision was made to the computer program used to reduce experimental data obtained during laboratory heat transfer experiments. The program calculates and lists significant engineering parameters (e.g., heat fluxes, flow rates, pressures, coolant conditions at various locations, and test section temperatures) using punched paper tape from the Thermal Hydraulics Laboratory data logging systems as input. Cases of liquid or two-phase mixtures (as provided by a preheater) at the test section inlet can be handled. Checks and options are built into the program to allow a variety of cases and experimental conditions to be handled, and to eliminate or replace questionable items of data on the input tapes. The program will handle data for the following types of electrically heated test sections:

- (1) Heated, internally cooled, single tube;
- (2) Tube-in-tube (annular flow) sections, with either the inner, outer, or both tubes heated.

The program is designated THOR-2 and is written for the 7090 computer.

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Run plans were developed for burnout experiments with a 32-inch long model of the annulus between the outer fuel element surface and the process tube. The electrically heated inner tube of this test section is eccentric toward the bottom of the "process tube", for a length of 23 inches, to approximate the case in which self-supports of one fuel element were sheared off, leaving only the weld tabs intact. The range of experimental conditions were chosen to allow comparisons with results obtained with similar, concentric, test sections. These experiments will complete the short-test-section burnout program for present N-Reactor fuel elements.

#### 6. Shielding Studies

Shielding studies were reactivated. A program evaluation is being made. One immediate goal is the re-establishment of shielding experimentations. As initial steps taken to accomplish this, the gamma spectrometer is being reconditioned for temporary service at the PRTR, and the No. 1 automatic sample changer is being restored to working condition. First experiments may be on barytes, iron serpentine, and borated concretes.

A sound theoretical program is also planned. Proposed areas of investigation include development of computer codes and design methods for biological shields which are suitable for an architect-engineer's use; exact-type analytical spectral and penetration analysis in biological shield materials (especially concretes); analysis of shields for fast reactors; and instrumentation for neutron spectral analysis.

#### 7. Graphite Studies

N-Reactor Graphite Irradiations. The two second-generation capsules, H-5-2 and H-6-2, in the series of long-term irradiations of N-Reactor graphite continue to operate satisfactorily in the GETR with all sample temperatures properly monitored.

Construction is nearly complete on the first third-generation capsule in the series, H-4-3. This capsule contains 20 previously irradiated samples, 11 of which have been in two previous capsules. A slight modification was made to the fifth sample position in which the four new samples are located. A pyrolytic graphite insulator was placed between the sample holder and the molybdenum spacer in an attempt to raise the irradiation temperature. The H-4-3 capsule will be installed in the GETR during the first week in April.

1103031

Measurements are continuing on the samples from the H-4-2 capsule. Length measurements were obtained on all samples including the four with broken ends. No further explanation is available on the cooling-ring displacement mentioned last month. Calibration of the three thermocouples from H-4-3 is being rerun due to large scatter in the results.

B. WEAPONS - O3 PROGRAM

Research and development in the field of plutonium metallurgy continued in support of the Hanford 234-5 Building Operations and weapons development programs of the University of California Lawrence Radiation Laboratory (Project Whitney). Details of these activities are reported separately via distribution lists appropriate to weapons development work.

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C. REACTOR DEVELOPMENT - O4 PROGRAM1. Plutonium Recycle ProgramFuels Development

PRTR Fuel Element Inspection. An irradiated Al-Pu, PRTR Mk-I spike fuel element (5088) that had been discharged from the PRTR as a suspected leaker was examined in the Fuel Element Examination Facility (FEEF) to more closely assess the condition of the element and possibly to locate the defect. The general appearance of the rods is good and no defects were observed. A loose spiral wire wrap on one of the fuel rods is an indication of mechanical core-clad interaction which has shortened the rod about 5/32-inch. The element will be disassembled and each rod individually inspected to locate the source of activity released during stagnant water tests in the PRTR storage basin.

As previously reported, the center support stem was accidentally twisted from the top end bracket gussets of a UO<sub>2</sub>-PuO<sub>2</sub> Mk-I element (5120) during manipulations in the FEEF. Subsequent tests of new Zircaloy end brackets showed that strength in tension was more than three times that for which they were designed, but that only moderate torsional forces were required to break the stems from the gussets in a manner similar to the failure of FE 5120. Metallographic examination of the failed, irradiated end brackets revealed no gross hydride. Hydrogen concentration was normal for autoclaved Zircaloy. Slightly increased hardness in the immediate vicinity of the fracture indicated a small amount of cold work. No metallurgical weakness was detected. The end bracket design allows failure in a component not containing fuel when movement of the element is restrained during twisting of the stem.

A transverse cross-section of a swage compacted UO<sub>2</sub>-PuO<sub>2</sub> PRTR fuel rod revealed that localized core temperatures resulting from PuO<sub>2</sub> segregation were sufficient to cause center void formation and columnar grain growth in a limited region. Previous examinations of incrementally loaded and swaged UO<sub>2</sub>-PuO<sub>2</sub> fuel rods operated at PRTR conditions revealed only low order sintering and equiaxed grain growth in PuO<sub>2</sub>-rich areas. The fuel rod, loaded with 157 increments, was included in an element (5096) that generated 1139 kw maximum power during accumulation of an average exposure of 300 MWD/T. A large population of lenticular voids in the columnar grains adjacent to the center void indicate that the center void and columnar grains were formed near the minimum temperature at which the phenomenon occurs. Had temperatures been higher, the columnar grains would be free of porosity. An unidentified, white appearing second

phase is dispersed throughout the  $UO_2$ - $PuO_2$ . There is no evidence of abnormal core-clad reaction in the region of increased  $PuO_2$  concentrations or of localized discoloration indicative of "hot spots" on the external cladding surface.

Destructive examination of a vibrationally compacted  $UO_2$ - $PuO_2$ , PRTR Mk-I fuel element (5113) that was discharged from PRTR as a suspected leaker revealed the first observed cladding failure of a standard PRTR fuel element. A hole, approximately 3/16-inch by 3/32-inch, was found 36½ inches from the top of a rod of the six-rod ring. Only a small amount of fuel washout and contamination release occurred, and there was no evidence of waterlogging as a result of three pressure and two power cycles after a leak was suspected. The average exposure of the element is approximately 370 MWD/T and maximum recorded heat generation rate is 1140 kw. An autoradiograph of the irradiated rod shows no localized high  $PuO_2$  concentration in the failure region. There is no evidence of excessive cladding temperature, or of association of the defect with the rod wire wraps or with fretting corrosion. The cladding has erupted slightly around the hole and the fracture surface appears brittle. Short radial cracks lead away from the hole. These observations suggest local hydriding of the cladding.

PRTR Fuel Element Fabrication. Four, swage compacted, mixed oxide, PRTR fuel assemblies were fabricated to meet April refueling needs. The method of attaching wire wraps to swaged fuel rods was modified to be identical to the method used on vibrationally compacted rods. A hole is drilled through each end cap, and the wire is inserted through the hole and spot welded on the opposite side. The new method was made possible by close length control,  $\pm 1/16$ ", achieved in swaging.

Two physics flux monitor fuel assemblies were fabricated for irradiation in the PRTR. The flux monitor assemblies contain three fuel rods with  $UO_2$ -0.48 w/o  $PuO_2$  pellets and 16 standard  $UO_2$ - $PuO_2$  vibrationally compacted fuel rods; the wire wraps are a Zr-Co alloy and the  $PuO_2$  contained 17 w/o Pu-240.

The gamma scan test unit for nondestructive determination of plutonium distribution in a fuel rod was set up in the 308 Building. Modifications were completed on the test bed and movable crystal head. Equipment checkout test scans continue to correlate with autoradiographs. Pellet standards, varying from 0.01% to 10%  $PuO_2$ , are being fabricated for quantitative calibration of the unit.

An engineering review of 308 Building autoclave facilities was started. Operating limits were established for those autoclaves in service, and equipment changes were initiated where necessary to assure that the facility meets HAPO and ASME codes. Calibrating or updating all instrumentation is in progress, and maintenance schedules and operating procedures are being prepared for each system.

Low Temperature Sintering of UO<sub>2</sub>. Ceramographic examination of a sample of PRTR cold swaged UO<sub>2</sub> fuel rod heated in the laboratory for more than 1000 hours at 550 C revealed no bonding between UO<sub>2</sub> particles. This supports the hypothesis that low temperature sintering observed in irradiated PRTR fuel rods is an irradiation effect.

Mark II-C Tubular Fuel Element. The PRTR nested tubular fuel element (1501) was destructively examined after being successfully irradiated to an exposure of 1360 MWD/Tj. Irradiation was terminated to measure the released sorbed and fission product gases from the fuel. Recent experiments at GE-APED indicate much greater quantities of sorbed gases are present in commercially fused UO<sub>2</sub> than indicated by conventional analytical techniques.

The fuel element was remotely disassembled in the PRTR storage basin. Gas pressures measured on each of the three components were less than expected.

Remote Fabrication Studies. Shakedown of the new manipulator system in the remote fabrication area was nearly completed. Operation of the system appears to be satisfactory.

Fuel Element Rejuvenation. The rejuvenation test fuel element (GEA 4-81), now cooled 60 days of the planned 90-day cooling time, was checked and gave a radiation reading of  $\sim 2 \times 10^5$  r/hr at one meter.

Repadding of PRTR Fuel Elements. Three irradiated mixed oxide Mk-I fuel elements were repadded in the PRTR basin. Twelve elements have been repadded and released for further irradiation during the last three months. Installation tools and clip-on wear pads were fabricated for repadding 13 UO<sub>2</sub> fuel elements selected for extended life tests.

Fretting Corrosion Studies. Preliminary work was initiated to provide an instrumented PRTR Mk-I fuel element to be used in determining how relative rod movement affects fretting in the Edel-1

loop. The 12 outer fuel rods will each contain a coil. Coils will be spaced in the element to provide signals at three different levels. Relative rod movement will be obtained by eddy current techniques. The coils will be driven by a 100 KC/sec oscillator. It is expected that the fuel element will be completed by May.

Fuel Element Design Studies. Fuel element end brackets having ring contact surfaces to alleviate fretting and wear are being tested on a  $UO_2$ , PRFR Mk-I fuel element in the TF-7 facility at PRFR coolant conditions. Only superficial wear occurred on one area of the 3/8-inch wide contact ring on the bottom bracket during two weeks of operation with a superimposed vibration on the test section. The oxide film was not removed from the inner surface of the Zircaloy simulated process tube, and no wear occurred on the contact area of the top end bracket. The test is continuing.

Fuel Refurbishing. Small hand tools, including pliers, a socket wrench, and a hole saw, were built to operate on 9-foot extension rods and tested in a tank constructed for testing underwater fuel element disassembly and reassembly. Underwater assembly of a newly designed replacement fuel rod in a 19-rod cluster fuel element was successfully completed.

Irradiation of Impacted  $UO_2$ - $PuO_2$ . Fuel capsules for the first irradiation test of impacted  $UO_2$ - $PuO_2$  were fabricated and shipped to NRTS. Fuel in the 0.505 inch ID by 3½ inch long capsules comprises HERI  $UO_2$  - 2.5 w/o  $PuO_2$  vibrationally compacted to a relatively low density (~ 80% TD). The capsules will be irradiated to generate surface heat fluxes of ~ 800,000 Btu/hr-ft<sup>2</sup>.

Irradiation Performance of  $MgO$ - $PuO_2$  and  $ZrO_2$ - $PuO_2$  Fuels. Ceramographic examination of irradiated  $MgO$ -3.05 w/o  $PuO_2$  pellets showed that the insoluble  $PuO_2$  was rejected from columnar grains near the thermal centers of the specimens.  $PuO_2$  was concentrated at the boundary between large and small columnar grains. This radial movement of  $PuO_2$  and/or fission products was previously detected by high resolution autoradiography.

$ThO_2$ - $PuO_2$  Studies.  $ThO_2$ - $PuO_2$  pellets (95-96% theoretical) containing 2 and 5 w/o  $PuO_2$  were completed for irradiation testing. Excellent pellet quality was obtained by dry pressing 900 C calcined powder and sintering in hydrogen at 1600 C for six hours.

Phoenix Fuel Experiments. P-3 physics calculations were begun to obtain more precise estimates of the heat generation in fuel elements being designed to demonstrate the stability of  $PuO_2$  cermets. Pre-

liminary calculations indicate that the high concentrations of plutonium in the fuel elements provide considerably more self-shielding than had been anticipated.

Particle Size Distribution for Vibrational Compaction. The optimum particle size distribution for vibrational compaction of a 1-3/8 inch OD x 1/4 inch ID test capsule was calculated from a mathematical model developed by Applied Mathematics. In spite of the odd geometry of this capsule, a very high density (93% TD) was achieved. This result, in conjunction with previous results on more conventional geometries, indicates that fuel elements of novel geometries can be fabricated without extended empirical determination of optimum particle size distributions.

Supporting Physics Studies. A number of calculations have been completed to obtain power distributions for proposed fuel element tests. Several of these designs involve materials and dimensions for which the P-3 calculation breaks down because of numerical difficulties. A cylindrical S-4 cell code has been compiled to treat these problems. Although the S-4 solution is slightly more time consuming than the P-3 code, it appears to give comparable precision. A slab geometry version of the same code is being compiled to treat the enrichment banding problem in mixed oxide fuels.

#### Corrosion and Water Quality Studies

Pitting Corrosion of Mild Steel Pipe. Selected sections of the PRTR secondary and process water systems have been radiographed to determine the extent of the pitting corrosion which caused failure of a section of the chemical tank mild steel fill line. All dead legs which were radiographed showed pitting with pit depths up to 0.085 inch. These pits are located in an area eight inches to 30 inches from the high flow piping. Radiographs of pipes containing high flow water showed no pitting. No low flow piping in these systems has as yet been radiographed. The observed dead leg pitting may result from a differential aeration cell.

Removal of Contamination and Films From Pressurized Water Systems. As previously reported, the AFACE procedure was ineffective in removing the films and activities from either the PRTR system or

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Wyandotte 5231 was of little value. Similarly, two-step procedures using alkaline permanganate followed by phosphoric acid (Turco 4512) or bisulfate (Wyandotte 5061) gave very low decontamination factors, less than 1.5. However, two-step procedures using alkaline permanganate followed by either sulfamic acid or oxalic acid gave decontamination factors ranging from 5 to 20. Both solutions prepared by mixing laboratory chemicals and proprietary solutions were effective. In these preliminary tests the best results were obtained using alkaline permanganate followed by an inhibited sulfamic acid (1.0 M sulfamic acid, 1.0 g/l proprietary inhibitor) for two hours at 70 C.

Additional tests are under way to evaluate effectiveness of ammonium citrate at higher temperatures in more dilute concentrations.

Fretting Corrosion of PRTR Fuel Elements in Ex-Reactor Loop. A PRTR fuel element with modified end brackets to permit a full 360° contact surface between the fuel element and pressure tube is being exposed to 530 F, pH 10 water in TF-7 Loop. Vibration is induced on the test section to promote fretting between the fuel element and pressure tube. After two one-week operating periods, fretting has been negligible. The oxide was found to be worn from the bottom fuel element bracket support over an area approximately 1" x ½". No metal penetration or oxide removal was found on the pressure tube. Based on the limited test information to date, the addition of the increased contact area between the fuel element and pressure tube appears to be a satisfactory method of reducing fretting corrosion.

Monitoring Zirconium Concentrations in PRTR. In last month's report, the zirconium concentrations in PRTR primary system water with operation of different pump combinations were reported. When the D<sub>2</sub>O was recirculated at high speed with pumps 2 and 3, audible vibration was reported. At this time (February 10 and 11), the concentrations of zirconium in the coolant were high, greater than 10 ppb. When a different combination of pumps was used, the audible signal disappeared and the zirconium concentrations decreased.

During March additional data were obtained on zirconium concentration levels in the PRTR primary coolant. A special test was conducted prior to a reactor startup to investigate the effects of different pump combinations at full-flow conditions. During this test samples were collected at approximately one-hour intervals to determine how rapidly the concentration might vary. Samples were also collected to determine the efficiency of the cleanup ion exchanger for removing zirconium from the coolant. Sample analysis did not indicate that any particular pump combination caused an appreciable increase in the

zirconium concentration, contrary to the earlier tests. It should be noted, however, that combinations of pumps 2 and 3, and 1 and 2 were only employed for about four hours each, and it is possible that this period was insufficient.

The cleanup ion exchanger was not effective in removing the zirconium from the coolant. Either the zirconium was present in a form that was not removed by ion exchange, or the resin was saturated with zirconium.

When the reactor resumed operation, pumps 1 and 3 were in service to circulate the coolant. There was a 32-hour period on March 8 and 9 when the zirconium concentration was consistently above 5 parts per billion. During operation concentrations greater than 10 ppb were observed on March 4 and 5. Samples taken during these periods (March 4 and 5) were rechecked at 8.6 and < 0.2 ppb, respectively. The marked reduction observed on the sample from 3/5/63 indicates that precipitated particulate zirconium may be present. The analytical procedure will be modified to dissolve any particulate zirconium present to evaluate the reproducibility of the procedure. Tests will also be continued to evaluate other procedures for measuring zirconium concentrations.

If the zirconium cannot be removed in the cleanup system, then redeposition is the mechanism controlling reduction of the coolant zirconium concentration. Under these conditions it is impossible to calculate actual zirconium release rates from concentration measurements in the coolant since the redeposition rate is unknown. Thus, at best, measurement of zirconium concentrations could only be used to detect the onset and cessation of serious fretting corrosion and could not be used to calculate the amount of fretting attack.

#### Reactor Components Development

Properties of Irradiated PRTR Process Tubes. The investigation of tube 5679 is continuing. This tube was removed from channel 1643 because it contained an ID flaw which was detected during in-reactor monitoring in May 1962. Burst tests of three, 2-foot sections of this tube have been performed. The test temperatures and calculated ultimate tensile strengths have been reported, as has the information that the flaw had no effect on the burst strength.

Metallographic examination of samples at and around the flaw location has revealed abnormal amounts of a second phase more evident in a transverse than in a longitudinal section. A phase segregation

has appeared in some portion of nearly all irradiated Zircaloy-2 pressure tubes examined. The second phase does not appear to reduce tube strength.

Measurements of the thinned wall and the stretched circumference of burst specimen 4B of tube 5679 have been made. They show that the hoop elongation and the wall thinning at the burst were between 30 and 40%.

The search for hydride in tube 0720 in the region of the MgO-PuO<sub>2</sub> fuel element failure has revealed a localized concentration of about 50 to 75 ppm. Estimates of the closeness of the hydrided area and the rupture were made. They indicate that longitudinal separation could have been as much as one inch; however, a record of the angular locations was not kept.

Burst test specimens from tubes 0720, 5540, and 5702 were prepared by PRTR personnel for shipment to Radiometallurgy. These specimens will be burst-tested at elevated temperature in the remote burst-test equipment at Radiometallurgy.

Tube 0702 will go in the PRTR in the near future as replacement for tube 5675 which will be discharged for burst-testing. The dimensions of tube 0702 were measured to provide for more accuracy in post-irradiation dimension dependent measurements. Specifically, the over-all length, the length of the small diameter section at the inlet end, the distance from the inlet end to the large end of the taper, and the OD at nine points along the large diameter section were measured.

All pictures of irradiated burst-test specimens were collected and arranged in a notebook. These pictures show that the majority of the bursts occurred at about one-quarter of the length from mid-length. A possible reason for this is that the amount of fast neutron exposure varied significantly from one end to the other and the end having the lower exposure would have the lower ultimate tensile strength. One of these specimens had a very small variation end-to-end in neutron exposure because it came from the location of the flux peak. It burst at mid-length.

Pressure Tube Monitoring. Eight process tubes were examined during the past month. Six of the eight channels operated with UO<sub>2</sub> elements since PRTR decontamination. Four of the eight tubes were believed to exhibit substantial vibration. Of the four tubes suspected to be vibrating, two operated with UO<sub>2</sub> elements and two with mixed oxide elements, one of which had been repadded. For the channels operating

with UO<sub>2</sub> elements, all tubes excepting one were in general found to have a number of new fretting corrosion marks formed since the June inspection of 1962. The bulk of the new fretting marks are the result of the spiral 19-rod bundle wrap wire contacting the inner surface of the process tubes. Of the tubes suspected to be vibrating, those which operated with UO<sub>2</sub> elements appear to have incurred more fretting corrosion than those tubes which operated with mixed oxide elements. There were no clear-cut differences in the amounts of fretting corrosion of suspected vibrating and non-vibrating tubes. More definitive vibration measurements are needed to determine whether or not there are gross differences in the magnitudes of vibration from one tube to any other in the PRTR. During this inspection, ID and gas gap were also measured. No deviations from operating specifications were found.

A radiation resistant television camera has been considered for in-reactor visual inspection of the PRTR Zircaloy-2 pressure tubes. Measurement of electronic properties of a special TV camera image tube (vidicon) with a radiation resistant faceplate showed no change after  $1.1 \times 10^9$  R accumulated gamma irradiation. This substantiates previous results in which no degradation in visual television picture quality was noted after this irradiation. This is the highest known successful irradiation of a television vidicon tube.

Second Generation Mechanical Shim Rod for PRTR. Detailed design of the driving head is complete except for the final assembly drawings and the heat sink details. Fabrication of the parts for the driving head is estimated 70% complete.

The selsyn position readout equipment was received. The initial order for this equipment was based on equipment for only one rod. Since it is desired to carry on out-of-reactor testing simultaneous with in-reactor testing and since the position readout equipment received appeared to be of high quality, a requisition was placed for additional units for the second rod assembly.

A satisfactory proposal was received for rotary type limit switches for the driving head. A requisition was placed for sufficient switches for two rod assemblies.

A requisition was placed for a set of zirconium lead screws to be evaluated along with the presently used aluminum lead screws. It is believed possible that zirconium screws might increase the reliability of the rods, although at a considerable increase in cost.

Shim Rod Environmental Test Facility. Fabrication and installation of the shim rod environmental test facility are complete. The facility has been operated satisfactorily for several hours at design conditions.

A new shim rod, procured as part of an order for 10 new spares, has been received and will be installed and tested in this facility.

EDEL-I Renovation. Vibration checks on the rebalanced pump-motor assembly indicated that the unit is now operable although the vibration still exceeds desirable limits.

Fabrication was completed and the system hydrostatically tested at 3000 psig in the presence of the third party inspector. A drawing identifying all welds and material certifications is being prepared. The loop is now ready for high temperature shake-down tests.

Fretting Corrosion Investigation. Delivery date for the oscillographic recorder to be used with the vibration and flow and pressure oscillation instruments is now May 10, 1963. The amplifiers originally intended to be used with the recorder have now become unavailable on a loan basis. Satisfactory amplifiers are currently being ordered on an emergency basis.

Fabrication of the fuel element containing the eddy current position indicating instrumentation will begin in approximately three weeks. Work is currently delayed pending receipt of the lead wire to be used with the eddy current coils.

PRTR Rupture Loop Components. A test to check leak rate and stud stresses for the rupture loop outlet jumper connection was attempted for the second time in EDEL-II. Prior to assembly, the connector hubs were round within 0.001 inch. High leakage was observed during the first temperature cycle. When the connector was disassembled, one hub was found to be oval by 0.009 inch and the other by 0.004 inch.

Investigation of the fit of the hubs and clamps revealed that the spacing between the clamp and one hub was gauged to be about 0.020 inch larger at the two clamp ends than at the center. The clearance on the other hub was approximately the same. This spacing should have been constant. Numerous checks and tests have been made on these deformed parts and the following facts are now apparent.

1. Two out of three connections tested by Equipment Development have leaked quite badly.
2. Contact between hub and clamp is confined to the center portions of the clamp on arc lines approximately  $22^\circ$  to either side of the center.
3. Line contact for the proper  $180^\circ$  of arc is not obtained until a total of 0.025 inch of shims is placed between the two hubs.
4. Connector parts conform to Gray Tool Company drawings.
5. Good performance has been obtained at Hanford and at Gray Tool on connectors of similar stainless materials.

At the present, the cause of this problem appears to be a design inadequacy relating to geometry and fit of the parts.

Initial contact has been made with Gray Tool Company and followup contacts are planned in an attempt to resolve this problem.

PRTR Pressure Tube Seals. Design is 50% complete on a three station pressure tube seal test assembly to be installed on EDEL-I. A small canned rotor pump and heat exchanger have been obtained to be used to circulate cool water through the test assemblies for the cool-off portion of the thermal cycle tests.

PRTR Gas Loop Components. Fabrication of the pressure tube-shroud tube gap detector assembly was initiated. Cleanup work on various tools continued.

#### Hazards Analysis

Hazards Reviews. The results of two PRTR nuclear safety analyses were presented to the Plutonium and Fast Reactors Subcouncil at the 10th meeting of the General Electric Company Technological Hazards Council. The subjects reviewed were proposed changes in PRTR operating limits and convection cooling of the PRTR following a total electrical power failure.

The Council accepted as consistent with reactor safety considerations the increased heat transfer flux limit, 650,000 Btu/(hr)(ft<sup>2</sup>), and tube power limit, 1800 kw. Previous limits were 400,000 Btu/(hr)(ft<sup>2</sup>) and 1200 kw. Review of convection cooling in the PRTR resulted in the conclusion that convection cooling can adequately

cool the fuel elements during a total electrical power failure provided that operating personnel perform the necessary operations at approximately the correct times and that the pressurizer relief valves open properly.

AEC approval has been received to operate the PRTR Fuel Element Rupture Testing Facility in accordance with the provisions of the safeguards analysis for this facility, "PRTR Final Safeguards Analysis, Supplement 5, Fuel Element Rupture Testing Facility," HW-61236 SUP 5.

Loss of Coolant Study. The effect of positive void coefficients on fuel temperatures in PRTR were determined in analog simulations of reactor transients. In these analyses it was assumed that the reactor was operating at 70 Mw, 87.5% of the coolant was lost in 0.1 second, and the safety circuit was tripped at 91 Mw. Credit was taken, also, for the negative fuel temperature coefficient in the studies. For various possible values of the coolant void coefficient the results are tabulated below.

Coolant Void Coefficient mk	Energy Release Mw-sec	Average Fuel Temp. Increase °F
+ 5	160	60
+10	830	600
+15	1200	1800
+20	8000	3000

Review of Light Water Injection System. In conjunction with the increased tube power limit in the PRTR, reanalysis of the light water injection was initiated. A review of the probable failure temperature of Zircaloy-2 clad, ceramic core fuel elements was also made; the published data on Zircaloy-2 strength versus temperature lead to the conclusion that these elements could fail at temperatures of about 1200 to 1400 F instead of at the melting point of zirconium, about 3300 F. Even with this lower temperature of jacket failure, both the high pressure and low pressure light water injection system would be adequate.

PRCF Conversion to H<sub>2</sub>O Moderation. The mathematical model has been developed for analog simulation of reactor excursions in light water moderated PRCF cores. In these studies inherent safety will be given by the negative fuel temperature coefficient and by the moderator void coefficient. The first cores to be studied on the analog will be PuO<sub>2</sub>-UO<sub>2</sub> mixed oxide core in support of the EBWR loading.

## 2. Plutonium Ceramic Fuel Research

Plutonium Nitride. Arc melting experiments were performed as part of a study of the thermal stability of PuN. Plutonium nitride, formed by nitriding plutonium hydride, was arc-melted under one atmosphere of nitrogen on a water-cooled copper hearth. Metallographic and x-ray diffraction data show that approximately 95 v/o of this arc-melted material consists of PuN dendrites in a matrix of alpha plutonium. The PuN thermally decomposes ( $\text{PuN} \longrightarrow \text{Pu} + \frac{1}{2} \text{N}_2$ ) at the melting point. The samples had a solid PuN surface layer.

Radiation Self-Damage of Plutonium Compounds. The lattice parameter of PuO<sub>2</sub> was found to increase at the rate of  $0.35 \times 10^{-3} \Delta a/a/100$  days due to alpha self-damage at room temperature. This value was measured after 503 days of exposure.

After 120 days of self-damage, beta-Pu<sub>2</sub>O<sub>3</sub> has exhibited the following rates of increase per 100 days:

$$\begin{aligned} \Delta a/a &= 0.70 \times 10^{-3} \\ \Delta c/c &= 1.16 \times 10^{-3} \\ \frac{\Delta(c/a)}{c/a} &= 0.43 \times 10^{-3} \end{aligned}$$

## 3. Uranium Ceramic Fuel Research

Thermal Conductivity of UO<sub>2</sub>. Preliminary measurements of heat transfer through UO<sub>2</sub> show that the conductivity of single crystals (relative to polycrystals) increases above 1600 C. A radial heat flow apparatus which compares the conductivity of hollow cylindrical specimens surrounding a central tungsten heater was used for these measurements. The ratio of the conductivity of the single crystal to that of the polycrystal was 2.0 at 850 C, 1.2 at 1600 C, and 1.8 at 2000 C.

These ratios may be low if the enhanced conductivity of single crystals arises from radiant heat transfer since the transmitted energy does not heat the UO<sub>2</sub> surface and thus passes through undetected. This effect is being investigated by using specimens plated with thin, opaque coatings of tungsten. The tungsten coating is expected to prevent loss of energy radiated through the UO<sub>2</sub>.

In-Reactor Measurement of Thermoelectric Power of UO<sub>2</sub>. The thermoelectric potential of UO<sub>2</sub> was successfully measured in-reactor. Of particular interest, a reversal of polarity (from p- to n-type conductance) occurred as the central temperature of the cylindrical element increased from 1000 to 2000 C. This suggests the possibility that greater amounts of thermoelectric power can be produced by controlling the UO<sub>2</sub> composition, and hence, the conduction mechanism. An average thermal emf of 500  $\mu$  v/C was measured at a core temperature of 1000 C.

Electron Microscopy of Autoradiographic Replicas. A method was developed of replicating UO<sub>2</sub> surfaces for electron microscopy with an alpha sensitive emulsion. Excellent detail of both the replicated surface and alpha tracks originating from the surface was observed at 40,000X. The direction the alpha particle travels, and hence the point of origin, can be determined by the spacing of the silver grains in the emulsion. Higher magnifications (up to 300,000X) were used to study detail of the alpha tracks.

Electron Microscopy of Thinned UO<sub>2</sub> Sections. Chemical thinning was successfully used to produce thin sections of UO<sub>2</sub> crystals for electron microscopy. The thinning is accomplished by directing a hot phosphoric acid jet on the sample. (The apparatus is based on that designed by Dr. Amelinckx at Mol). Thinned areas have averaged about 400 square microns and are expected to increase in size with improvements in the technique.

Sections thinned from sawed wafers of UO<sub>2</sub> crystals show a complex network of mobile dislocations and extinction contours. Thinned natural cleavage platelets showed little dislocation structure initially but developed an immobile network upon prolonged exposure to the electron beam. Additional thinning of the cleavage platelets occurred during examination, each break-through hole displaying a similar hexagonal shape.

UO<sub>2</sub> Microcores. UO<sub>2</sub> microcores, approximately 0.015 inch OD x 0.085 inch long, were extracted from UO<sub>2</sub> specimens using a special, small core drill, and ultrasonic drilling techniques. Specimens were taken from UO<sub>2</sub> single crystals, impact formed UO<sub>2</sub> and sintered UO<sub>2</sub>. These small cores allow precise sampling of fuel materials for fundamental studies of fission product migration and analyses of impurity phases.

Impaction of UO<sub>2</sub> Scrap to Ultra-High Density. The high densities (99.0% TD) reported last month for UO<sub>2</sub> impacted with a 4-inch diameter tool steel punch at an unknown impact pressure, have been

shown to be the result of higher impact pressures than previously believed possible using tool steel punches. These results have been duplicated using calibrated 4-inch and 2.5-inch diameter tool steel punches, at measured impact pressures of 250,000 psi.

Impacted densities approximately 0.4% higher were obtained using pulverized sintered scrap roasted to a composition of  $UO_{2.01}$ , compared to densities obtained using pulverized sintered scrap without roasting. Mechanical strength of the material impacted from roasted  $UO_2$  is greater than that of material impacted from unroasted scrap, indicating better inter-particle bonding.

Electron Microscopy of UN-W Cermets. A UN-W cermet was observed by reflection electron microscopy during controlled electron beam heating, and while it was being subjected to a low-pressure oxygen jet. Severe thermal etching occurred, especially in the UN, at surface temperatures of approximately 1000 C. Introduction of oxygen at that temperature caused rapid oxidation of both UN and W surfaces, but UN-W interaction did not appear to be appreciable, and the surface retained its general two-phase character. Investigation is continuing with other gases and other uranium cermets.

Fabrication of Cermets. A variety of powdered metals has been procured for impaction of uranium and plutonium cermets. A technique has been developed for fabricating 4-inch diameter solid cermet discs approximately 1-inch thick by impaction. Tungsten- $UO_2$  cermets completely encased in and bonded to tungsten cladding have been formed.

Magnetic Force Resistance Welding Studies. Two pieces of tungsten were joined successfully without the occurrence of the large recrystallization grains that normally appear in the weld zone. The rapidity with which the bonding takes place limits recrystallization to an extremely narrow band in the weld area. The recrystallization grains that do form are smaller than the grains of the parent metal, indicating the joint may be stronger than the parent material.

The first successful joining of tungsten- $UO_2$  cermet material was achieved. No  $UO_2$  vaporization occurred during the bonding of the pieces because of the extremely short welding time and high current density used to produce the bond. The pressure applied during welding produced slight cracking in the parent material, but this can be eliminated.

Preparation and Properties of  $UOS$ . A method was developed for preparation of  $UOS$  (uranium oxysulfide) by deposition from a molten

salt solution.  $US_2$  and  $UOS-US_2$  mixtures have also been prepared. The melting point of UCS prepared by this method was  $1880 \pm 50$  C. After solidification, two unidentified phases were present in the UCS.

UO<sub>2</sub> for Savannah River Laboratory. UO<sub>2</sub> materials are exchanged with other sites for evaluation and information exchange. In response to a request from SRL, a 50-pound lot of (-12 +20) mesh arc-fused UO<sub>2</sub> for HWCTR testing was treated at high temperature in moist hydrogen to reduce the impurity content, particularly that of the nitride. Preliminary data indicate water content approximately 5 ppm, nitrogen content less than 100 ppm, and density greater than 99% TD.

Basic Materials for Research. Standardized uranium ceramic specimens were prepared for other sites, including Tokai (Japan), CANEL, LASL, ORNL, and NMPO.

HGSR Fuel Element Study. The research and development effort required to produce a fuel element meeting the functional requirements outlined in HW-73130, "Power Reactor Design Study and Evaluation - Hanford Graphite Superheat Reactor (HGSR)," is being evaluated at the request of Reactor Design Analysis, N-Reactor Department. The proposed fuel elements are 18 or 26 feet long, re-entrant, and include both boiling and superheating heat transfer surfaces. Problem areas include cladding material, fabrication (vibrational compaction) technology, cladding support, eccentricity, cladding and fuel creep and differential expansion, fission gas accommodation, high heat flux, corrosion, hydraulic stability and effect of operational variables.

#### 4. Basic Swelling Program

Irradiation Program. Two general swelling capsules have continued their irradiation. The reworked Minneapolis-Honeywell instrumentation is being employed to control the temperatures of these capsules. Due to an unexpectedly large amount of fission heating, some difficulty was experienced in maintaining precise control on one capsule and a momentary excursion into the beta occurred. This excursion is expected to have a small influence on the results that might otherwise have been obtained. The second capsule experienced a temperature drop when the reactor went down because of an error in the wiring of the heating power supply. This temperature drop with the reactor down is not expected to influence the behavior of the specimens. The problems have been rectified and both capsules are now operating at a constant temperature.

Two additional capsules are being constructed. These will each contain eight different uranium specimens in lieu of the six (three of which are duplicates) heretofore employed. Uranium with various additions of Fe + Al and Fe + Si will be included with the high purity uranium. Various heat treatments will also be represented to permit greater insight into the influence of metallurgical conditions on the irradiation behavior of uranium.

Post-Irradiation Examination. The uranium specimens that were recovered from two controlled temperature capsules irradiated to 0.15 a/o B.U. at 575 C are being processed for metallography. One capsule contained specimens enriched to 1.44% U-235, whereas the second contained natural (0.72% U-235) uranium specimens. The density values and the calculated volume changes are as follows: as extruded, natural uranium,  $\rho = 12.3 \text{ g/cc}$  ( $\Delta V/V = 0.54$ ); beta-quenched, natural uranium,  $\rho = 15.3 \text{ g/cc}$  ( $\Delta V/V = 0.24$ ); as extruded, 1.44% enriched uranium,  $\rho = 14.2 \text{ g/cc}$  ( $\Delta V/V = 0.33$ ); and beta-quenched 1.44% enriched uranium,  $\rho = 13-16 \text{ g/cc}$  ( $\Delta V/V = 0.43-0.15$ ).

The density values agree qualitatively with the macro-appearance of the specimens and verify the previous conclusion that the natural uranium specimens suffered slightly more damage than did the enriched uranium specimens. The density measurements on the enriched specimens were taken initially using tetrabromoethane as the immersion media and were repeated with carbon tetrachloride. The values on the as-extruded samples were the same with both fluids but the density was much higher (16.47 versus 13.26 g/cc) in  $\text{CCl}_4$  for the beta quenched specimen. This is believed to be due to the fact that better wetting of internal cracks and pores was achieved with  $\text{CCl}_4$ . The lower density value is probably more representative of the true condition of the specimen. The value obtained on the beta-quenched, natural uranium specimen also fails to indicate the magnitude of the damage that actually occurred due to probable wetting of internal cracks and pores.

Both optical and electron metallography have been completed on the enriched samples. The natural uranium specimens have yet to be completely processed. Large differences were observed between the samples. Sample A-2 (natural uranium, as extruded) had tears, pores (some of which appeared to be aligned crystallographically) and pseudo second-phase that has been detected previously. This "phase" also appears to be aligned crystallographically. No grain structure could be detected by either optical or electron metallography. Profuse porosity was observed in the electron microscope; pores

varied in size from  $\sim 1\mu$  to  $\sim 0.03\mu$ . Specimen B-2, same state as A-2, looked much more sound. Grain structure was clearly developed and was quite similar to the pre-irradiation structure. A few tears were present as was a little of the crystallographic porosity and pseudo second-phase. The general porosity appeared to be less than was the case for specimen A-2. The variation in pore sizes seemed to be about the same; however, the density of large pores in specimen B-2 is much less. The pore size density distributions will be determined with the quantitative metallographic techniques developed on this program. Specimen C-2, which had been beta-quenched prior to irradiation, varied a great deal within the transverse section -- one end was very lacy whereas the other end contained only a few tears and large holes. Appreciable amounts of pseudo-second-phase were also present. It is extremely difficult to distinguish reality from artifact in these samples. Porosity and tearing can interfere with the normal polishing and etching characteristics of the uranium, producing structures that are completely false. A constant effort is being made to improve the techniques employed.

Some large patches of the unidentified pseudo-second-phase present in specimen A-2 were removed with a micro drill and the drillings were collected with a vacuum pickup onto plastic filter paper. Several attempts were made to obtain an x-ray diffraction powder pattern from the material collected, but no trace of a pattern was obtained. It would appear that the material is amorphous. Other micro techniques will be employed to identify the elements present.

The Zircaloy-2 clad, uranium-uranium diffusion couple specimen that had been irradiated to 0.4 a/o B.U. in the enriched shell and to 0.02 a/o B.U. in the depleted core and then annealed at 950 C for 100 hours has now been reprocessed for metallography after an additional anneal at 600 C for 1000 hours. The 600 C anneal produced essentially no change in either the size or distribution of porosity in the depleted zone. The enriched zone is too porous for effective replication. No evidence for diffusion was obtained; hence, it is doubtful that the rest of the annealed specimens of this series will be examined. There are still 12 specimens representing two exposure levels that are in the irradiation capsules. It is anticipated that these will be used to study diffusion at ultra-high pressures.

Restrained Irradiations. In order to gain insight into the influence of restraint on the swelling of uranium, Zircaloy-2 clad uranium rods with selected uranium temperatures, cladding thicknesses

and exposure are being irradiated in NaK-filled temperature monitored capsules. Recent irradiations showed marked changes in the micro-structure and second-phase distribution of beta heat-treated U-2 w/o Zr fuel irradiated at temperatures of approximately 520 C. Out-of-reactor annealing studies have been started to separate the thermal and radiation effects on the phase changes. This information is important to the understanding of the role that phase transformations may have in the mechanics of swelling of U-2 w/o Zr. Additional irradiations of Zr-2 clad U-2 w/o Zr fuel in NaK capsules are now under way. The volume average temperature of these samples is approximately 475 C.

In addition to the above alloy fuel irradiations, one U-2 w/o Zr rod clad with Zr-2 of 0.025 inch nominal thickness and two Zr-2 clad unalloyed uranium rods, one with 0.025 inch nominal cladding thickness and one with 0.035 inch nominal cladding thickness, are being irradiated in NaK capsules. These three rods are each approximately six inches long and have one end open to permit possible extrusion of the fuel out the end. It is anticipated that these samples will provide qualitative data on: (1) the relative plasticity of the unalloyed and alloyed fuels under similar irradiation conditions; and (2) the relative restraint offered by 0.025 inch and 0.035 inch thick Zr-2 cladding. These samples are now at approximately one-half goal exposure.

Thorium. Two thorium specimens irradiated to 0.18 and 0.92 a/o B.U. have been processed for metallography, hardness, and density. The hardness and density values are indicated below.

Sample No.	Source of Metal	a/o B.U.	Hardness		Density, gms/cc Avg of Two Determinations in CCl <sub>4</sub>
			R <sub>B</sub>	Avg of 10 Readings	
B-2	Battelle	0.18		78 ± 1	11.67 ± 0.01
S-3	Hanford	0.92		(Center) 65 ± 2	11.50 ± 0.01
				(Edge) 73 ± 2	

Both specimens have increased appreciably in hardness as compared with the pre-irradiated hardness of about R<sub>B</sub> 13. It is interesting to note that specimen S-3 is significantly softer in the center

region than in the edge region. The reason for this is probably due to the higher operating temperature of S-3 which permitted some recovery of irradiation hardening to occur. A few small pores (0.02-0.1 $\mu$ ) located primarily at grain boundaries were observed in S-3. There was no marked difference between the edge and center regions, but these areas will be examined in greater detail to ascertain whether or not real differences in either the size or number of pores exist. The density measurement on specimen S-3 indicates a volume increase of about 1.5%. It is doubtful that the porosity observed can account for this much volume change. Since specimen S-3 contained a large crack, it is likely that this has contributed significantly to the observed density change. The density of sample B-2 was no different from unirradiated control material and, as expected, no porosity was observed with the electron microscope. These specimens will be annealed at successively higher temperatures and the above processing will be repeated.

#### 5. Irradiation Damage to Reactor Metals

Alloy Selection. Procurement of materials to be used as test specimens for the Irradiation Effects on Reactor Structural Materials Program is continuing. Received on site during the past month were approximately 18,000 pounds each of A212B and A302B pressure vessel steel plate. This material was four inches in thickness and closely duplicates the standard heat material obtained by the United States Steel Corporation for the nuclear industry.

Specifications for precipitation hardenable alloy A-286 have been prepared and a requisition for procurement of strip and rod initiated. This chromium, nickel, iron alloy has good strength and corrosion properties at high temperatures and its properties before and after irradiation will be compared with other precipitation and solution hardened alloys.

An order has been placed with Carpenter Steel Company for AISI 406 stainless steel in the form of strip and bar. In addition, bid review has been completed for quantities of plate, sheet, and bar of AISI 304, AISI 348, and AM-355. This material will be obtained from Allegheny Ludlum Corporation.

Tensile specimens of Inconel 625, R-235, and Hastelloy N have been prepared and submitted for irradiation in a cold water (175 F) irradiation facility of the ETR. Data from this irradiation test will be compared with that of other tests for specimens irradiated

at 540 F and at 1300 F in order to determine the temperature effect upon the properties of irradiated structural materials.

In-Reactor Measurements of Mechanical Properties. The test in progress is the first of a series to determine the stress dependency of Zircaloy-2 creep during irradiation. The present test is being conducted on 20% cold worked Zircaloy-2 at a temperature of 350 C and a stress of 20,000 psi. The test has accumulated 1300 hours of operation during which time three reactor outages have occurred. The creep rate during irradiation at 350 C and 20,000 psi stress has been tentatively established at  $1.7 \times 10^{-6}$  in/in/hr. The in-reactor test during a 10-day outage, which occurred shortly after charging, exhibited a rate essentially the same as that during reactor operation. The ex-reactor test has not accumulated enough hours to determine the creep rate accurately, but it appears to be about  $1.9 \times 10^{-5}$  in/in/hr. Except for the in-reactor rate during reactor outage, the creep curves follow the same general pattern established by the series of creep tests conducted at 30,000 psi stress. Other tests will be conducted to determine if this deviation from the pattern is due to stress dependency.

Development of the differential control system for the temperature control of the in-reactor capsule heaters described in the January monthly report is continuing. Construction of the creep capsule 96-point data logger is also progressing. Ninety percent of the parts have arrived and assembly work is in progress.

Negotiations were completed this month for the procurement of 20 additional creep capsules. The specifications call for 10 capsules identical to those now being used in the program and parts for an additional 10 capsules that can be tailored to particular environmental tests. The assembled capsules can be operated between temperature limits of 200 and 700 C, with stresses between 0 and 80,000 psi and strain recorded for one-half inch elongation. The unassembled capsules can be altered to extend the above limits as a particular test or environmental condition may dictate.

Tensile testing of heating element wires is progressing. The tests are being performed on irradiated and unirradiated Ni-Cr-Fe-Al, Ni-Cr, and 406 stainless steel wires to determine the cause of in-reactor heater failures and to establish a basis of selection for future in-reactor use. Grip slippage of the 0.020-inch diameter specimens has been eliminated and the techniques discussed in the January monthly report have proved satisfactory. The unirradiated samples are presently being tested. No comparisons to irradiated samples can be made at this time.

Irradiation Effects in Structural Materials. The purpose of this program is to investigate the combined effects of irradiation and reactor environment on the mechanical properties of structural materials. Special attention will be given to the determination of mechanical property changes produced in metals by irradiation at elevated temperatures.

During the month 146 specimens were tested. The specimens tested included 104 irradiated specimens, 33 control specimens exposed in the ex-reactor hot water loop, and nine specimens in the as-fabricated condition. Of the specimens tested, 93 were Zr-2, six were Zr - 2 Sn - 2 Nb, and the remainder were stainless steels of the types AISI 410, 406, 348, and AM-350. Bend tests were performed on 24 irradiated Zr-2 specimens; the remainder of the tests were performed on plate tensile specimens.

Specimens of AISI type 348 and 304 stainless which received exposures of approximately  $6 \times 10^{19}$  and  $1.8 \times 10^{20}$  nvt (fast) were given a one-hour quench in liquid nitrogen before testing at room temperature. No change in mechanical properties was noted as a result of this treatment.

One hundred tensile specimens have been fabricated from rolled sheet stock of Hastelloy X-280. Specimens oriented transverse to the rolling direction were given a variety of aging treatments after a solution anneal. Both the aging temperature and the time at temperature were varied. Specimens representing all aging conditions are to be charged in both the ETR G-7 loop and the ex-reactor hot water loop.

Six capsules containing 48 tensile specimens have been sent to the ETR to be irradiated in the C 3x3 G-6 position. Specimens irradiated in this position are exposed to reactor ambient temperature, process water and flux similar to that in the G-7 loop. Materials included in these capsules were AISI type 304, 348, 410, and AM-350 stainless steels, as well as nickel alloys R-27, R-235, Hastelloy N, Hastelloy X-280, and Inconel 625.

The G-7 loop specimen hanger assembly was fitted with six iron-constantan thermocouples for monitoring specimen temperatures during ETR Cycle 52. This was the first time specimen temperatures in the loop had been monitored directly during reactor full power operation. Results of this test indicate that specimen temperatures range from 540 F in the lowest flux positions to approximately 580 F in the higher flux positions. This is approximately 45 F higher than the loop outlet bulk water temperature when

the reactor is operating at full power. From these results a better thermal history of specimens irradiated in this loop will be obtained.

Slow bend tests on notched Zircaloy-2 beams were performed at several temperatures. The Zr-2 beams were fabricated from 23% cold worked sheet stock and had the following dimensions: gross beam depth - 0.70 inch; width - 0.46 inch; length - 6.0 inch; notch depth - 0.245 inch; root radius - 0.002 inch; and a flank angle of 45 degrees. Specimens fabricated from both the transverse and longitudinal directions with respect to rolling were tested in four-point bending.

The specimens were fatigued in a half cycle cantilever manner to produce a dead sharp fatigue crack approximately 0.06 inch deep in 5000 to 7000 cycles. The fatigue crack, which extended inward from the notch root, was delineated by heat tinting the entire specimen to a straw or light blue color. The testing was carried out at various temperature intervals from boiling liquid nitrogen to room temperature. The various temperatures were achieved by insulating the mid-section of the specimen, immersing the specimen in liquid nitrogen, and then letting it warm to the appropriate temperature for testing. The temperature at fracture was used in all cases. Iron-constantan thermocouples imbedded at the notch exit corner was chosen as the correct spot for imbedding the thermocouple head because two surfaces free from traction meet at an angle of 112.5 degrees at this point. A minimum disturbance in the remaining stress field of the beam occurs when a shallow (~ 0.0625 inch deep) hole is drilled at the notch exit corner.

A dead sharp crack was used to most closely simulate crack propagation requirements rather than crack initiation requirements. The two differ in that the former is the energy requirement to cause an increment of crack area to be formed in front of a running crack. The crack initiation energy requirements must fulfill the conditions for the formation of a crack from intersecting slip planes, slip planes and grain boundaries, included foreign matter, etc. The crack initiation requirements are much greater than propagation requirements. As a result, once a crack is formed more than enough energy is available to cause propagation. In these studies, however, the question is not, will a crack form. Rather, once a crack has formed and propagated a short distance, is the material of such a nature that the excess energy will be absorbed by plastic deformation and the crack stopped before serious failure to a structure has occurred. The dead sharp fatigue crack approximates a running natural crack more closely than does a machined notch.

It was found that both the rolling and transverse directions undergo a sharp transition in fracturing mode and behavior between -30 C to -85C for the size specimen and cold work used. From room temperature down to -60 C the fracture occurs in a ductile manner by fibrous tearing. Many of the specimens tested at room temperature did not fracture rapidly but failed entirely by slow crack propagation. Large shear lips and severe gross deformation could be seen on the fracture surface. Also clearly defined on the surface were many pits indicative of void formation prior to separation. Below -80 C the fracturing behavior is quite different. No gross deformation occurred and the shear lips were nearly nonexistent occupying only two to five percent of the surface area. The fracture surface was flat with a crystalline appearance indicating failure in a brittle manner.

Nominal stress calculations were made utilizing simple elastic beam theory and the net beam depth. The maximum nominal stress at the base of the fatigue crack above the transition temperature was found to be approximately 190,000 psi and 180,000 psi for the transverse and rolling direction, respectively. Just below the transition the nominal stress values fell to 95,000 psi and 100,000 psi for the transverse and rolling directions, respectively. Also, below the transition the nominal stress varied linearly with temperature with values of 79,000 psi and 86,000 psi for the transverse and rolling directions, respectively, at -196 C (boiling liquid nitrogen).

Critical elastic energy release rate or  $G_c$  values were found to be quite low at -196 C, 84 in-lb/in<sup>2</sup> for the transverse direction and 102 in-lb/in<sup>2</sup> for the rolling direction. Plastic bend angle measurements indicated that the  $G_c$  values become obscured by gross deformation at a temperature of -80 C for both the transverse and rolling directions. The plastic bend angle is determined from the deflection increment which deviates from the linear portion of the load-deflection curve. Below -80 C the plastic bend angle is essentially zero but increases rapidly above -80 C with increasing temperature. This is conclusive evidence that gross deformation ceases to exist below -80 C and the transition from ductile to brittle behavior is essentially complete.

Reactors for space auxiliary power applications operate at temperatures too high for common high temperature alloys. Refractory metals must therefore be used in these applications. It is desirable to learn what effect irradiation and reactor environment have upon the properties of these refractory alloys. Sheet and rod stock of columbium, columbium-1 w/o zirconium, tantalum,

tantalum-10 w/o tungsten, and TZM materials are presently on site. During the past month the initial design of a liquid metal convection loop capsule was completed. This capsule will contain 21 round tensile specimens. It is estimated that these specimens will operate initially at a temperature of 1300 F in liquid potassium. A prototype of the capsule is being designed in order that flow rates and heat transfer characteristics of the capsule may be studied. In addition, specimens of these alloys will be irradiated in helium-filled capsules at temperatures to 1400 F.

Damage Mechanisms. The objective of this program is to establish the nature of the interaction between defects present prior to irradiation and those produced during irradiation with emphasis on the role played by interstitial impurities. The investigation is presently concerned with high purity iron and its low carbon and nitrogen alloys.

Hardness recovery curves and photomicrographs of cold worked Ferrovac "E" and zone refined iron show two striking differences in behavior. The zone refined iron recrystallizes without hardness recovery and no grain growth was observed below 850 C, while Ferrovac "E" shows a large recovery peak upon recrystallization and accelerated grain growth above 650 C.

Tensile testing of Ferrovac "E", zone refined Ferrovac "E", and MRC zone refined iron has revealed marked differences in the yielding behavior. Ferrovac "E" exhibited a sharp upper yield point followed by an instantaneous drop in stress of 18,000 psi to the lower yield stress and then a region of Lüders strain. The zone refined irons showed pre-yield microstrain before a broad rounded upper yield point followed by a slow drop in stress of 50-700 psi to the lower yield point and then a region of Lüders strain. The pre-yield microstrain indicates some dislocations are mobile before unpinning from the atmosphere takes place at the upper yield stress. However, interrupted tensile tests with aging at 200 C for one hour show a reproducible yield drop in both the Lüders strain region and in the flow curve region indicating at least some atmosphere pinning. This behavior differs from the classical behavior as formulated from studies of lower purity materials and presents an interesting problem in explaining the propagation of the Lüders band with such small yield drops.

Foils of high purity iron, received from the Johnson-Matthey Co., have been irradiated to  $3 \times 10^{18}$  and  $1 \times 10^{19}$  nvt (fast). These foils will be stored until excess radioactivity has decayed. One

disk has been submitted for radiochemical analysis to identify trace impurities present in the metal.

Zircaloy Corrosion and Hydriding. An investigation of the effect of Zr-2 work history on the fractional pickup of theoretical corrosion hydrogen out-of-reactor was continued during the report period. Duplicate specimens representing the same levels of cold work and cut from the same plate stock as used for the previously reported in-reactor tests and accelerated out-of-reactor tests in 425 C steam were exposed to steam at 400 C and 1500 psig in a refreshed autoclave for 42 days. As in the earlier test, some of the specimens entered the autoclave with an initial oxide film equivalent to about 11 mg/dm<sup>2</sup>, while others were in the as-etched condition. Forty-two-day weight gains of all coupons were in the neighborhood of 45 mg/dm<sup>2</sup>.

Hydrogen pickup fractions for the precoated specimens were ~ 12% for both temperatures as were those for the as-etched material at 400 C. Hydrogen pickup fraction for as-etched material at 425 C was higher, about 23%. No significant trends with metal work history were observed.

Hydrogen analysis for irradiated Zr-2 specimens, quadrants 63 and 64, continue to show reduced pickup for material representing about 10% cold work. However, the differences are much less than shown previously for coupons irradiated at lower levels of flux intensity and integrated flux. Instead of differences approximating a factor of two, the current data show a range from negligible reduction to about 25%. As with the earlier data, comparison of fractional pickup results for individual coupons show considerable variation.

The possibility that crystallographic orientation in Zr-2 may contribute to observed variations in fractional pickup of corrosion hydrogen in-reactor was explored to a limited extent by the x-ray diffraction technique using a pole figure goniometer. Basal-plane pole figures were constructed from reflection count-rate data for program plate material representing annealed and 10% cold work conditions. Annealed material from plate stock 6515 shows a high intensity of preferred orientation with the basal plane parallel to the rolling plane. For 10% cold worked material there was a noticeable reduction of intensity of orientation in this mode. Comparative orientation data are being collected for the remaining plate material and other levels of cold work.

Super Alloy Corrosion in High Temperature Helium. Oxidation tests of Hastelloy X, a heat resistant nickel base alloy, in laboratory air and pure oxygen have been made at 1000 C on as-received and abraded sheet specimens. Neither oxidant pressure nor surface preparation have shown a significant effect on the oxidation rates of Hastelloy X. Weight gain curves for all tests deviate no more than 15% from one another in the 48-hour duration period of the tests.

Irradiation Damage to Inconel. The remaining portion of the Inconel pressure tube from the helium cooled DR-1 Gas Loop was removed. When the first attempt was made in 1962 to remove the entire loop assembly, the tube had broken in half. A 10-foot long portion could not be removed until special equipment was fabricated. This length of tubing has seen the highest neutron flux and operating temperatures up to 1150 F. Metallographic examination will be done on several sections from this portion of the tube in order to determine if further testing is warranted.

Irradiation Damage to Stainless Steel. Metallography samples from 316 SS inner liner of the helium cooled DR-1 Gas Loop were examined. Samples from the high flux-high temperature region (1000-1200 F) showed a large amount of second phase precipitate at the grain boundaries. Samples from the lower temperature (800-1000 F) no-flux region were free from this second phase.

Compatibility of Hot Calcium Metal with Structural Materials. Hot calcium metal in particulate form is a highly reactive agent used in the purification of inert gases. Mild steel pipe is frequently used to contain the calcium bed, but it has obvious limitations as a structural material at the commonly employed temperature of 650 C, especially in a pressurized system.

Three materials were considered as candidate structural materials for calcium bed containment: Haynes 25, Hastelloy X, and 302 stainless steel. All of these have strength and corrosion properties at elevated temperature which are much superior to mild steel.

Duplicate disks of sheet stock of each material were placed in a 304 stainless steel tube, alternating with layers of calcium metal particles. After assembly, the calcium and the metal samples were pressed together by means of screws in either end of the assembly. The system was then encapsulated under conditions of high vacuum and held at 650 C for 14 days.

The samples of 302 stainless steel and Haynes 25 showed neither deterioration or lack of ductility after the test. The Hastelloy X samples showed a marked loss of ductility over the as-received sheet. The resistance of the 302 stainless steel to the calcium metal implies that austenitic stainless steels of simple composition would be useful structural materials in contact with hot calcium. For pressurized systems where high temperature strength is an important consideration, Haynes 25 would be an appropriate choice.

Gas Loop Development. Methods of joining superalloys, stainless steels, and TD nickel in unusual combinations are being devised for construction of the model ATR high temperature gas loop at Hanford. The technology developed will also serve in the design and construction of the ATR at Idaho Falls.

Hastelloy X and C, Haynes Alloy 25, and AISI stainless steels 316 and 347 have been butt welded in pipe form to themselves and to each other by the tungsten-electrode, inert-gas process. As reported previously, all welds were sound as judged by visual, radiographic and macrostructure examination and fluorescent penetrant testing.

Weld joint ductility at room temperature has been determined by bend tests. Samples were prepared, tested, and judged in accordance with Section IX of the ASME Boiler and Pressure Vessel Code. Essentially, the test consisted of bending strips of the butt welded pipe through 180 degrees. The bend was of  $\frac{1}{4}$ -inch radius and the face and root of each type of weld was individually tested. To qualify under code, no cracks greater than  $\frac{1}{8}$ -inch in any dimension are permitted in the weld or weld zone.

Weld joints of Hastelloy X and Haynes Alloy 25 to themselves, to each other, and to 316 and 347 passed the bend test. The dissimilar weld joint between 316 and 347 also proved to be ductile. However, approximately half of the welds of Hastelloy C to Hastelloy C, Haynes Alloy 25, and stainless steel 316 failed the bend test. Hastelloy C welded to AISI stainless steel 347 was ductile.

The room temperature weld joint efficiency is being determined by tensile testing. Although testing is not complete, a pattern has been established. Dissimilar joints of the superalloys to each other or to AISI stainless steels 316 and 347 generally have a 100% joint efficiency. All these welds were made with Hastelloy W filler wire. The weld efficiency of the superalloys welded to themselves is high, but it is less than 100%. Filler metal in this case is of

the same composition as the base metals. The weld efficiency of the AISI 316 to 347 with 316 L filler metal is less than 100%.

Elevated temperature joint efficiency of welds involving only superalloys are being determined by stress rupture tests. An initial test of Hastelloy X welded to Haynes Alloy 25 was conducted at 1800 F in a helium atmosphere. Rupture occurred in the Hastelloy X away from the heat affected zone thus demonstrating a 100% weld joint efficiency. The elongation was about 6%, occurring almost exclusively in the Hastelloy X portion of the specimen. The stress necessary to rupture the Hastelloy X apparently was considerably greater than that indicated in published data, i.e., 3600 psi for rupture in 100 hours at 1800 F. In this test rupture was produced after the following stress-time sequence: 3600 psi for 166 hours, 5200 psi for 100 hours; 8000 psi for 22 hours. No explanation has been found for the apparent high strength of the metal. A stress-rupture specimen of Hastelloy X welded to Hastelloy X is now in test.

The environment of model loop piping will include thermal cycling between room and elevated temperatures. Butt welded pipe sections of the superalloys and stainless steels are being thermal cycled to detect any tendency for the weld joints or heat affected zones to crack. After 20 cycles between room temperature and 2000 F, the welds involving only superalloys exhibited no cracking as judged by visual and fluorescent penetrant inspection. After 20 cycles between room temperature and 1400 F, the welds involving stainless steels exhibited no cracks.

The microstructure of the superalloy base metals and welds has been examined by optical microscopy at magnifications up to 750X. The superalloy pipes were in the solution heat treated condition when welded. Actually, the normal commercial solution heat treatment of these alloys at a high temperature for a short time followed by an air quench results in the presence of precipitates. Examination showed the Hastelloy C with considerable precipitation in the matrix, the Hastelloy X with less and the Haynes Alloy 25 with the least. According to published literature, both the nodular and angular precipitates are mainly carbides of the  $M_{23}C_6$  and  $M_6C$  type where M represents carbide forming elements such as chromium, molybdenum and tungsten. Precipitation at the grain boundary of the three superalloys was apparent at 750X while not apparent at 100X. Some sources credit this type of precipitation with enhancing high temperature strength properties while other sources feel that it degrades the strength properties.

The heat affected zones of the superalloys as determined by grain structure and micro-constituents were only 10 mils thick. Within these zones there was a limited amount of grain coarsening and some solution and incipient melting of the second phase constituents during welding. The heat affected zone of the stainless steels appeared to be about twice as thick, or 20 mils. There was some grain coarsening and what little precipitate was present apparently tended to go into solution and to undergo some incipient melting.

The fused zone of all welds contained second phase precipitates in dendritic and acicular form.

There is little information on the change of high temperature strength properties of these three superalloys and particularly their welds with long exposures at high temperatures. Micrographs of Hastelloy X exposed for 10,000 hours at 1750 F are available in the literature. These show an agglomeration of the carbide both in the matrix and at the grain boundaries. Micrographs of Haynes Alloy 25 aged at 1800 F for times up to eight hours show this same tendency. Since the disposition of the precipitates strongly influences the high temperature strength properties, a change in properties would be expected with long time exposures - a thermal condition of the model loop. Therefore, superalloy weld samples for bend, tensile, stress rupture and metallographic examination are being prepared for long time exposure at temperatures in the range of 1700 to 2000 F.

Two general type test section designs for the model high temperature gas loop were evaluated during the month by scope heat transfer calculations. Both cases investigated utilized double pressure tubes on the outside with concentric tubes and gas annuli inside, the only difference between the two being the number of layers.

General results show that since the heat removal capacity of the helium coolant stream is limited, much of the specimen heat must be removed through the test section walls into the surrounding water coolant. This requirement makes the following necessary:

- (1) The volume or density of materials used for the test specimens and specimen holder assemblies should be minimized to permit the helium coolant stream to remove as much of the specimen heat as possible.

- (2) No stagnant gas annuli can be allowed. Therefore, either the double pressure tubes should be eliminated or some means provided to insure adequate heat transfer from the inner tube to the water surrounded outer tube.

Neutron Dosimetry and Radiation Effects. Analyses of the space-energy distribution of neutrons which cause damage in structural materials are in progress for several reactors where test irradiations are made on these materials or where the materials will be used in service. These calculations provide a means to interpret flux-monitor activation data and to correlate exposure between dissimilar irradiation facilities.

One analysis is that of the northwest quadrant of the Engineering Test Reactor. This quadrant includes the Hanford hot-water loop, which is used for irradiation of reactor metals, and several graphite irradiation facilities.

The analysis was made using the 2DXY Transport-theory Code with 18 energy groups, 16 of which are above 0.183 Mev. Spatially, a 19 x 19 array was used with one-inch intervals. Thus, 361 solutions were obtained at each energy level. Results for several positions in the water loop show that variations in exposure occur over a small distance and demonstrate the need for this type of analysis. The average cross-section values for the Ni-58 (n, p) Co-58 and Fe-54 (n, p) Mn-54 reactions do not vary over about 10%; however, the flux intensity computation showed a factor of three. Thus, in such a facility, there is a serious difficulty in interpreting exposure data if monitors are at one position and specimens at another even though the intervening distance is small.

## 6. Gas-Cooled Reactor Studies

EGCR Graphite Irradiation. The experiments in the series of long-term irradiations of EGCR graphite are proceeding successfully. The fifth capsule in the series, H-3-5, completed the fourth cycle of irradiation satisfactorily and was removed from the EGCR on February 26, 1963. The samples received 84.1 effective days of exposure. The maximum exposure received by samples which have been irradiated in all five H-3 series capsules is estimated to be  $1.4 \times 10^{22}$  nvt,  $E > 0.18$  Mev or approximately 90,000 MWD/AT equivalent EGCR exposure.

Upon disassembly the H-3-5 capsule was found to be in excellent condition. All samples were recovered intact, returned to HAPO,

and measurements are currently under way on them. All flux monitors were recovered from the capsule and are being returned. The typical small amount of carbon deposit was found on the lower part of the inner wall of the aluminum shell. Thermocouple number 3, which failed during the last cycle of irradiation, was inspected and a break was found in one of the wires at the right angle bend where the thermocouple enters the sample.

Construction has begun on the sixth capsule of the series, H-3-6. A design modification is under way in an attempt to lower the irradiation temperature of the two bottom sample positions. Since these two positions are in the higher flux region of the capsule, higher exposures can be attained in a shorter period of elapsed time. Thus, information on graphite contraction at the temperatures of particular interest in the EGCR can be attained more rapidly. Heat transfer calculations to aid making the necessary design changes are in progress on the 7090 computer. The H-3-6 capsule is to be installed in the GETR during the first week of April.

Effect of Additives on Dimensional Stability. Graphites prepared with additives by Speer Carbon Company under Contract DDR-118 have been discharged after a second exposure at 600 to 650 C in a Hanford hot test hole. The rates of contraction between 1745 and 4200 MWD/AT<sub>K</sub> for transverse samples of the 17 additive systems were in every case equal to or less than the 0.005% per 1000 MWD/AT<sub>K</sub> observed for the transverse control which contained no additive. Significant reduction of contraction rate was observed in the following samples.

Additive (1 w/o original mix)	Contraction Rate, %/1000 MWD/AT <sub>K</sub>
Fe <sub>3</sub> O <sub>4</sub>	0.0004
V <sub>2</sub> O <sub>3</sub>	0.0004
CrCl <sub>3</sub> ·6H <sub>2</sub> O	0.001
Al <sub>4</sub> C <sub>3</sub>	0.002

The comparatively low contraction rates for all samples reflect the transition from growth through nonlinear contraction to linear contraction, which occurs during the exposure interval in which data were obtained. Comparison of rates at this point should be indicative of the relative rates for linear contraction at higher exposures; however, it is expected that the ultimate linear rates will be higher than those determined above. Irradiation of these samples will be

continued and samples from Contract DDR-136 will also be charged. The latter will provide information on additive effects in graphites which are of interest in applications requiring high strength or impermeability but which have the disadvantage of high contraction rates.

Graphite Compression Test. Discharge of the 300 psi compression-test boats and measurement of the samples have been completed. An increase in difference between the length changes of the stressed and unstressed samples was noted. This increase in difference, which was less than that occurring during the first irradiation period, is similar to that observed for the samples under 150 psi compressive stress.

The five 150 psi boats have been recharged in 2C test hole at KW-Reactor for additional exposure.

The parallel NC-8 graphite samples irradiated under 100 psi tensile stress have been measured and are ready for recharging during the next reactor shutdown. The stressed samples increased in length while the unstressed samples decreased in length during the first irradiation period of about 940 MWD/AT<sub>K</sub> at 650 C.

Graphite-Water Vapor Reaction Under Gamma Irradiation. Measurements of the rate of oxidation of graphite by small partial pressures of water vapor in the Co-60 gamma facility have continued. A TSX graphite sample weighing approximately 8 grams is currently being employed. At sample temperatures of 600 or 680 C and a helium flow rate of 0.5 cu ft/hr, oxidation rates obtained at a dose rate of  $3.04 \times 10^6$  r/hr were about  $2.2 \times 10^{-6}$  hr<sup>-1</sup> and were independent of water vapor concentration (200-6000 ppm) and temperature.

A comparison of the present data with those obtained previously for TSX graphite but at a lower dose rate, about  $8.1 \times 10^5$  roentgens/hr, indicates that the oxidation rate is approximately proportional to the dose rate.

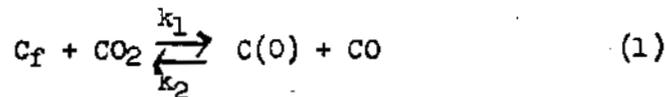
Inhibition of Graphite Oxidation. The oxidation of graphite by oxygen presents a temperature limitation on air-cooled reactors, and a gas-purity limitation on reactors cooled by inert gas. Moreover, accidental ingress of air, such as has been considered in the EGCR and HTGR hazards studies, could have serious consequences. The importance of controlling oxidation or extinguishing combustion once it has started has prompted studies of the kinetics and mechanism of gas phase inhibitors.

Recent results on inhibition tests with dichlorodifluoromethane ( $\text{CF}_2\text{Cl}_2$ ) and  $\text{CF}_4$  were reviewed and further tests have been conducted. Modifications of the equipment, especially the use of a recording balance, provided more accurate results. When graphite oxidizes in air the rate of weight loss gradually increases to a constant value after a few hours. With the addition of 0.5%  $\text{CF}_2\text{Cl}_2$  to the air, the reaction is more complex. With the same sample size and gas flow rate, there is an initial rapid weight loss as much as three times faster than the equilibrium rate in air, but the rate continuously decreases until in several hours the rate is one-third less than the equilibrium rate in air. After a sample has been partially oxidized in air and  $\text{CF}_2\text{Cl}_2$  a change to air alone will give a prompt increase in rate.

Previous results on the effectiveness of chlorine can be used as a basis for evaluating inhibitors. A unit concentration of a compound decomposing to give unit concentration of chlorine should be equally effective as the chlorine added directly. Direct inhibition and effects of other pyrolysis products would naturally give differing results. In the present test at 600 C, it was found that 0.5%  $\text{CF}_2\text{Cl}_2$  in air was about twice as effective as  $\text{Cl}_2$  in inhibiting the steady-state oxidation rate of EGCR graphite. On the basis of a preferential absorption mechanism for the inhibiting effect of chlorine, fluorine should be at least as effective an inhibitor since it is more strongly adsorbed.

The effectiveness of inhibitors in a radiation flux remains to be tested and for this reason the possible reactions with fluorine and other pyrolysis products of halocarbons are of special importance. Just as radiation can provide the activation energy for oxidation at low temperature and produce rates equivalent to a higher temperature, radiolysis of an inhibitor may generate products comparable to those from pyrolysis at elevated temperature and complicate the reactions occurring.

Graphite Oxidation Studies. An equation derived from the accepted mechanism of graphite oxidation by carbon dioxide adequately predicts the observed effect found when oxidizing a sample which has had an initial outgassing treatment. The mechanism is:



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where  $C_f$  is the number of unoccupied active sites and  $C(O)$  is the number of active sites occupied by an oxygen atom. The total number of active sites,  $C^*$ , is assumed to remain constant.

The fraction of active sites that are occupied by adsorbed oxygen atoms,  $\theta$ , is:

$$\theta = \frac{C(O)}{C^*} \quad (3)$$

The time dependence of  $\theta$  derived from the above equation is:

$$\theta = \beta/\alpha (1 - e^{-\alpha t}) \quad (4)$$

where  $\beta = k_1 P_{CO_2}$

$$\alpha = k_1 P_{CO_2} + k_2 P_{CO} + k_3$$

and  $\beta/\alpha$  is the fraction of active sites that are occupied at steady state.

In the present experiments the oxidation rate was determined in a flowing system where  $P_{CO_2}$  remained constant and  $P_{CO}$  was effectively zero; hence,  $\alpha$  and  $\beta$  were independent of time.

It has been observed experimentally that after a steady state concentration of  $C(O)$  has been attained the rate of weight loss is constant, and since the oxidation rate is proportional to  $\theta$ ,

$$-\frac{dw}{dt} = a \theta = \frac{a\beta}{\alpha} (1 - e^{-\alpha t}) \quad (5)$$

Integrating (5) gives:

$$w_0 - w = \frac{a\beta}{\alpha} \left[ t - \frac{1}{\alpha} (1 - e^{-\alpha t}) \right] \quad (6)$$

Equation (6) has been tested by oxidizing a few samples for over 100 hours each in the temperature range from 840 to 900 C. The results appear to agree with an equation of this form.

Under the more commonly studied steady-state conditions where  $e^{-\alpha t} \ll 1$ , the oxidation rate determined from (5) is the same as that derived from (1) and (2):

$$k_3 \theta = -\frac{k_3}{a} \frac{dw}{dt} = \frac{k_1 P_{CO_2}}{1 + \frac{k_2}{k_3} P_{CO} + \frac{k_1}{k_3} P_{CO_2}} \quad (7)$$

mechanical cleaning, and sandblasting all failed to leave the thorium surface oxide free. Cathodic etching was successfully used, but the process was temperamental and redeposition occurring during etching often left the surfaces unusable. Finally, cleaning was accomplished by brushing the thorium surface at high speed with a zirconium wire brush. The brush cleans the oxide film off and leaves a thin film of zirconium metal on the surface which prevents re-oxidation of the thorium. Excellent bonds resulted between the Zircaloy - 5% Be braze and the thorium core. Less thorium contamination was found in the braze when the surfaces were prepared by Zr-wire brushing than when the surfaces were cleaned by cathodic etching. All the brazed elements have been radiographed, machined, electron-beam welded, and are being autoclaved in preparation for irradiation in the ETR.

Thorium-Uranium Alloy. Two 23-pound ingots of Th -  $2\frac{1}{2}$  U (normal) -  $1\frac{1}{2}$  Zr have been double vacuum arc melted. This material will be coextruded to Zircaloy-2 clad rod stock for visual autoclave defect testing.

Extrusion will be done at 760 C from the  $2\frac{1}{4}$ " container to 0.525" OD with a 16.8 to 1 reduction ratio. Clad thickness will be 0.025". These conditions very nearly match those used on the previously reported enriched fuel material of the same composition.

Wire Drawing. A small bench type wire drawing machine has been built and has been successfully used in the drawing of fine (10-mil) high purity iron wires. The bull block for this device was made from the drive mechanism of an excess high pressure autoclave pump.

A wire lubricating device has been built and attached to the draw bench. This device was primarily designed to experiment with high temperature lubricants. However, it also serves the purpose of pre-heating wires which have a brittle phase at ambient temperature. A die box with an electrically heated reservoir will accommodate such lubricants as molten salts with and without additions of solid lubricants, low melting oxides and eutectics of oxides and some of the high temperature oils. It is also planned to experiment with asphalts and additives at various temperatures. Information gained from such experiments will benefit a much broader field of exotic and refractory metal forming than the initial wire drawing.

Refractory Metal Fabrication. Sintered molybdenum powder compacts of thick walled tubular shape have been successfully swaged at 800 C

to 50-60% reduction in area. The starting pieces, 1.5" OD with 0.375" wall, were made by cold compacting the powder in rubber molds followed by hydrogen sinter at 1600 C for 16 hours.

10. USAEC-AECL Cooperative Program on Development of Heavy Water Moderated Power Reactors

19-Rod Boiling Burnout Studies. The boiling burnout data from the 0.050-inch spaced 19-rod bundle which had warts to maintain the rod spacing have been reduced to a usable form. The data were obtained with an electrically heated test section made up of 19 Inconel tubes each 0.587-inch in diameter and  $19\frac{1}{2}$  inches long. The spacers or warts were made of  $Al_2O_3$  about 0.5 inch long, 0.10 inch wide and 0.050 inch thick. Two sets of spacers were used, a set about  $6\frac{1}{4}$  inches from each end. Each of the heater tubes had a thermocouple installed at its downstream end to measure an average inside wall temperature. Five thermocouples were installed between rods at the downstream end to provide some measure of the differences in local coolant temperature. The experiments were made with the test section in a horizontal 3.25-inch pressure tube and were done at 1200 psig.

The boiling burnout heat fluxes coincide very closely with those obtained with a test section identical to the one used in these tests except with rod spacing maintained with a 0.050-inch OD wire wrapping on a 10-inch pitch on 12 of the rods.

The rod temperature data were used to calculate heat transfer coefficients at the downstream end of the rod. The average liquid phase heat transfer coefficient for all of the rods ranged from about 1500 B/hr-sq ft/ $^{\circ}F$  at a mass flow rate of 500,000 lb/hr-sq ft. These heat transfer coefficients show the dependence on the 0.8 power of the flow that is predicted by standard correlations.

The liquid phase heat transfer coefficients of individual rods reflect not only the temperature drop across the water film on the heat transfer surface, but also the elevation or depression of the water temperature around that rod above or below the bulk water temperature. It has not been found possible to separate the true heat transfer coefficient effect from the local water temperature effects. However, if the assumption is made that the average heat transfer coefficient is a good measure of the local heat transfer coefficient of each rod, a calculation of the average local water temperature around that rod can be made. Such calculations show that the water seen by the inner seven rods increases in enthalpy

from inlet to outlet about 1.9 times that of the water seen by the outer 12 rods. This spread appears to be independent of the flow rate.

The five thermocouples installed to measure local water temperatures show a much larger deviation. The enthalpy increase of the hottest channel was about five times that of the coolest channel. This is a larger value than would be predicted if no mixing were assumed and indicates probably that the thermocouple locations were such that they did not provide a good average temperature measurement.

The heat transfer coefficients improved significantly when local boiling started. The improvement continued with increasing local boiling and reached values 5 to 10 times the liquid phase heat transfer coefficients when bulk boiling conditions existed.

Two-Phase Pressure Drop Studies. An experimental program investigating two-phase pressure drops in standard pipe fittings was started. The experiments included use of 2-inch straight pipe, a branch Tee, a 3-inch radius ell, a 3x2-inch reduction, a 3x2-inch expansion, and a gate valve. Flows from 500,000 to 4,000,000 lb/hr in a 2-inch pipe and qualities up to about 25% were covered at a pressure of 1200 psia. The program is about one-third completed and will continue to cover other pipe fittings and a few studies at other pressures. Approximately 1990 data points have been obtained to date.

Analysis of the data has not been started. However, the raw data show that the two-phase pressure drop of most components is about 60% of that predicted by the Martinelli-Nelson correlation. The 3-R elbow had a two-phase-to-liquid phase pressure drop ratio about 25% larger than that of straight pipe. The 3x2-inch expansion had a two-phase-to-liquid phase pressure drop ratio about two-thirds that of straight pipe. The 2x3-inch expansion showed the pressure increase that would be expected from the change in velocity.

Dome Seal Type Closure. The retaining lugs on the dome seal were placed and the assembly reinstalled in the test loop for pressure and temperature cycles with disassembly-assembly of the seal between each cycle.

After five cycles the flattening deformation at the dome center was 0.076 inch. After eight cycles the center dome deformation was 0.135. Also, during the 2 hours and 50 minutes the eighth

cycle required, one cc of leakage was collected. This was the only leakage observed during the entire dome seal test to date. The disassembly torque for these eight tests was between 75 ft-lb and 30 ft-lb, and the force required to remove the assembly averaged about 400 lb.

Testing has been halted pending a recheck of calculations. The original calculations indicated stress values below yield.

#### 11. Advanced Reactor Concept Studies

Fast Supercritical Pressure Power Reactor. Further 18-group diffusion theory calculations were completed to investigate power distribution in the present core design. It was found that one of the two moderating regions within the core could be removed and placed in the radial blanket. This resulted in a significant improvement in both power distribution and enrichment as well as a negative coolant void coefficient. In anticipation of a batch-fuel cycle, redistribution of enrichment to flatten the radial distribution was also investigated.

Radial dimensions of a near final core design are as follows:

Inner Core	42.6 cm
Flux Trap	
Depleted U	50.6
ZrH <sub>x</sub> Moderator	53.6
Depleted U	61.6
Outer Core	88.0
Blanket	
Depleted U	92.0
ZrH <sub>x</sub>	95.0
Depleted U	125.0

The critical mass for this core design was calculated to be near 1350 kg fissionable plutonium for a 5-foot core length. The reactivity loss on total coolant voiding was 1%  $\Delta k/k$  and voiding of each core region independently was also found to result in a reactivity loss. The initial conversion ratio, not including the axial blanket was calculated to be about 1.6. The radial core power distribution is characterized by a peak to average value of 2.0.

The effect of burnup on the reactivity, power distribution, and coolant coefficients is next to be examined. At present there is some difficulty with the manner in which the HFN code normalizes the power density to unit length. Calculations on the burnup effects will proceed when this difficulty is resolved.

The preliminary run of the simplified version of the Fast Super-critical Pressure Power Reactor core with the transport theory code Program S (11th Revision), using an 18-group cross-section set uncorrected for U-238 self-shielding has been completed. The code converged to within 0.01%  $\Delta k/k$  of the fission eigenvalue after 69 iterations using a first-order-perturbation-theory convergence acceleration of dual adjoint-and-flux field and verified by a free-run of 12 more iterations. The fission eigenvalue for this case, predicted by transport theory, was 3.8%  $\Delta k/k$  higher than that predicted by diffusion theory (HFN) for the same case, rather than 2%  $\Delta k/k$  lower as estimated in the February monthly report. The previous estimate was based on an extrapolation prior to convergence after only 15 iterations. The large number of iterations required to obtain the above-mentioned convergence and the deviation between the transport theory and diffusion theory fission eigenvalue predictions appear to be due to the non-ideal mesh-spacing (memory limitation) in the transport theory calculation. The adverse effect of the two core regions acting as a system of two loosely coupled reactors is also increased by the nonideal mesh spacing. A second transport theory calculation with Program S has been started on the present FSPPR core configuration using the 18-group cross-section set corrected for U-238 self-shielding and a more ideal mesh spacing.

Tentative heat balance of the FSPPR steam cycle indicates a net plant efficiency of about 43.8% compared to 43.0% reported for the SPPR. Although the FSPPR and SPPR cycles are based on identical throttle and reheat conditions, the advantage in the fast reactor concept is gained by better utilization of the secondary reactor heat removal system. The SPPR concept employed a low pressure, low temperature, light water moderator, which removed about 39 Mw of heat from the reactor. Only 24 Mw of this heat was transferable to the steam cycle; hence, 15 Mw was rejected. Total heat loss to the gas is about 10 Mw, and all heat is transferable to the steam cycle.

Studies of piping layout between the core and the main feed and steam headers were initiated. It is desired that the layout permit fuel handling of single elements and the use of minimum numbers of jumpers from the core to the headers. However, the compact core

size may necessitate compromise of the desirable features with practical considerations of space required to perform the necessary operations of disconnecting and connecting the fuel element to the jumper.

A preliminary estimate of the fabrication cost of FSPPR core elements is \$300/kg U + Pu; similarly, for blanket elements, \$250/kg U. This relatively high fabrication cost is based on a production rate of one (1) loading every 4 years. Even with these fabrication costs, a loading of 2000 kgs total Pu, and enrichment of 20% total Pu, the fuel cycle cost will approach 1 mill/kwhr at 100,000 MWD/MT (5-yr life). Comparatively, costs will increase about 3/4 if core life is halved and will decrease about 1/3 if life is doubled.

Fuel Re-use. Conceptual design drawings have been made by Ceramics Research and Development for the proposed fuel re-use demonstration experiment irradiations in PRTR. A new pressure tube would be permanently fitted with a liner to provide a square fuel channel duplicating the Fermi blanket cans. Stainless rods and end-fittings for a hydraulic take-apart model of the experimental fuel element are being procured. A new fuel shipping cask being designed for carrying a single PRTR fuel element is being modified to be suitable for off-site shipment of PRTR-Fermi interchange elements.

Plutonium Fuel Spacecraft Reactor. The document describing the Plutonium Fuel Spacecraft Reactor concept will be published in April.

Moon Base Reactor. Work is proceeding on establishing system parameters for a 10 Mwe epithermal reactor concept, fueled with "Phoenix Fuel" plutonium isotopes, to provide power for a base on the moon. This reactor concept utilizes plutonium nitride fuel, yttrium or zirconium hydrides as moderator, and is lithium ( $Li^7$ ) cooled. Power is recovered through a potassium secondary coolant system. Reactor coolant temperatures will exceed 2000 F although a final temperature has not been settled on. It will probably be necessary to provide a separate coolant for the moderator, to prevent excessive losses of hydrogen. Heat removed from the moderator could possibly be utilized to preheat potassium in the secondary loop.

## D. DIVISION OF RESEARCH - 05 PROGRAM

### 1. Radiation Effects on Metals

This program is aimed at establishing the combined effect of impurities and neutron irradiation on the properties and structure of specific metals and deducing from thermally activated recovery processes how the damage state can be altered.

Single Crystal Molybdenum. X-ray lattice parameters of molybdenum single crystals are being measured in a diffractometer modified for the purpose. A standard diffractometer was rotated 90 degrees and moved away from the x-ray tube to permit measurement of peak positions on each side of the 180 degree  $2\theta$  position. The collimator consists of a pair of pinholes, one being a length of 0.1 mm ID tubing very near the tube focus, the other a 0.5 mm diameter hole in a lead foil very near the sample. Separation between the two pinholes is about 30 cm, so the angular width of the beam is approximately 0.05 degree, or three minutes at half maximum. In theory, this arrangement should permit lattice parameters to be determined to better than one part in 100,000. For molybdenum, this would be  $\pm 0.00003$  A.

The measurements of lattice parameter revealed that there is a variation from the exterior of the single crystal rod to the interior. Lattice parameters of unirradiated low-carbon molybdenum (10-20 ppm C) were 3.14706 A at the edge, 3.14708 A at a point halfway between edge and center, and 3.14735 A at the center. An irradiated crystal ( $10^{18}$  nvt,  $E > 1$  Mev) of the same carbon content gave the values 3.14691 A at the edge, 3.14681 A at the halfway point, and 3.14715 at the center. A medium-carbon crystal (100-200 ppm C) irradiated to  $10^{18}$  nvt gave values of 3.14704 A, 3.14732 A, and 3.14741 A at the corresponding locations. An unirradiated crystal of this carbon level has not yet been studied.

The number of measurements is too small for any but the most tentative conclusions. It would appear that the effect of the irradiation on the low-carbon single crystal is to reduce the lattice parameters. A possible mechanism contributing to the observed decrease is a relaxation of microstresses during irradiation. Further measurements are being made.

Tension testing of molybdenum single crystals has continued through the month. A total of 41 crystals has been tested and the flow curves computed; of these tests, 20 have been conducted with photographic recording of the process of deformation. In this technique

the crystal specimen is thoroughly degreased and coated with a 0.0001-inch thick photographic emulsion. A photographic negative of a grid, with 0.002-inch thick lines spaced on 0.020-inch centers, is placed in tangential contact with the gage section of the crystal. After exposure to light from a mercury arc lamp, the emulsion is developed and the resultant image is dyed. In order to bond the emulsion to the crystal, the emulsion must be fired. This is done by heating rapidly to 250 C; the combination of relatively low temperature and rapid heating are believed to minimize any annealing of radiation damage during firing. During tensile testing, deformation of the grid is recorded by the use of a 35-mm time-lapse camera. One frame is exposed for every 0.001-inch of crosshead travel in the interval from the proportional limit to the ultimate load. During the remainder of the test the exposure interval is 0.005-inch of crosshead travel.

Preliminary examination of the photographic recordings obtained indicate that, in addition to precise determination of tensile strains, it is possible with this technique to observe the intersection of slip lines with the surface, the onset of nonuniform deformation, the occurrence of multiple necking, and other anomalous deformation characteristics.

The tensile tests conducted on crystals with exposure of  $10^{19}$  nvt ( $E > 1$  Mev) have confirmed the tendency for drastic restriction of the number of operative slip systems after irradiation. The extent of the easy glide region becomes very great at  $10^{19}$  nvt, with glide strains in easy glide of the order 0.80. This suggests that the "channeling" observed by transmission electron microscopy at exposures of  $10^{19}$  nvt should be more pronounced than in specimens with exposures of  $10^{18}$  nvt.

Polycrystalline Molybdenum. Defect structures have been observed in high purity molybdenum which contains  $< 10$  ppm C as well as  $> 100$  ppm C after neutron irradiation to  $10^{19}$  nvt (fast). These bulk foils (0.003-inch thick) have been strained axially in the range one to two percent to study the interaction between defect structures and dislocations which have been induced to move as a result of the deformation. Evidence for such interaction has been observed by transmission electron microscopy in the form of straight line channels from which the irradiation-produced defects have been partially or completely removed. The origin of the channels is presumed to be the grain boundary. In some instances areas free of defects were observed at grain boundaries; in these regions defects had apparently been swept away by moving dislocations originating at the boundary. Large carbide particles have been observed in some of

the channels of foils containing 100-200 ppm C. It is possible that the carbides, under stress, will act as sources of dislocations which move in the glide plane sweeping out defects. Dislocation tangles are present at some of the intersections of the channels indicating complex interactions with each other. Occasionally, relatively large dislocation loops are also found in the channels. They presumably are formed by the complex interaction of dislocations. Tilting of the specimen in the electron microscope disclosed a change in the orientation of some of the channels with respect to the matrix material. This contrast effect may result from a reorientation of the material within the channel, due to the passage of many dislocations, or the formation of microtwins. Electron diffraction patterns of the channels and the matrix should show the degree of misorientation. The occurrence of channels observed in the stressed samples is in good agreement with one theory of radiation hardening, i.e., an increased stress is required to move a dislocation through a field of radiation-produced defects. Since essentially all clustered defects in these channels have been eliminated, many dislocations must have passed along these channels. With the elimination of the defects subsequent dislocation motion in the channels requires less stress. Attempts at observing the formation of channels in the electron microscope have been unsuccessful. Micro-tensile samples could not be punched from the irradiated 0.003-inch thick disks, as embrittlement of the metal had occurred to such an extent that the irradiated disks shattered. Preformed micro-tensile samples are therefore being prepared for neutron irradiation.

Operational tests of the stored energy calorimeter are continuing. Certain modifications were made which allow the specimen stand, the internal heaters, and the thermocouples to all be removed from the calorimeter as an integral unit. Also, the Pt-Pt 10% Rh differential thermocouples were replaced with chromel-alumel couples to provide greater sensitivity in differential temperature measurements. An improved specimen heater support, fabricated from high purity  $Al_2O_3$ , was installed. Polycrystalline molybdenum rod,  $\frac{1}{2}$  inch diameter, has been upset at 500 C in a four-step operation to a final diameter of  $\frac{3}{4}$  inch. The rods are now being swaged and heat-treated to produce  $\frac{1}{2}$  inch diameter specimens with different levels of cold work for use as test specimens.

Other Metals. Nickel, a face-centered cubic metal, and rhenium, a close-packed hexagonal metal, will be investigated in a manner analogous to the molybdenum studies.

Nickel foils 0.003-inch thick have been prepared by rolling 1/8-inch thick strip, which was in turn rolled from 1/2-inch diameter rod material. The material is broken down into three groups, according to the purity level of the starting material. These purity levels are (1) 99.97% Ni, (2) 99.6% Ni, and (3) 99.2% Ni. The foils were prepared by cross-rolling, with the reduction per pass kept less than 20% in order to retard the formation of a pronounced rolling texture. Electrolytic thinning techniques for these foils are under development, and examination by transmission electron microscopy will follow.

The effects of a high supersaturation of vacancies on the electrical resistivity of nickel is being investigated. Wires 0.010-inch in diameter have been prepared from the three materials mentioned above, and preliminary quenching experiments have been conducted. The specimens used are coils of 1/8-inch diameter with a gage length of 30 to 36 inches. The wire is heated at a pressure of  $10^{-6}$  mm Hg to 1400 C by direct resistance heating and held for five minutes to anneal out the cold work damage incurred during winding of the coil. Then the vacuum chamber is backfilled to 10 psig with pure helium and the chamber is immersed in a liquid nitrogen bath. The specimen is then heated to  $1430 \text{ C} \pm 20 \text{ C}$  and quenched by turning off the current. The quenching rate is so fast as to appear instantaneous, but it is planned to measure the rate by determining the rate of decrease of gas pressure with a pressure transducer sensing system.

Rhenium wire (0.010-inch diameter), rod (0.100-inch diameter), and single crystals (1/8-inch diameter) are on order. An electrolytic machining device for strain-free machining of single crystal tensile specimens is being fabricated and will be completed shortly.

## 2. Plutonium Physical Metallurgy

The objective of this program is to determine some of the basic physical metallurgical properties of high purity plutonium and to establish the effect of certain specific alloying additions on these properties. Two areas are under study: mechanisms of phase transformations and mechanisms of deformation and recovery.

Phase Transformations. Studies on the effect of plastic deformation on the beta  $\rightarrow$  alpha transformation, and the effect of compressive loading on the microstructure of specimens undergoing beta  $\rightarrow$  alpha transformation have continued.

Specimens were beta heat-treated, deformed by compression at a temperature of 150 C, and then permitted to transform at a temperature of 79 C. Changes in the thickness of the compressed specimens were in the range 60-97%. The plastic deformation associated with the beta phase compressions increased the time for the start of transformation into alpha. The incubation times were in the range 3000-6000 seconds. Undeformed specimens with the same thermal history required only a 200-second incubation period at 79 C for transformation into alpha. A marked difference in final density of the deformed and undeformed specimens after transformation was indicated. This effect will be investigated more fully.

The normal effect of plastic deformation on nucleation and growth reactions is to increase the rate of transformation whereas the effect on nucleation and shear type reactions is to lower the  $M_s$  temperature. The observed decreased rate of transformation after plastic deformation supports the contention that the beta  $\rightarrow$  alpha transformation is a nucleation and shear transformation.

Experiments were performed in which specimens were allowed to partially transform from the beta to alpha phase under an applied compressive stress of 5000 psi. The transformation at 79 C was interrupted by quenching to -40 C with no applied stress. The alpha grains which formed under the applied stress were columnar and in bands. The bands were observed at 90 degrees to the applied stress and the major axes of the columnar grains were parallel to the direction of the applied stress. The bands which appeared to have nucleated at the center of the specimens increased in number as transformation proceeded, rather than in size.

The bands appeared to have nucleated at the center of the specimens. This is also evidence that the beta  $\rightarrow$  alpha transformation may be a nucleation and shear reaction.

Electron Microscopy. Initial attempts at obtaining electron photomicrographs of delta stabilized plutonium have been successful. Grain structure and microcracks are readily observed. A plutonium sample, delta stabilized with four a/o Al, was etched by argon ion bombardment at 3500 volts for 12 minutes. Six successive cellulose acetate replicas were taken and decontaminated to < 500 D/M smearable contamination by ultrasonically cleaning in alternate baths of six normal HCl and water. The cellulose acetate was shadowed with palladium and replicated by carbon evaporation. After dissolution of the acetate in acetone, the carbon replicas were immersed in water and then retrieved on 200 mesh copper grids. No contamination

could be detected on the grids. Rinsing the carbon in  $\text{HNO}_3$ , as described in the previous monthly report, was found to be unnecessary for replicas taken from a cathodically etched surface.

#### E. CUSTOMER WORK

##### 1. Radiometallurgy Laboratory

Examinations. The split failure from Tube 3584-C was examined and photographed as-received. The uranium fuel was badly fragmented over the entire length. Visual examination of the spire indicated that failure was not the result of water penetration along the spire. (RM C-417)

Three I&E elements from B Area were replicated to study the corrosion patterns of the outer can wall. On two of the elements deep galvanic type corrosion pits were found in which the uranium was exposed. The agent that caused the corrosion in these pits has not yet been determined. The can walls were also badly pitted with groove corrosion, but the two distinctly different types of corrosion did not appear to be associated. (RM C-416)

Physical and Mechanical Properties Testing Cell. Installation of the Instron straining frame with power track and accessories has been completed. Final checkout and adjustments were performed on the Instron positioning device. Installation of the electrical system for the rotating beam fatigue tester has also been completed. Final adjustments were made on the cell door drive mechanism. Equipment functional tests will commence following a complete checkout of the installed equipment.

Equipment. The design of high temperature tensile grips was completed and the work request for fabrication was issued. The grips are required for testing process tube specimens at temperatures up to 1500 F.

Several modifications were made in the micro sampling equipment in "D" Cell and this equipment is now available for use. The installation of the pulse annealing furnace in "D" Cell is also complete.

Inspection reports that the vendor has completed the basic optical system drawings and is working on the basic mechanical drawings for the stereo zoom microscope. Detail drawings are expected for approval about April 15, 1963.

An order was placed for two light-duty CRL Model G Manipulators for evaluation purposes.

Testing of the resin column portion of the filtering system for the basin is complete. A larger capacity pump was ordered to replace the existing filter pump when the resin columns are installed.

## 2. Metallography Laboratories

An electropolishing-etching electrolyte consisting of one part 60% perchloric acid and 10 parts glacial acetic acid has been shown, over the past few months, to produce good results in the preparation of uranium bonded to Zircaloy. Recent experimentation with this electrolyte has shown that it is also very effective for polishing such diverse materials as aluminum-lithium alloys and several of the super alloys, e.g., Hastelloy-X, and the stellite alloys.

## 3. N-Reactor Design Testing

### N-Reactor Charging Machine

Modifications. The new end thrust stabilizer rod anchors have been modified to lower the line of action.

Installation of the television and communication equipment is complete.

Testing. Four more complete charges of fuel elements were made in addition to the ten made last month under Design Test 22-A. These were:

- Test 11 - 18 elements, 24" long with suitcase handle type supports.
- Test 12 - 18 elements, 24" long with suitcase handle type supports.
- Test 13 - 36 elements, 12" long with suitcase handle type supports.
- Test 14 - 24 elements, 18" long with suitcase handle type supports.

Both charge 11 and 12 suffered some visual damage to the fuel element feet. This was described by NRD personnel as gouging of varying degrees, probably caused by the nozzle to process tube transition and attributed to the soft iron shims which were used extensively in these two charges under the shoes to provide the proper support height. No apparent damage occurred to the last two charges.

Preparation for Design Test 22-B is estimated 70% complete. This test is similar to Test 22-A in that charges are pushed over the same magazine-nozzle-pressure tube transitions but with the addition of a resisting force on the fuel to simulate possible column action resulting from forces necessary to push an irradiated charge of fuel from the pressure tube.

#### NPR Fuel Handling Tongs

The fabrication of the tongs is 90% complete.

#### 4. Special Plutonium Fabrication

High Exposure Plutonium-Aluminum Fuel for Physics Tests. One hundred, 3-foot long, Al-Pu rods were fabricated for physics tests in the PCTR and PRCF. Billets were cast for another 50 rods.

High Exposure PuO<sub>2</sub>-UO<sub>2</sub> Fuel for Flux Monitor Tests. Seven fuel rods containing UO<sub>2</sub>-PuO<sub>2</sub> pellets were completed, and pellets for five additional rods were sintered. A total of 18 rods with pellets are needed. During the month the hydrogen sintering furnace heating element burned out; replacement of the element delayed completion of this work.

Low Exposure PuO<sub>2</sub>-UO<sub>2</sub> Fuel for Physics Tests. This work has been postponed to permit completion of the flux monitor pellets described above.

High Exposure PuO<sub>2</sub>-UO<sub>2</sub> Fuel for Physics Tests. Use of high energy impact formed PuO<sub>2</sub>-UO<sub>2</sub> vibrationally compacted into rods for physics tests continues to look promising as an alternate to pellets. Reasonable bulk density (86% TD) was achieved in the required 3-foot long rods by vibrationally compacting 99% TD impacted UO<sub>2</sub>. Preliminary analyses of impacted UO<sub>2</sub>-1.3 w/o PuO<sub>2</sub> indicated greater Pu segregation than normal for this material; however, the sampling technique and analyses are suspect. Repeat samples are planned.

Plutonium-Aluminum Fuel for Corrosion Tests. Fourteen coextruded 5-inch Al-Pu fuel rods with X-8001 cladding passed bond testing and are ready for autoclaving. A rod clad in KYZ aluminum was coextruded for cutting into the second set of fuel rods.

EBWR Plutonium Fuel Elements. The complete detail design package for the vibration compaction facility in 308 Building (Project CGH-992) was delivered to AEC-RLOO for approval. With AEC approval of the design, funds will be released to start construction and

field engineering on the project. Existing equipment in Room 138 is being removed and construction of the pit will start as soon as funds are available.

Development work on the oxide loading process for EBWR began. A series of mixed oxide fuel rods were loaded by vibrational compaction to evaluate blending and loading techniques for "bottle loading" of  $UO_2$ - $PuO_2$  powders. Feed material for these rods was subjected to various blending actions (V blender, roller blender, and end-for-end tumbling) for periods ranging from 5 minutes to 2 hours. Evaluation of these rods by autoradiographs and gamma scanning is under way.

Uranium dioxide was prepared for blending with  $PuO_2$  and impaction of prototypical EBWR fuel material. Depleted uranium was reacted with pressurized water, yielding a fine powder of the composition  $UO_{2.25}$ . The powder was converted to a poorly-sinterable stoichiometric  $UO_{2.00}$  by reduction with hydrogen at 1200 C. The  $UO_2$  was impacted at 220,000 psi to 97% TD, pulverized to -48 mesh, and air roasted to the composition  $UO_{2.01}$ . This type of  $UO_2$  can be mixed with  $PuO_2$  and impacted to 99% TD at 1200 C and 250,000 psi impact pressure, using a tool steel punch.

A program for irradiation testing prototypic  $UO_2$  - 2 to 3 w/o  $PuO_2$  EBWR fuel rods was prepared. The GETR Boiling Water Loop, currently empty, would accommodate a 36-inch long, 3x3 array of the vibrationally compacted, 0.42-inch diameter, Zircaloy-clad rods. The loop can be operated to simulate EBWR conditions. The desirability of conducting irradiation tests in that facility is being determined.

#### 5. Fission Product Impaction

In cooperation with Chemical Laboratory personnel, simulated fission product oxides were impacted in both 2.5- and 4-inch diameter containers. Materials impacted included  $SrTiO_3$ ,  $SrO$ ,  $CeO_2$ , and  $Nd_2O_3$ .



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B-1

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PHYSICS AND INSTRUMENTS LABORATORY

MONTHLY REPORT

MARCH 1963

FISSIONABLE MATERIALS - O2 PROGRAM

REACTOR

N-Reactor Lattice Parameters

Work has continued on the re-analysis of experimental data. A systematic error in the measurements of "p" and "e" has been discovered which was due to a coding flaw in the foil activity correction program. Changes resulting from this error are small. The measurements in the mockup and condensed lattice are now being corrected to a common graphite density.

Optimization of Re-Tubed Lattices

The overbore (CVIN) fuel has been removed from the "C" exponential pile and replaced with air-cooled regular (CIIN) fuel. Measurements of the material buckling and control strength in this lattice have begun.

Computational Programming Services

A program which exceeds computer core space presents one or both of two questions: why is the deck too large, and how much must be cut to make it fit. The answers to these questions, as well as additional useful information about the deck, may be obtained by using PADOED (Program and Data Overlap - Execution Deleted). PADOED will examine a binary deck and list the name, length, and transfer vector of each routine included. The library tape is searched and the name (and secondary entry names, if any), length, and transfer vector of each required library routine is given. A summary sheet lists total lower and upper memory required, tells how much was wasted by duplicate routines, and points out names of all missing sub-routines. The program will also be useful in determining the maximum permissible dimensions for variables in a deck that does not exceed core space, since the exact amount of unused core space is given. PADOED is completely written and about 50 percent debugged.

A source program listing of the Reich and Moore multilevel Breit-Wigner formulation has been obtained from Phillips Petroleum Company for possible inclusion in program GROUSS as an option to the single level subroutine presently contained therein. Since GROUSS performs Doppler broadening

1103083

numerically, the asymmetry of fission resonances due to interference will cause no difficulty.

### Instrumentation

Continuing efforts are being made to evaluate neutron pulse-type detectors for use in special NPR startup instrumentation. A Helium-3 neutron counter tested for pulse-height distribution showed a sharp cut-off point at about 0.75 Mev when tested with a neutron source. Its gain increased by a factor of about 2.5 per 100 volt increase of applied voltage, while the pulse rise times varied from 1 to 3 microseconds. Other measurements included pulse height distributions from standard and pressurized (at three atmospheres) fission chambers.

Work continued on instrumentation for the NPR Fuel Rupture Testing Loop at PRTR. Progress has been made in the preparation of layout sketches for the flow sensing and control valve arrangement. Revised specifications were completed for the general purpose oscilloscope, an oscilloscope camera, and an X-Y recorder. Modifications were made to the multichannel pulse height analyzer to permit pulse observation at the analog to digital converter. General equipment layout work was started.

Gain, linearity, and stability test data for the dual-channel preamplifiers of the gamma energy monitor of the NPR Fuel Rupture Monitor were received from GE-APED and reviewed along with the test methods used. Acceptance test procedure methods were started, and plans were established for the next witnessing of tests at GE-APED.

Theoretical calculations were completed regarding a single time-to-power meter presentation of flux level and period data for use in reactor start-ups. An invention report was prepared and submitted.

The development work on the graphite channel bore-gage for reactors was suspended due to apparent lack of customer interest.

Experimental evaluation of the cable continuity acceptance test procedure for the NPR high level flux monitor has been started. The test method experiment was requested by Instrument and Electrical Design, NRD.

### System Studies

Simulation of the NPR pressurizer-injection system was completed this month. This simulation was used to determine automatic controller settings and to determine the transient effects of various types of disturbances such as process tube diversion, loss of power to one or more injection pumps, reactor

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power level increases and decreases, and various scram conditions. Highly satisfactory results were obtained from the investigation of more than two hundred separate and distinct cases. Both computers (EASE and GEDA) were used. The simulation was operated two shifts each day for approximately two weeks.

Separate simulations of the NPR injection pump and pressurizer-injection systems are being designed to allow system studies using only the EASE analog computer. A mathematical model of the pressurizer which includes in-surge mixing effects has been formulated and is being prepared for programming. The simulation circuits are being designed in such a way that repetitive operation may be performed at a rate ten times faster than real time in order to provide more rapid problem solutions.

A single-node, six-delayed group simulation of the NPR is nearing completion. However, some of the instrumentation time responses and xenon constants of the simulation, which includes both temperature and xenon effects, have not yet been determined.

A single amplifier simulation of a proportional-plus-reset controller has been designed which facilitates repetitive mode computer operation. Designed for NPR simulation studies, the reset (integral) circuit consists of an external RC switching circuit which is connected to the EASE computer patch-board. The remaining controller circuitry consists of the normal built-in computer components.

A mathematical model of the NPR pressurizer has been formulated which describes its operation as a function of water surges. The model takes into account steam condensing and flashing as governed by the amount of water surging into or out of the pressurizer. A signal flow diagram based upon the model was constructed which shows the relationship between problem variables. Constants are being formulated to be used in the basic programming of the pressurizer.

An analog program for the NPR cooling gas composition study was placed on the computer and made operational. The purpose of the study is to determine the effects of changing the input cooling gas composition, and to determine optimum gas composition for various reactor operating conditions. Cyclic gas temperature fluctuations brought about by localized temperature variations of individual process tubes were considered in addition to the effect of more general, over-all temperature variations.

Production Test IP-569-C authorizing 100-D reactor experiments in which outlet coolant temperature will be determined as a function of control rod position has been approved. Tests at that reactor are tentatively planned

1103085

for the first week in April, or as soon as possible after in-core neutron chambers are calibrated. As temperature changes for each rod will be expressed in matrix form, and since it will be necessary to convert this matrix into an analog simulation, the corresponding analog setup has been structured in matrix form. A solution to the matrix equation has been derived and programmed for the IBM 7090.

A single node, closed loop simulation of an automatic controller for 100-D reactor has been completed. Results indicate that the controller can be driven directly by the voltage outputs of an analog computer. Plans are to move the controller to the computer laboratory upon completion of the 4-node reactor simulation now in progress.

## SEPARATIONS

### Experiments with Plutonium Solutions

Additional critical mass measurements were performed with plutonium-nitrate solutions in an 11.5-in. diameter water reflected sphere with criticality being studied as a function of the Pu concentration and nitrate. Pu concentrations were in the range of  $\sim 220 - 80$  g Pu/l; the acid molarity of the solutions was  $\sim 0.3$  or less.

Some difficulties were encountered with the dump valve mechanism for the criticality vessel when a leak developed permitting solution to drain into the dump tank (in the event of a scram, the solution is automatically drained to the dump tank). On disassembly the valve was found to have a damaged O-ring seal which was replaced. An extension skirt was also installed on the dump valve actuating mechanism for improved contamination control.

### Pulsed Neutron Source Experiments

The first pulsed neutron source experiments at Hanford were conducted at the Critical Mass Laboratory on March 6. In these experiments, short bursts of fast neutrons were injected into the multiplying material, and the exponential decay of the prompt neutrons from fission was observed. In the pulsed neutron source experiments, one must have a neutron detection system capable of measuring the intensity of neutrons as a function of time within intervals in the microsecond to millisecond range. For these experiments a scintillation detector with a high speed liquid scintillator is being used in conjunction with wide band amplifiers to drive a multi-channel analyzer. Some pulse pile-up problems have occurred, but the preliminary results are encouraging.

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B-5

Pulsing a subcritical assembly with neutrons in no way affects the reproduction factor, and hence the state of criticality is not changed. Of course, the neutron level is increased during the injection--as it would be if any constant non fissile source were added, but this does not in any way create a criticality situation. (The fission neutrons resulting from the neutron pulse can in no way interact with the pulse tube to increase its yield--hence, in a subcritical system the only effect is to raise the neutron level by a constant value). However, the rate of decay of the neutron population, following the burst, is a function of the neutron lifetime, delayed neutron fraction, and  $k_{eff}$  (nearness to criticality of the system).

Plans are to use the pulsed neutron source technique in criticality studies with plutonium systems--and to measure effective multiplication constants of prototype process equipment relative to the in-plant applications and the development of a "criticality instrument".

In the first preliminary test of the pulsing device, the decay constant of the thermal neutrons in the tamper tank was measured. The lifetime of the neutrons in the water reflector was in qualitative agreement with the expected value,  $\sim 200 \mu$  seconds. A plutonium solution system was then pulsed at successive steps during an approach-to-critical in further preliminary testing of the pulsing mechanism and time analyzer equipment. The prompt neutron decay constant was measured at several different solution volumes ( $\sim 90$  g Pu/l) in the 11.5-in. water reflected sphere. A plot of the decay constant versus solution volume gave a value for the prompt critical volume of  $\sim 12.6$  liters; from the inverse multiplication curves, the delayed critical volume was  $\sim 12.5$  liters. A tenth of a liter between delayed and prompt critical was to be expected for this system.

With the vessel half full ( $\sim$  half the critical volume), the measured decay constant implied a  $k_{eff}$  of about 0.93.

Further analysis and interpretation of the data are required; additional experiments are planned during April.

## Experiments with Plutonium Oxide-Plastic Mixtures

The remote split table machine was installed within the second hood of the critical assembly room in preparation for criticality experiments with  $PuO_2$ -plastic fuels. These fuels are being used to cover the range of densities of plutonium precipitates and polymers which can form in separation plants.

1103087

Tests are currently being performed to determine the delay time of the control and safety rods for the split table machine in the event of a scram. The time for the fuel removal rods to move six inches after the controller trip point has been reached was measured and found to be approximately 200 milliseconds. The time from the radiation incident to the controller trip point will also be measured to yield a total delay time.

The neutron source drive is currently being installed in the hood.

#### Bucklings of Uranium Tubes and Rods in Light Water

Safe mass and volume limits are given in the Nuclear Safety Guide (TID-7016) for slightly enriched uranium rods. However, if tubular (hollow rods) fuel elements are being handled, one may question if these same limits can be safely applied to tubes of the same enrichment. To help determine the answer to this question, lattice parameter calculations were made for several rod and tube sizes for 1.03 percent enriched uranium in light water.

For the tubes the outside diameter was held constant, and the inside diameter varied to determine the ID giving the maximum buckling; several different outside diameters were used in the calculations ranging from 0.2 to 1.2 inches.

The calculations indicate that up to an OD of 1.2 inches, for 1.03% enriched uranium in light water, there is no tube size which has a higher buckling than the maximum obtainable for solid rods of optimum diameter with the same enrichment. However, for solid rods of non-optimum diameter, there are obviously tube sizes which do have higher bucklings.

#### Buckling of Partially Filled Spheres

The computer program is being revised to get better approximations for the average of the derivative terms representing leakage at the surface of a small volume element. Further revisions included corrections for the small irregular bumps on the top surface due to the spherical coordinate system. These corrections also lead to a more consistent approximation for the flux and its derivatives; the flux is a double quadratic in  $\theta$  and  $r$  coordinates, the first derivative component is linear in the parallel direction and quadratic in the perpendicular direction. Derivatives are still found using the Lagrange interpolation formula; however, this is a much better approximation than a simple linear first derivative. A great deal of hand calculation is necessary in the debugging of these changes as each one represents the evaluation of two definite integrals

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for each partial derivative at each of the nine points on the double quadratic flux fit.

A trimmed version of the code was used to solve the problem of a hemisphere. The value of B was found as 4.52, whereas 4.49 is the analytic value. The error of .6 percent is of the same order as the error in some of the derivative terms near boundaries. The calculation was hindered because the CROUTS simultaneous equation program in the library appears to "peak" in the exactness of its solution, that is, more iterations give a worse solution after a certain point is reached.

## Subcritical Interactions

Program INTERSET is being generalized somewhat to account for interaction between results of various concentrations and vessels with surrounding reflection. Changes in concentration can be handled by iterative solution of the matrix for  $k_{eff}$  instead of albedo.

An equivalent extrapolation distance replaces the albedo in each different concentration and  $k$  is found from  $k = k_{\infty} / \Sigma(1 + \tau_1 B^2)$  for each unit. Success of the method may be checked by using the various albedos to predict  $k$  for each unit by an HFN calculation.

Data from Oak Ridge are being used for comparison with the "constant flux in the void" approximation for an outside reflector. These data provide an extreme case for comparison, since (1) reflector coefficients for the reflectors used are poorly known, (2) the arrays were actually cubic, (3) cylinders are very closely spaced (where interaction probability values are poorest), and (4) unequal reflectors on the six sides of the array were used.

Comments were made on a safety report for the PURE facility.

## Consulting Services on Nuclear Safety - Criticality Hazards

### Nuclear Safety in HLO

The following nuclear safety specifications were reviewed and issued:

#### Experimental Reactors Operation

- B-2 (Revised) Rules for the Storage and Handling of 93% U<sup>235</sup> Enriched Metal Foils
- B-3 (Revised) Rules for the Storage and Handling of Pu-Al Alloy Rods and Foils

1103089

- B-4 Rules for the Storage and Handling of PCTR Fuel Elements  
B-5 Rules for the Storage and Handling of TTR Fuel Elements

### Thermal Hydraulics

- P-1 Rules for the Storage and Handling of Slightly Enriched Uranium Fuel Elements

### Nuclear Safety in CPD

Participation on the Recuplex Deactivation Committee continued. Efforts to remove 4-8 Kg of plutonium from the J-26A steam stripper with hot acid under slight pressure (Procedure A-35) have to date been unsuccessful. It is now planned to cut the vessel and remove the plutonium mechanically. Procedure A-45 was issued March 15, 1963, to cover the dismantling operation.

A hazards evaluation of the new Plutonium Recovery Facility (CAC-880) is nearing completion, and a hazards report is under preparation.

### NEUTRON CROSS SECTION PROGRAM

#### Scattering-Law Measurements for Light Water at Elevated Temperatures

A new  $\text{He}^3$  proportional counter was placed in operation as the scattered-neutron detector for the triple-axis spectrometer. This counter has a counting efficiency three times that of the  $\text{BF}_3$  counter assembly previously used with no increase in background counting rate. Measurements have been started on the inelastic scattering of neutrons from water at  $95^\circ\text{C}$ . Measurements have been completed for neutrons of two different initial energies with an energy after scattering of 0.114 ev.

#### Study of Systematic Errors in Scattering-Law Measurements

Attempts to calibrate the efficiency of the analyzing-crystal spectrometer for Scattering-Law measurements revealed some systematic effects which were previously either unobserved or not present. These effects manifested themselves as systematic differences in the measurement of the elastic scattering from vanadium using different orientations of the analyzing spectrometer. Measurements are in progress to determine the magnitude of the systematic errors which may result in Scattering-Law measurements.

#### Rotating-Crystal Spectrometer

Development of apparatus to measure slow-neutron inelastic scattering by

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B-9

time-of-flight was continued. Design work was done on a mechanical rotor to be phased with a rotating-crystal to eliminate unwanted reflections. Further studies were made of the phasing of two rotors using two separate hysteresis synchronous motors. A phase-difference stability of 0.1 degree has not been achieved with the system possibly because of poor alignment of one motor which has suffered repeated bearing failures. Efforts have been made to improve the performance of this motor. One commercial and one HAPO built BF<sub>3</sub> counter were tested for possible use with the time-of-flight system.

Debugging of the 6144-channel time-of-flight analyzer has been held up by late arrival of parts. Wiring of the system subsections has been completed and testing will get under way as soon as the digital modules arrive. Equipment racks have been received, assembled, and wired.

#### Fast-Neutron Cross Sections

Work is in progress to subtract the effect of oxygen from the measured fast-neutron total cross section of Pm<sup>147</sup>-oxide. Nine new samples of different elements are ready for total cross-section measurements and material is on hand to prepare samples for 14 other elements. Some progress was made on obtaining five samples of separated isotopes from Oak Ridge. A three-dimensional visual display was prepared of the total cross sections measured on this program.

#### REACTOR DEVELOPMENT - 04 PROGRAM

##### PLUTONIUM RECYCLE

##### High Exposure PuAl Lattice Studies

The measurements in the PCTR on the three cell by three cell array of 19 rod clusters (20.6 w/o Pu-240) on an 8-3/8" pitch were begun and completed during the month. The mass of copper required to poison the central cell to a multiplication of unity was determined. Foil irradiations were made for both the poisoned and unpoisoned lattices in the case where the cadmium ratio of gold was matched in the central cell and surrounding eight buffer cells. In these irradiations the fine structure of the flux in the central cell was measured using copper foils for cell traverses and PuAl, U-235 Al, Cu, and Au foils to obtain the relative activation rates in each fuel ring, at the cell boundary and in the thermal column. The flux variations in the copper poison packets and in the fuel rods were measured in detail. The cadmium ratios for PuAl, U-235 Al, Cu, and Au were also measured in each fuel ring and on the cell boundary.

1103091

The reactivity coefficient of a rod in each ring of the 19 rod cluster was measured in both the poisoned and unpoisoned cases. In the unpoisoned case, the reactivity coefficient of the cluster was found to go down, the reactivity coefficient of a U-235 sample around the center rod was also less, but the reactivity coefficient of copper around the center rod increased by a factor of three.

The foil data for the 10-1/2 inch lattice, both poisoned and unpoisoned cases, has been processed through APDAC and checked, and is now being analyzed. The foil data for the 8-3/8 inch poisoned lattice has been processed through APDAC.

### Plutonium Recycle Critical Facility

#### a) Startup Experiments

The loading of PuAl fuel clusters into the PRCF was authorized on March 5 and the approach-to-critical was begun. Critical was achieved in 13 working days--at 1615 hours on March 21. The gross characteristics of the critical loading were: Moderator height of 93.00 inches; 25 UO<sub>2</sub> and 30 PuAl fuel clusters in approximately a two-zone loading; excess reactivity of 18 cents ( $\sim 0.84$  mk or a 51.5 sec. period) with all control and safety rods at their most reactive positions.

Twenty-four of the UO<sub>2</sub> fuel clusters were located in the innermost lattice cells, the 25th UO<sub>2</sub> was placed in one of the six outermost lattice cells.

The moderator level coefficient (at  $\sim 94$  inch water height) was found to be  $\sim 0.8$  mk/inch which is approximately half that of the FRTR. This is tentatively attributed to the PRCF control rods that extend into, and reduce the worth of, the top D<sub>2</sub>O reflector.

Equipment has been made ready for the calibration of the control and safety rods by the rod drop method.

#### b) Experiments in H<sub>2</sub>O Moderator

Assistance was provided in preparing the final draft copy of the "Plutonium Recycle Critical Facility Criteria for Light Water-Moderated Experiments." This document is completed and will be issued next month.

The document which describes the scope of the modifications to the PRCF has been reviewed and comments forwarded to the authors.

DECLASSIFIED

B-11

This document is now complete and will be distributed for approval.

Estimates have been made for the worth of the three control and safety rod combinations and on the worth of the three safety sheets. The control and safety rod combinations are expected to be worth approximately  $0.9\% \frac{\Delta K}{K}$  per rod and each sheet about  $0.7\% \frac{\Delta K}{K}$  or a total worth of approximately  $4.8\% \frac{\Delta K}{K}$ .

Three group cross-sections are being completed for calculating critical mass information for zoned loading experiments in the FRCF with H<sub>2</sub>O moderator.

c) Experiments in D<sub>2</sub>O Moderator

The programs REL35L and REL26L which have been used for kinetics studies in the FRCF have been combined into one program STEP. The program STEP combines the desirable feature of REL35L which allows calculations to be made with precursors which are not in equilibrium to that of REL26L which allows calculations to be made with a final excess reactivity of zero. A brief description of the use of this program has been prepared for distribution.

A study of the kinetics of reactivity and anti-reactivity in the FRCF has been completed. A summary of this work is being prepared for inclusion in the Physics Research Quarterly Report.

A subroutine has been written for the Generalized Least Squares Fitting Program GLEX. The purpose of the subroutine is two-fold. It will allow data from the kinetics studies in the FRCF to be analyzed by the computer more accurately and faster than the graphical methods which have been used in the past. The second use will be in fitting data from reactivity transient measurements from irradiated fuel samples.

Approach-to-Critical Experiments Using High Exposure PuAl

Experiments similar to the ones performed on the 1.8 w/o Pu-Al fuel rods are planned for 2.0 w/o Pu-Al with 16% Pu-240 content. Six hundred and fifty of the 1,000 fuel rods expected have been received. The first approach to critical will use a lattice with the fuel spaced 0.66" center to center. This lattice is being assembled in the tank in the TTR reactor room in preparation for the experiment. Additional lattices that are being prepared for this experiment have fuel rod positions spaced 0.75, 0.80, 0.85, 0.90, and 0.95 inches center-to-center.

1103093

### Neutron Thermalization - Theoretical Scattering Kernels for Water

If slightly modified Gaussian's are used to fit the energy level structure of water, the resulting auto-correlation function can be expressed in terms of simple functions. If one then follows the usual procedure of using the appropriate short and long collision time approximations in obtaining the neutron differential scattering cross section from the auto-correlation function, one obtains for a final result a double series. While this double series is more complicated than the first and second order width correction results, it is more exact, since the width corrections are mathematical approximations to the double series. Consequently, evaluation of the double series is a more appropriate test of the theory and such an evaluation is being programmed for the 7090. Should experiment and theory then show good agreement, various mathematical approximations such as the width expansions can possibly be used to reduce computer time.

### Plutonium Utilization Studies

Studies to compare PuN, PuO<sub>2</sub> vs. UO<sub>2</sub> as fuels for compact, fast spectrum space reactors are continuing. The present calculations are being carried out with the transport theory S-XI code. A wide range of reactor void fractions (20 to 80%) and tungsten cermet compositions are being investigated.

### Phoenix Fuels for Compact, Water Moderated Reactors

The work on compact, Phoenix fuel cores is continuing at a considerably accelerated pace. Life-studies for a wide range of reactor types and fuel loadings are in various stages of completion. Fuel loadings extend from 20 to 300 kg, plate thicknesses from "infinitely thin" to 0.150". Plutonium composites varying in Pu-240 content from 5 to 30 a/o are being considered.

The present methods of calculation indicate the need for considerably higher Pu-240 content fuels (~30%) than was previously assumed. Thinner fuel plates also seem indicated to enhance the effect of the Pu-240 cross section.

A variety of check calculations are under way to get more assurance on the treatment of the Pu-240 capture resonance. In these calculations, the GROUSS code has been utilized. In addition, an eight group calculation is also under way to be compared with the four group treatment presently in use.

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## FRTR - Fuel Cycle Analysis

Work to improve cross sections for the one-dimensional version of MELEAGER code continues. These improved cross sections are required to obtain better agreement between measurements of flux, burnup, and multiplication factors in the FRTR and the calculated quantities.

A  $P_3$  calculation (Program  $I_2$ ) was performed for a heavy water moderated, natural uranium-fueled reactor. IDIOT calculations for the same configuration, included in the study, "The Three Reactor Plutonium Optimization Study," predicted an increasing flux from the middle ring to the center of the 19-rod fuel cluster. The  $I_2$  calculation predicted a flux which decreases monotonically toward the center, as would be expected. Although the flux shapes do vary, the thermal utilization factor for each case is not significantly different. The calculated values are 0.95276 (IDIOT) and 0.95337 ( $I_2$ ).

## FRTR - Burnup Experiments

Burnup analysis has started on the Mark-II  $UO_2$  fuel element. This element is a concentric tube and rod type element.

Two of the instrumented high exposure mixed oxide fuel elements have been delivered to the FRTR for insertion into the reactor.

## Mass Spectrometry

The heavy-element mass spectrometer was used to provide isotopic analyses in support of Plutonium Recycle Program studies. Analyses were provided on eight samples from FRTR Al-Ni-Pu fuel element No. 5051, four samples from No. 5095, and two plutonium samples from  $UO_2$  fuel element No. 1041.

## Code Development - RBU

At this time, all known discrepancies have been resolved satisfactorily and the associated system modifications are completed. Following a final inspection of program alter decks, the system checkout will begin on the machine. In addition to the modifications, several of the more important calculations done by the system have been checked thoroughly. Two programming errors were detected during the checkout as well as some corrections to present definitions of problem data.

The Monte Carlo flow charts to be included in the RBU system document are 80 percent complete while the document itself is about 40 percent complete.

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Calculation of the diffusion coefficient continues to present some difficulties. However, the problem is one of making the transition of Monte Carlo estimates (analogous to transport theory estimates) to diffusion theory application. Several methods of computing diffusion have been derived and the limitations of each explored. It is quite probable that the expressions now available will be adequate.

Reports covering the work performed by RBU in support of resonance interference studies at Phillips Petroleum Co., Idaho Falls, Idaho, indicate that RBU Monte Carlo resonance cross section estimates are quite good.

#### Code Development - CALX

The complete CALX system consisting of programs GAM, TEMPEST, SIGMA-3C, CALX, and two small control programs chain linked together has been successfully run as a unit with CALX doing a burnup based upon the cross sections generated by GAM and TEMPEST and assembled by SIGMA-3C. Multiple TEMPEST-GAM runs have also been made to write a TAM library (combined TEMPEST-GAM output) with stacked cases which are then analyzed by SIGMA-3C. These codes have been converted to enable them to utilize the composite library tape; this conversion obviates the necessity of supplying as card input the appropriate library deck every time the code is called into memory.

Those portions of subroutine LOOP or CALX which relate to the endpoint search, either in time or reactivity, have been extensively recoded to eliminate basic logical errors and are presently 80 percent debugged.

A new code, SLOPE, which will be included in the CALX system has been coded and is 90 percent debugged. SLOPE calculates the slope of each microscopic 9-group isotopic cross section as a function of burnup when supplied with a double TEMPEST-GAM-SIGMA-3C run. The first set of these cross sections correspond to the initial concentrations, the second set to estimated end of burnup concentrations. These slopes are written on the CALX data tape and subsequently used by SPECTRUM subroutine SIGMA of CALX to do a linear interpolation to provide microscopic cross sections during burnup.

#### Code Development - RBU Cross Section Updating

The final edit of the cross sections that were to be updated on the RBU nuclear data tape is complete. Upon making the new basic library tape, the corresponding cross section libraries (i.e., GAM-1, TEMPEST, SPECTRUM, THERMOS, and MAXIE) will be made from the RBU tape. A document is being written describing the methods and references used in this compilation of cross sections.

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B-15

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Instrumentation and Systems Studies

Further PRCF hazards studies require determination of reactor behavior following accidental loss of light water coolant. The PRCF simulation has been programmed and checked out on the EASE analog computer. One of four runs which are to be made for this study was completed.

Tape recording and noise analysis equipment was specified for reactor lattice physics studies.

Specifications were completed for a high level radiation detection system for use with the cells of FRPP. One picoammeter, ordered for testing purposes, has not been received.

The printed circuit card file and detector probe were completed for the experimental final model scintillation solid state liquid effluent gamma monitor for FRTR. Detailed drawings were completed for the probes.

The complete electronic instrumentation system was established for the underwater gamma scanner for FRTR fuel rods and wire. Preliminary tests of the electronic system were favorable.

HIGH TEMPERATURE REACTOR LATTICE PHYSICS PROGRAM

The Project Proposal for design funds and its attached Summary Design Criteria were completed and circulated for approval signatures.

Work on the Final Design Criteria is progressing. A completion date of May 1, 1963, is scheduled. Criteria for the nuclear instrumentation, data acquisition, data logging, and nuclear safety systems were developed for this document.

NEUTRON FLUX MONITORS

Production Test IP-564-D has been formally approved for the irradiation of cobalt, plutonium and uranium samples in conjunction with the experimental evaluation of the regenerating detector concept. The first group of samples was charged into the central, water-filled tube of the 2B Magazine facility at KE Reactor early in the month. At the present time, the samples have received a sufficient exposure and will be discharged at the next outage. Data from this irradiation test will be used to determine the neutron flux parameters  $r$ ,  $T$ , and  $\phi$  such that subsequent regenerating detectors can be fabricated with the optimal composition for a particular environment. Fabrication techniques for construction of actual regenerating elements are being studied.

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were used as reference settings. Shim thicknesses of .002 to .006 inch were used which gave fairly linear readings from 1.1 to 3.6 volts. The d-c readout was adjusted to give readings from two to six volts with these shims. The cores were tested at one position over their full length. Only one of the 109 gave a reading that was considered larger than what would have been obtained from a .005 inch thickness of paper. All the rest of the elements tested are believed to have not more than .001 inch of oxide.

A new retractable probe for this and similar applications was conceived, fabricated and tested. Use of the new probe results in greater reproducibility of readings, and it can be inserted and withdrawn from the cores with less wear on both the core and the test coil mounting.

#### Heat Transfer Testing

Experiments are being performed to determine optimum conditions for heat transfer testing the end closures of N-reactor fuel elements. A time of three seconds from the time heat is applied is required before signals due to end-cap to core bonding defects appear at the surface where they can be detected. Variations of rotational velocity did not affect the signal-to-noise ratios. Good sensitivity to 1/4 inch diameter artificial defects positioned at nominal unbond depth in a dummy test piece was demonstrated.

Test pieces used in these experiments were fabricated by drilling longitudinal, flat-bottom holes of the desired depth in the end of Zircaloy-2 cylinders of the same geometry as N-reactor fuel elements. Since the thermal diffusivity of Zircaloy-2 is approximately equal to that of uranium, these solid Zircaloy cylinders should give approximately the same result as Zircaloy clad uranium fuel elements having defects in the end-cap to core bond.

#### Zirconium Hydride Detection

The breadboard eddy current equipment for detection of hydride in Zircaloy-2 has been further improved. Tests are under way to determine the magnitude of background variations in signal caused by both local and general differences in non-hydrated Zircaloy-2 process tube samples. Signals expected to result from 2000 ppm hydride content are about twice as great as background signal variations detected within each sample. Differences in signals from different tube samples having identical history and residual cold work, were about 1.6 times as great as the 2000 ppm hydride signal. Compensation for such background signal variations should be possible either by using direct electronic means, or by comparing signals with pre-service signal recordings. Results of tests on three process tube samples, having different residual cold work levels and histories than the

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others, indicated that cold work and history did not affect the signal as much as other factors. Fabrication of prototype eddy current hydride detection equipment suitable for testing larger numbers of process tube samples is partially complete. A commercially available hydrogen detector was evaluated, but was found not applicable to nondestructive detection of the amount of hydrogen picked up by Zircaloy-2 process tubes.

Variations in output of the breadboard circuits due to stray capacitive coupling were reduced by improvements in circuit layout. Solid state diodes, used in the phase detectors that determine real and imaginary deflections in the complex plane signal display, were found to be a source of noise and long-term drift. Replacement with vacuum diodes improved the performance of the system.

Differences in eddy current signals for different samples were calibrated in terms of apparent differences in electrical conductivity. The maximum difference between process tube samples having the same level of residual cold work and history was about four micro-ohm centimeters. Maximum differences in resistivities of two tube samples having 35% residual cold work and those having 15% residual cold work was about 1.5 micro-ohm centimeters.

## Fundamental Ultrasonic Studies

The analytical studies describing the propagation behavior of plane, continuous ultrasonic waves incident on a liquid-liquid interface are complete. Three conditions pertaining to the wave behavior at the liquid-liquid boundary were analyzed and compared. The three conditions studied were the cases of no attenuation in either liquid, attenuation present in only the refracting liquid, and attenuation in both liquids. Comparison of the reflection and transmission coefficients and the attendant phase shifts for the three cases showed only minor differences. The presence of attenuation in the refracting liquid, however, was found to change the form of the refracted waves. In the case where neither liquid was attenuating, it was found that when the incident angle exceeds the critical angle, the refracted wave becomes an inhomogeneous boundary wave with an amplitude which decreases exponentially along the wave fronts. An important difference exists when the effect of attenuation in the refracting liquid is included. It was found that in this case the refracted wave was inhomogeneous at all angles of incidence, indicating that boundary waves are not formed. The inhomogeneous refracted waves always have an exponentially decreasing amplitude along the wave fronts in addition to attenuation in the direction normal to the wave fronts. The combined effect results in a plane wave which has attenuation both in, and normal to, the direction of propagation. Experimental investigations of these wave behaviors are

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being undertaken. As a first part of the experimental work glycerine and kerosene are being separately measured for their respective acoustic properties. The fabrication of the equipment for the detailed liquid-liquid boundary studies is complete and the components are being assembled.

#### USAEC-AECL COOPERATIVE PROGRAM

##### Nondestructive Testing of Sheath Tubing

Time scheduling and program planning of the AECL cooperative program are being accelerated to ensure that the results of this program may be applied with maximum benefit to the over-all Canadian nuclear fuel program. Under the new schedule, a complete electronic and mechanical prototype test system will be completed by year's end. Test specifications and test procedures will be documented early in CY 1964, followed by a comprehensive terminal report by the end of FY 1964. An additional report discussing theoretical aspects of ultrasonic waves propagating in thin curved metal sections is planned for early 1964.

Principal experimental investigations are on small, sharply focused transducers as applied to 0.017 wall thickness tubing. To date, best results are obtained with 3/16 inch diameter spherically focused lithium sulphate crystals whose radiated ultrasonic beams are only eight mils, or about one-half the tubing wall thickness, in diameter. Under this experimental arrangement reproducible results have been obtained which substantially confirm a previous conclusion that circumferential waves propagate in a "saw-tooth" pattern rather than in a Lamb wave mode.

With the beam aligned so as to generate shear waves whose direction of propagation is forty-five degrees with respect to the tube surface, one-half mil deep notches are detected with a 10-to-1 signal to surface noise ratio.

Inner and outer surface notches are detected with nearly the same signal amplitude when the beam is adjusted such that it enters the tube surface at its focal point.

A 3 mil wide, 5 mil deep notch responds with nearly the same amplitude as does a notch 17 mils wide and 1 mil deep. Signal response as a function of defect size appears to be fairly linear, if one takes into account the effective defect area that is seen by the ultrasonic beam.

In practice, however, such a response curve is of little value since natural flaws may occur with random orientation. Therefore, other inspec-

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tion techniques whose sensitivity is less dependent upon defect orientation are being explored.

One technique being evaluated orients the beam normal to the tube surface, such that pulses of longitudinal energy are excited in the tube wall which reverberates between the surfaces for a relatively long period of time. As they emanate from the transducer, the pulses contain a wide frequency band width; once inside the metal, however, they are filtered by the tube. Only those frequencies which resonate with the wall thickness are reflected back to the transducer, hence the frequency characteristics of the decaying reverberation pattern changes with time. It is observed, however, that this reverberation pattern is characteristically altered by small defects located either on the surface or in the tube wall. Attempts are being made to interpret and utilize this change for defect detection purposes.

The second method under consideration uses two crystals, one to send a beam of shear waves at  $45^\circ$  with respect to the tube wall, and the other to receive this energy after it has traversed the tube. The geometry of the beam path makes this system relatively insensitive to defect orientation.

A third system using ultrasonic reflectors is also being evaluated. This method positions the crystals on the inside of the tube permitting the tube to serve as its own container of coupling liquid. If successful, this approach will simplify crystal alignment and mechanical scanning requirements.

A basic objective of the AECL Cooperative Program is to develop a theoretical understanding of the behavior of ultrasonic waves in tubular geometries. To this end extensive analytical efforts are being made to develop a frequency equation which describes waves moving circumferentially in a hollow cylinder. The approach is patterned after Lamb's solution to the flat plate problem, except the equations are expressed in cylindrical coordinates to accommodate tubular geometries. An exact, but extremely cumbersome equation involving sixteen Bessel function terms has been obtained. When the variable radii in this expression are allowed to go to infinity, the expression reduces to Lamb's flat plate expression, as it should. Attempts are presently being made to find methods of putting this equation in more usable form.

A second derivation by somewhat different mathematical steps, starting with the basic stress and displacement equations of elasticity, gives the same answer. The equation is lengthy, and there are several possible ways of simplifying it that are being tried. This work indicates that the fre-

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quency equation may be factorable--at least under some circumstances. When (or if) the factorization can be accomplished, it would show there are two types of waves in the hollow cylinder, which cannot coexist, analogous to Lamb's symmetrical and asymmetrical waves in a flat plate.

#### BIOLOGY AND MEDICINE - 06 PROGRAM

##### Atmospheric Physics

Analysis of atmospheric dispersion data collected over the past three years continued with emphasis on relating measurements of dispersion to meteorological variables. One of the notable results of field tests using the source near ground-level was the finding that the cross-wind dispersion sometimes increases as a power function of distance, and sometimes does not. A partial resolution of this dichotomy has been found by analysis of wind measurements taken downstream from the source. When the wind direction variance increased or decreased markedly with distance, the cross-wind dispersion was generally not a power function of distance. On occasions in which the wind direction variance remained essentially constant with distance, the power function provided a good estimate.

Previous reports have noted a lack of good correlation between diffusion parameters and atmospheric stability. Earlier attempts to stratify diffusion parameters with atmospheric stability used the Stability Ratio; however, an improved stratification resulted when the bulk Richardson number was used and the results summarized in terms of time of travel rather than distance. Correlation equations are being sought that better represent the physical processes involved in the dispersion phenomena.

In work on tracer technology, differences between calculations of the Rankin counters were studied in relation to their application to multi-tracer assay problems. The total effect on the counting rate of tracer material impacted on the filter retaining ring was shown to be less than 2 percent, even when minimal filter face velocities were used. There was no difference between counters in the retaining ring effect. The studies also showed that impaction filters could not be assayed by Rankin counter techniques.

Precipitation scavenging studies progressed through successful completion of a difficult scavenging field trial during the month. Gross counting results indicate that approximately 85 percent of the particles scavenged by the rain were larger than 3.0 microns, whereas only about 39 percent of the particles in the diffusing cloud were larger than 3.0 microns. These results further confirm the earlier finding that very small particles are difficult to scavenge with normally occurring raindrop sizes.

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Dosimetry

Study of the detection of beta rays emitted from the surface of the body continued. Two counters were made and tested. Use of the large detector of the whole body counter as an anticoincidence shield was very effective in reducing background counting rates.

Calibration of the Shadow Shield counter for measurement of Zn-65 was completed. This work was begun at the Swedish Hospital last Fall.

The study of X-ray scintillation counters continued. The effect of the earth's magnetic field on the response of the photomultiplier tubes and the amount of magnetic shielding necessary to offset it were determined. Some of the parts for a large counter were ordered.

A high voltage surge (probably from the belt charge power supply) damaged several parts of the terminal assembly of the Van de Graaff. Three days were necessary for the repairs. Aside from this the accelerator operated satisfactorily during the month. In the past, one part of the accelerator plumbing has had to be changed back and forth to suit the needs of two different classes of experiments; a study was made that resulted in a method of eliminating the need for this change; this will save time and work.

The precision long counter that was sent to Mound and Argonne laboratories for comparison studies was returned to Hanford so that we could see if any detectable change had taken place in its characteristics. To our surprise a small but detectable change had taken place. The situation is not entirely clear because two different tests indicated changes, but the changes were in opposite directions.

Two plutonium-238-beryllium sources were received. It is hoped to use their predictable rate of decay to monitor the rate of change of neutron emission of other sources and to use these very small mass sources to obtain undegraded neutron spectra. Tests indicate that the sources are mechanically stable.

The study of heat amplifier characteristic curves at dry ice temperatures was ended when it was found that to obtain appreciable amplification it was necessary to use the thermistors under conditions in which they became unstable.

Calorimetric measurement of the specific power output of one promethium-147 source was completed. The calibration of the calorimeter is being checked. The result was lower than expected so another sample will be measured.

### Instrumentation

Further calculations were made to evaluate the use of multiple solid state detectors, with moderators and foils, for neutron dose meter sensors. Results to date confirm the feasibility of this novel technique. An invention report was prepared and submitted.

The platinum fiber and conducting Teflon center rod sensor of the recharging pocket dose meter were exposed to a Cobalt-60 source for eighteen days of continuous recharging before any fiber sticking occurred. This period equates to 17,000 recycling operations or a total dose of 1700 roentgens. Several dose meters are now being assembled to conclusively test the new sensor configuration.

Methods of modulating the biology data telemetering transmitter with heart beat and respiration information have been favorably tested. In order to circumvent the need for unavailable transducers, test signals were generated which simulated the desired inputs. In addition, work was started on a breathing rate and volume measuring device to be employed in the same system.

Continuing experiments are investigating the use of thin CsI scintillations detectors for airborne alpha monitoring applications. A series of tests were made during several atmospheric temperature inversions to determine the effects of radon-thoron level changes. In addition, the detector and a pulse height analyzer were used to determine the alpha energy spectrum of several millipore filters on which plutonium was deposited. By lowering the discriminator level and widening the window width, a plutonium channel counting efficiency of 12 percent was obtained.

A second analyzer was adjusted to view the higher alpha energy radon-thoron contribution and a difference meter with alarm was connected between the count rate meters of the two channels. Further tests will be carried out when a large atmospheric inversion occurs.

Work was started regarding the three experimental in-field, real-time, phosphorescent particle detection instruments for use in air diffusion and particle transport studies.

An experimental instrument was completed which continuously measures fission product contamination on air filters under high gamma background conditions. The prototype is ready for calibration. The instrument was developed to permit detection and signaling within ten minutes for a fission product concentration of  $1 \times 10^{-9}$   $\mu\text{c}/\text{cc}$  with an air flow of 10 CFM in gamma fields exceeding 50 mr/hr. Laboratory tests have been successful.

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General detector and circuit design requirements were established for the measurement of microgram quantities of filter entrapped beryllium and experimental fabrication of the system was started. The detection head, consisting of two opposed neutron and gamma scintillation detectors, will be installed in a glove box because of the potential hazard associated with the Polonium-210 alpha source to be used for beryllium activation. A solid state amplifying and coincidence counting system will be used. Calculations indicate that less than one microgram of beryllium can be so detected on the filter.

An experimental 100 megacycle solid state amplifier was fabricated for fast counting investigations.

A novel, linear, solid-state count rate circuit was developed using a conventional diode pump modified with a transistor emitter follower. The measured linearity was  $\pm 1$  percent of full scale. An invention report was prepared and submitted.

Two final model emergency type pocket dose meters, suitable for use in gamma dose measurements from about 5 to 100 roentgens were fabricated and laboratory tests were started. The instruments use a 10 cc ionization chamber, one electrometer tube, and solid state circuits for meter indication of accumulated dose and for trip-level audible signaling purposes. The miniature exponential horn used with the audible signal portion was redesigned to provide an improved signal level with a 12 decibel improvement obtained. In addition, tests were carried out to determine the leakage resistance properties of a polystyrene capacitor. The leakage resistance was determined to be greater than  $5 \times 10^{14}$  ohms.

Redesigned detector probe light pipe shells were obtained for the reexperimental scintillation beta-gamma hand and shoe counter. The latter instrument uses automatic gamma background subtraction to improve sensitivity under relatively high background conditions. Six probes, four for hand counting and two for shoe counting, were assembled and general instrument testing started.

Methods of reducing noise in photomultiplier tubes are being investigated. Reduced noise in EM/US9536-B tubes are being obtained with anode currents of  $7 \times 10^{-11}$  amperes and 1500 anode volts by freeing the anode lead from the base material. Further tests employing vibrating capacitors are planned.

General cabling and wiring was completed for the main portion of the experimental portable mast system for Atmospheric Physics studies, and wiring was completed for the weatherproof cabinet to be located at the base of the mast. New relays were tested and installed in the thermocouple

1103105

temperature measuring circuits. One failure occurred during long-term testing of the paper tape punch coil driving circuits; the fault was located and corrected.

Wind speed and direction are being recorded from eight levels on the 400 foot Meteorology tower near 200-W Area. The data required are 5-10 second RMS deviations of the rectangular coordinate components of wind velocity from a 5-20 minute mean velocity vector. These data, now being calculated by hand, are used for maintaining climatological records and estimating the diffusive capacity of the atmosphere. Studies are being made to determine relative cost and technical merits of analog and digital methods of automatically handling atmospheric data. In addition, programs are being written to definitely establish the technical adequacy of the analog system. Recordings of wind speed and wind direction will be taken in order to simulate actual wind conditions.

#### WASHINGTON DESIGNATED PROGRAM

##### Isotopic Analysis Program

Isotopic analyses of program samples were provided at an accelerated rate during the month through the concentrated use of the vacuum-lock source mass spectrometer.

Work continued on the study of the scintillation-type ion detector using the ion test-bench. Experimental data were obtained to determine optimum design parameters. Final design of a detector for use with the mass spectrometer is in progress.

A new ion source for the mass spectrometer was designed and is being fabricated. This ion source is designed to have a higher transmission to improve the sample size sensitivity of the mass spectrometer.

A vacuum lock device to allow rapid changing of samples is being designed to increase the rate at which samples can be analyzed on this mass spectrometer.

#### TEST REACTOR OPERATIONS

Operation of the PCTR continued routinely during the month. There was one unscheduled shutdown caused by faulty bypassing technique. The experiment to determine  $k_{\infty}$  data for Pu-Al (20%  $^{240}$ ), 19 one-half inch fuel element clusters in an 8 3/8-inch graphite lattice was completed. Null reactivity measurements and foil activation traverses were made. The core is to be changed to a 6 1/2-inch graphite lattice.

Operation of the TTR was on an intermittent basis during the month. There were no unscheduled shutdowns. The TTR was made available to the University of Washington Graduate Center three nights during the month. Late in the month the TTR control and safety circuitry was transferred to the critical approach tank. Preparations are being made for a new series of runs using Pu-Al fuel, 1/2 inch rods in light water.

#### CUSTOMER WORK

##### Weather Forecasting and Meteorological Service

Consultation service was rendered on meteorological and climatological aspects of 1) oxides of nitrogen and acid mist release in 300 Area to IHO, 2) low altitude sampling of fallout to CR&D, 3) NPR Hazard Appraisal, and 4) environmental consequences of a major production accident. Climatological data summaries were made, including 1) hourly temperatures during the winter season for calculating power requirements on overhead process lines to IPD, 2) pressure and temperature data for correlating with cosmic ray data to RPO, 3) pressure data for pile atmosphere regulation to IPD, 4) hourly temperatures for 5-year period to Airefco, Inc., Portland, Oregon, 5) June and July temperature and wind data to Safeco Insurance Company, Pasco, Washington, and 6) full record of July wind speeds to cooperative observing station, Mesa, Washington. The 1963 Outlook for discharge and temperature for the Columbia River at Hanford was distributed.

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

On March 26, Hanford synoptic weather data began to be transmitted at 6-hourly intervals over the National Service "C" Teletype network. The data are telephoned to the Seattle office of the U. S. Weather Bureau for teletype transmission and for use in preparation of weather forecasts for the Tri-City Area.

##### Weather Summary

<u>Type of Forecast</u>	<u>Number Made</u>	<u>% Reliability</u>
8-Hour Production	93	85.3
24-Hour General	62	85.6
Special	170	88.2

March was considerably warmer and slightly wetter than usual. There was a high wind with gusts to 65 mph on the 28th.

Instrumentation and Systems Studies

Unsatisfactory temperature control of a uranium swelling capsule at 100-KW again caused the loss of an experiment; however, the cause of the over-temperature excursion was in capsule design and not instrumentation. This fact, coupled with a history of poor temperature control, has given impetus to a previous recommendation that the system be analyzed thoroughly and simulated on the analog computer. Computer simulation studies would provide pre-irradiation determination of optimum control settings as well as elucidate the effect of various parameter changes. Moreover, a system analysis and computer simulation would produce valuable data for use in a new capsule design.

Advice was rendered Reactor Metals Research Operation on the design of a system to measure the activation energy of various irradiated metals. The proposed system will apply a tightly controlled ramp temperature increase to a specimen located in a calorimeter and continuously measure the power to the specimen to determine and measure energy releases.

The digital control and readout system developed for use with the creep capsule micropositioner continued to perform on-line at 100-KW without incident.

Construction of the creep program 96 point data logger is continuing. Poor quality printed circuit negatives has caused some delays in construction of the printed circuit relay boards. Small pits in the copper-cladding of the finished board is the symptom.

The Hanford Test Reactor simulation contains five spatial nodes for which power and temperature are calculated. The problem consists in introducing various reactivity step and ramp functions into the five reactor regions and recording the resulting power and temperature excursions. Some early attempts to run this problem on the computer were unsuccessful due to incorrect scanner operation. Useful results have been obtained without the use of the scanner, however, and these have been given to the customer.

A task network computer demonstration problem was programmed on the GEDA and demonstrated with good results. The task network computer concept is for an on-the-job instrument which can quickly and automatically select the critical path of a number of tasks and will permit rapid assessment of the effects of changing task durations and inter-relationships. A nine-task sample problem was demonstrated on the general purpose analog computer.

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Fabrication-assembly continued on two air filter counting systems, one for Radiation Protection Operation use and one for Chemical Development for use at FRPP.

Assistance was rendered to CPD personnel regarding their interest in, and acquisition of, a coincidence-count alpha particulate air monitor as a copy of the present field prototype in use in the 308 Building.

Assistance and consultation were rendered to CPD Radiation Monitoring groups regarding the conversion of their present scintillation alpha filter counters to the coincidence-count method. This conversion would eliminate the present required long delay for radon thoron decay before plutonium contamination can be determined.

Assistance was rendered to Biology personnel regarding operation and maintenance of the portable, scintillation, solid state field analyzer and monitor.

#### Optics

Tests of the Electrical Readout Traverse Mechanism continued in the Optical Shop. After several electromicroimeters were unsuccessfully tested in an effort to properly match the sensing transducer in the ERTM sensing head, an electromicroimeter was modified such that it functions over the desired range. Linearity was improved to better than one percent of full range by adjustment of the position of the sensing transducer.

A carrying case was designed and fabricated which fits on process tube nozzles and protects the ERTM from damage due to dropping or excessive bending forces. Push rods and a second universal joint were also added to the system.

Specifications were written for two low threshold laser rods which lase in the infrared. Studies were continued to determine a workable method for using gamma rays to pump these rods.

Past month's shop work included:

1. Repair of two crane periscope heads for Purex. These heads were installed and the Purex crane periscopes were serviced and adjusted. Darkened erector lenses were replaced with clear lenses.
2. Fabrication of a complex ultrasonic sample and probe manipulator was completed for Physical Measurements Operation.

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3. Twelve quartz rings were fabricated for Radiometallurgy.
4. Four camera shutters were repaired for Radiometallurgy.
5. Twenty small lenses were made for the automatic film badge densitometer.
6. Several aluminum ceramic bearings were sectioned for Metal Fabrication Development.
7. Five quartz insulators were fabricated for Ceramic Fuels.
8. Nearly 80 man-hours assistance was given to Redox to clear up difficulties in the operation of their crane periscope.
9. Two cylinder lenses were fabricated for Ceramic Fuels.

#### Physical Measurements

Development of a two-channel prototype instrument for monitoring PRTR fuel assembly vibrations has been completed. Fabrication has begun on the driver and receiver sections of a six-channel motion analyzer which will be used, at least initially, in the 314 Building mockup facility. Discussions have been held with representatives of the Fuels Development groups for the purpose of finalizing the design of the special fuel element assembly with their "built-in" sensing coils. Vibration testing of the coils is expected to be accomplished during March to assure that these elements will withstand the vibrational compaction process. Work is continuing on the design of the readout switching and display circuitry.

A brief investigation is being conducted to evaluate the possibility of using a Model C cyclograph eddy current tester to obtain a reliable classification of fuel elements with respect to gross differences in grain size and/or heat treatment history. Test coils are presently being fabricated for this test and the evaluation is expected to be completed during April.

Work on the evaluation of micro-displacement readout systems to be used by Reactor Metals Research for in-reactor creep measurements has been completed on all systems supplied for evaluation to date. Analysis of data obtained during "Zero Shift" testing of a Physical Science Corporation Model TI-A-03 transducer used with a Model 801-D excitation and readout system, was completed and the results submitted to the customer in an evaluation report. Most of the performance characteristics for this system were unaffected by offsetting the electrical zero by as much as 0.035 inch. One significant exception was the full scale range which was found to increase about 7.5% for 0.035 inch of zero offset.

Checkout of the ultrasonic plutonium tester is complete and the equipment is presently being installed in a hood. Inspection of a plutonium specimen is expected to begin near month's end. A document, HW-76866, Ultrasonic Plutonium Tester, describing this instrument, is nearing completion.

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Equipment for making tensile measurements of specimens at elevated temperatures is being ordered. Two methods being considered are: (1) Measuring the resonant frequency of vibration to obtain Young's modulus, and 2) measuring the longitudinal and shear wave velocities of propagation to obtain Poissons' ratio.

### Physical Testing

Testing service continued to increase during the month with emphasis on K-reactor process tube preinstallation examinations. A total of 13,392 tests were made on 4,828 items representing 147,652 feet of material for a total volume work load twice that of the previous month. Services rendered to HAPO operating departments remained steady at thirty-two, not including two AEC contractors.

Tests were completed on all K-reactor process tubes on hand. Van Stone flange work is being performed by reactor personnel prior to the final inspection.

Field activities included magnetic particle inspection of PRTR eye-bolts, fluorescent penetrant examination of converted fuel element nozzles, zirconium tube Van Stone flanges and Parker fittings on the rear face of 105 DR. Vibration tests were made on PRTR process tube nozzles. Radiographic work was performed on assorted piping in 100-B, 100-D, at the PRTR, 1706 KER cells, 165 KW valve pit and 1240 building. In addition, radiographic services were rendered on the PRTR overhead crane, and on the air receivers in 100-F Area. Thickness measurements were made on raw and filter water lines in 100-D and on one trailer transport tank.

Modification of the ultrasonic test system was completed and testing of 100 tube lot of 0.422 inch O.D. Zircaloy-2 tubes continued. Ten of 38 tubes were rejected on the basis of signal indications exceeding the amplitude of reflections received from a one mil standard. All indications were verified visually. Modification to the Curtiss-Wright Immerscope was made to allow higher test speeds for the longitudinal test. A Sanborn 650 recorder was installed to record high frequency ultrasonic information.

A technique was developed to bond test co-extruded aluminum clad, aluminum-plutonium fuel cores which are scheduled for the C-1 loop. Mechanical problems, which had been limiting factors in previous test applications, were eliminated with the application of a specially designed weir test tank.

Specimens and specimen holders for uranium oxide-tungsten vibrapacked materials are being designed for the purpose of performing compression, tensile and torsion tests at high temperatures.

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Linear motion radiography development was completed and tests were made on a 12-foot aluminum GERT Graphite Irradiation capsule. This application insured proper alignment of swage locking rings and other internal components.

#### ANALOG COMPUTER FACILITY OPERATION

Analog computer problems considered during the reporting period included:

1. NPR Pressurizer-Injection System
2. NPR Gas Coolant Flow
3. HTR Hazards Analysis
4. PRTR Hazards Analysis
5. Optimization Example Problem
6. Task Network Computer
7. 100-D Reactor Two-Dimensional Simulation

Bids on the new analog computer were received and reviewed during the month.

Eighty-three percent of the GEDA and eighty-eight percent of the EASE equipment were in good operating condition this month. Computer utilization was as follows:

<u>GEDA</u>	<u>EASE</u>	
192	208	Hours Up
32	32	Hours Scheduled Downtime
<u>32</u>	<u>16</u>	Hours Unscheduled Downtime
256	256	Hours Total

Since the computers are new in use on a two-shift basis, a third maintenance shift was added. This action has improved the general reliability of the computers.

#### INSTRUMENT EVALUATION

A majority of the evaluation tests were completed on the experimental solid state portable radiation survey instrument which employs a rechargeable nickel-cadmium battery and both an aural signaling circuit and a multi-range count rate meter. The unit can be operated 60 hours following a recharge, and performance is acceptable from minus 10°F to plus 130°F, although some improvement in reading error may be required below 0°F to be fully satisfactory. The instrument can employ as detectors regular GM tubes, miniature halogen-quench GM tubes to extend the range to 500 mr/hr or more, alpha scintillation probes, and various scintillation and

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proportional counter neutron detector probes. Reliability and versatility are emphasized.

The complete drawings were updated and modified for the scintillation alpha-beta-gamma solid state hand and shoe counters. Seven are being fabricated for Radiation Protection Operation by a commercial instrument fabricator. A trip was made to the off-site plant to observe the work progress and tests being done.

The drawings for the 2 inch by 7 inch, plastic housing, scintillation alpha detector probes were checked and revised so Radiation Protection Operation can secure about \$5,000 worth from an off-site fabricator before July 1.

General acceptance-evaluation tests were completed on a production prototype solid state scintillation alpha "poppy" portable instrument. Thirty of the units are being made by an off-site commercial instrument fabricator for Radiation Protection Operation.

Evaluation testing was started on a Victoreen Model 440 portable, vibrating reed type, beta-gamma dose rate meter.

*RS Paul*

Manager

PHYSICS AND INSTRUMENTS LABORATORY

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## CHEMICAL LABORATORY

### RESEARCH AND ENGINEERING

#### FISSIONABLE MATERIALS - O2 PROGRAM

##### IRRADIATION PROCESSES

###### Effluent Water Radioisotope Studies

The test comparing zirconium-clad fuel elements in deionized-water cooled aluminum and zirconium reactor tubes was continued. Although a steady state condition may not be completely established as yet, steady state values should not differ much from the present values. In the all-zirconium system the Cu-64 and Np-239 concentrations were down a factor of 2.5, Mn-56 was down a factor of 4, As-76 down a factor of 5, P-32 down a factor of 9, and Na-24 down a factor of 40, compared with the same zirconium tube with aluminum-clad fuel elements. The aluminum tube with zirconium-clad fuel elements had P-32, As-76, Np-239 and Cu-64 concentrations essentially unchanged, Mn-56 down a factor of 2, and Na-24 down a factor of 2.5, compared with values obtained with aluminum elements.

The half-reactor test at 100-F Reactor comparing high-alum treated water with low-alum treated water attained test conditions of 18 ppm alum vs. 6 ppm alum on February 18, 1963. Since that time the Ga-72 concentrations in the effluent from each side have differed by less than 6 percent while the Na-24 concentrations have shown no difference. Insofar as these two radionuclides are indicators of corrosion rate, the results indicate no significantly different corrosion rates as a function of the different amounts of alum used in the water treatment processes.

Operation of the water treatment pilot plant has continued to be plagued with instrumental and other operating difficulties. Because of erroneous instrumentation, water flow measurements of the water treatment pilot plant were found to be high by about 30 gpm. Since sufficient water from this plant was not available for the two tubes, service water was added to the clear wells. As a result of this, the suspended solids concentration in the clear well was two to three times that of the filter effluent and effectively compromised the test. On March 4, the aluminum baffle which separates the filter into two sections was removed. It was found that the sand had again shifted, resulting in uneven filtering efficiency. Redistribution of the sand in this fashion resulted

1103114

in increased filter effluent turbidity. During this period zeta potential measurements were made periodically on all shifts which should allow better control of the process.

The search continued for a coating which would effectively reduce the corrosion rate of aluminum surfaces exposed to reactor cooling water. Ten new candidate inhibitor compounds were added to 95 C water passed over aluminum turnings at 3 ml/minute for several days, following which As-76 adsorption measurements were made. Two of these compounds showed promise. Rust-Lick P7 and Rust-Lick C both reduced adsorption by a factor of 10 when used at 100 ppm. These two compounds are organic, water dispersible compounds. Further studies to determine the effectiveness of lower concentrations of these materials will be made.

#### Effluent Monitor

The iodine monitor was placed back in operation after a prolonged outage due to a failure of the sample supply pump. Samples of the effluent were submitted for iodine analysis for calibrating the monitor. An automatic sampling device is being designed which will collect samples during indicated high iodine activity such as might be expected during a rupture.

#### Release Studies

The experiments to determine the release of Po-210 from irradiated, aluminum-canned bismuth cylinders were started. Tests are being run in an air atmosphere at 1300 C for various periods of time. Preliminary results indicate that the release of polonium will be significant even for very short heating periods.

#### SEPARATIONS PROCESSES

##### Disposal to Ground

Water samples were obtained and analyzed from eight farm wells located on the Columbia Basin Irrigation Project just east of the Hanford Project. These wells are believed to be of sufficient depth to tap a deep confined or semi-confined aquifer known to exist near the plant shore of the Columbia River, if, in fact, the formation does extend into the locality of the wells. Analytical results to date give no indication that contaminants of Hanford origin have entered the wells. The maximum gross beta concentration observed, 20 femtocuries per cc, was identified by gamma energy scan to be natural-occurring K-40. Nitrate ion concentrations

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were less than 3 ppm, and the tritium content was less than 2 picocuries per cc. Additional samples will be obtained in the future to detect possible changes in concentrations.

All vertical monitoring wells in the 241-A tank farm were logged with the highly sensitive gamma scintillation probe on March 15, 1963, to determine changes in location and magnitude of above-background (nominally 1100 cpm) counting rates previously observed. In addition, the probe was lowered down the stairwells of the two monitoring caissons. Little change was noted since the wells were last logged on February 7, 1963. Well No. 11, not previously available for monitoring, showed a counting rate of 43,000 cpm above background at the 20-foot level; this well passes within several feet of the cascade pipe between the 105 and 106 tanks at the 20-foot level. Both caissons showed counting rates of about 4,000 cpm above background within several feet of the bottom (~68-foot level), and the No. 2 caisson, located southwest of the 106 tank, had a counting rate of 48,000 cpm above background at the 16-foot level, the same elevation as nearby waste transfer pipes. The accumulated data are being analyzed for their significance.

Core sampling of the 216-Z-9 Recuplex crib was continued using split drive-barrél samplers in place of the unsuccessful rotary core barrels. To date, continuous 6-foot cores have been obtained from four of the eight planned sampling locations. The cores are being processed by Chemical Processing Department analytical services personnel to evaluate the deposition of plutonium with depth.

## Redox Process Water Treatment

The study of the Redox demineralizer system was completed. The primary recommendation, contained in the report to be issued, is to replace the degraded strong-base anion resin with a weak-base anion resin.

## Use of Uranium(IV) in the Purex Partition Column

Further studies were made on the rate of oxidation of plutonium(III) to plutonium(IV) in the scrub section of the Purex 1B column under conditions expected when using uranium(IV) as partitioning agent. Results from experiments made at 25 C were reported last month (HW-76596 C). Further studies concerned the effect of temperature since the plant column operates in the 35 - 45 C range. Experimentally, an aqueous phase simulating that in the scrub section and with plutonium initially all plutonium(III) was contacted with an organic phase pre-equilibrated with nitric acid to simulate the

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scrub section organic. The plutonium distribution ratio was measured as a function of contact time and used as an indirect measure of plutonium oxidation. After 30 minutes' contact time (estimated aqueous residence time in the Purex 1BX column) at 45 C, the distribution ratio ( $E_a^0$ ) for plutonium from an aqueous solution 4 M in nitric acid and containing ferrous sulfamate (present operating conditions) was 0.09. With 0.02 M hydrazine instead of ferrous sulfamate the  $E_a^0$  was 1.1. For an aqueous 2 M in nitric acid and 0.02 M in hydrazine, the  $E_a^0$  was about 0.6. These data indicate that, at 45 C, significantly greater reflux of plutonium will occur in the 1BX column with hydrazine as the only reducing or holding agent present than occurs with ferrous sulfamate present.

### FUEL PREPARATION PROCESSES

#### Uranium Surface Electrochemistry

Continued use has been made of EMF measurements of uranium as an index of the cleanliness and activity of uranium surfaces. This index has been applied to a study of variables of importance in the desmutting step following anodic etching in 5.4 M  $H_3PO_4$  - 0.28 M HCl.

Rates of desmutting and passivation of uranium were measured in 10 and 12 M  $HNO_3$  at 25 C and in 6 and 8 M  $HNO_3$  at 40 C. The rate of passivation of the desmuted surface was found to be the same in all four cases (0.005 volt/min) although the rate of desmutting increased with temperature and nitric acid concentration: 8, 3, 7 and 4 minutes, respectively, being required for the attainment of clean surfaces. The clean surfaces obtained at 25 C were more active (-0.37 volts vs. saturated calomel electrode in 0.5 M  $NaClO_4$  at pH 5.5) than those obtained at 40 C (-0.30 volts).

The effects of increased anodic etch time and of the concentration of nitrite in the desmutting solution on the behavior during desmutting in 8 M  $HNO_3$  at 40 C were investigated briefly. Increasing the anodic etch time from 6 to 20 minutes (at a current density of 55 amp/ft<sup>2</sup>) had no effect on the behavior during desmutting. Addition of 0.1 M urea (to consume the nitrite initially present and that formed during the reaction) did not affect the rate of desmutting but did increase the rate of passivation by a factor of 2.

### WASTE MANAGEMENT AND FISSION PRODUCT EXTRACTION

#### Trilauryl Amine Extraction of Neptunium and Plutonium from Purex LW Solution

In the present study flowsheet, hydrazine is added to LW to reduce

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neptunium(V) to neptunium(IV). Neptunium and plutonium are co-extracted into an equal volume of 0.3 M trilauryl amine (TLA)-Soltrol and co-stripped with one-half volume of 0.1 M oxalic acid. Successful reduction of neptunium(V) to neptunium(IV) and subsequent extraction was accomplished by adding hydrazine to an intimate mixture of extractant and LWW solution. This procedure may be necessary in plant operation.

Destruction of oxalic acid in the strip solution with nitric acid is planned in a plant test of the recovery procedure. The reaction of oxalic acid with nitric acid in the 7 - 11 M range was found to be first order with respect to oxalate concentration. About six hours were required to reduce oxalate concentration from 0.01 to 0.001 M in boiling 10 - 11 M  $\text{HNO}_3$ .

The solubility of TLA in the strip solution (0.1 M oxalic acid) was found to be negligible when measured by a technique developed previously at ORNL (ORNL-1734). Contamination of Purex solvent by TLA due to the solubility of TLA in the oxalate strip solution does not appear to be a problem. However, because of the possibility of introducing TLA into Purex solvent via entrainment of extractant in the oxalate strip, studies are in progress to determine the effects of small amounts of TLA on the performance of Purex solvent. Plutonium retention tests indicate that up to one volume percent TLA in the Purex plant solvent will not impair plutonium stripping in the 1B column.

#### CSREX Process-Laboratory Studies

Further tracer-level mixer-settler runs were made testing CSREX Flowsheet No. 2 conditions. Under extraction column conditions, chromium decontamination factor was only 18. This is lower than expected since chromium decontamination factors greater than 1000 have been obtained previously in the D2EHPA flowsheet (BMBP not present). Yttrium loss in the extraction column was about seven percent; less than 0.5 percent of the yttrium was stripped in the partition column. Behavior of yttrium under strip column and solvent washing conditions is currently under study.

#### CSREX Process-Engineering Studies

An unsuspected degradation reaction of the CSREX solvent was observed during a routine 1C column run. After about one hour of operation using a stripping solution of 1 M  $\text{HNO}_3$ , the solvent began to darken and soon became a very dark reddish brown. A trace of  $\text{NO}_2$  gas was observed in the vapor space of the column. No reaction

1103118

was observed in either a preceding or a following run using 0.5 M  $\text{HNO}_3$ . The BAMBP concentration in the degraded solvent was found to have decreased from the nominal 0.5 M to 0.14 M, confirmed both by analytical titration and by the cesium distribution ratio.

Laboratory studies and a review of the pilot plant solvent history suggest that the degradation was caused by nitrous acid, apparently introduced into the system by a synthetic alkaline waste supernate heel left over from earlier supernate-BAMBP runs (see the January monthly report, HW-76315 C).

It has been postulated that the degradation involves a two step reaction: first with nitrous acid to produce a nitroso compound and then with nitric acid to form a nitro compound, releasing nitrous acid for further reaction. This explanation agrees with the apparent auto-catalytic effect observed in the pilot plant and laboratory studies. Nitrous acid removal techniques are currently being sought as a means of protecting the solvent from degradation.

#### BAMBP Extraction of Cesium - Laboratory Studies

High stripping column losses were experienced in previous pilot plant pulse column studies on the use of 0.75 M BAMBP-Soltrol to remove cesium from synthetic stored waste supernate. The solvent was suspected to contain some dipicrylamine (DPA)-nitrobenzene from earlier studies and cesium loss was attributed to reduced cesium stripping rate because of the DPA. Recent batch contact studies with laboratory solvent showed that the presence of a small amount of DPA-Soltrol in BAMBP-Soltrol does not alter the cesium stripping rate. Cesium stripped as rapidly from the pilot plant solvent as from the laboratory solvent but the pilot plant solvent retained more cesium, perhaps due to solvent degradation products.

#### Treatment of Purex Stored Waste Sludge

Efforts toward developing a flowsheet for the treatment of Purex process stored waste were continued. Experiments this month emphasized leaching the sludge with complexing agents such as citrate, tartrate and EDTA in an effort to remove strontium without dissolving the iron. The sludge was leached first with water to remove much of the sodium salt content. The remaining sludge was leached with a solution containing the complexing agent. Analytical data are available only for citrate leaches. Complexant concentration, leaching time, temperature and final pH were variables studied. Maximum strontium removal obtained was 75 percent; about seven percent of the iron was also dissolved. It is of interest that about

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15 percent of the strontium in the sludge is very resistant to leaching. Even hot 6 M  $\text{HNO}_3$  fails to remove this strontium.

## Waste Packaging

Experiments were conducted to provide data for estimating the magnitude of off-gas clean-up requirements in a waste packaging system employing granular inorganic ion exchange media. In the system, a loaded zeolite bed is to be dehydrated by passing hot, dry air through it at about 20 bed volumes per minute. Some fines can be expected to be carried into the off-gas lines during this dehydration step.

Three different zeolites were evaluated at 800 F with a gas flow rate of 20 scfm through a one-cubic-foot, 8-inch diameter bed. Isokinetic off-gas samples were taken on Millipore filters over a 17-hour period. Low off-gas dust loadings of  $1 \times 10^{-5}$ ,  $6 \times 10^{-4}$  and  $9 \times 10^{-5}$  grains/cu.ft were obtained for Linde 4A, clinoptilolite and Linde 13X, respectively. Particle size distribution information indicated that the synthetic zeolites are more stable than the naturally occurring clinoptilolite.

## Cesium Removal from Alkaline Supernates

Revised estimates of high level supernatant waste compositions for A, SX and S tank farms were made, based on recent analytical results. The revised composition for A farm supernates is based on a stored waste volume of 150 gallons per ton of uranium and includes a small expected dilution. The sodium concentration of this waste is 4.4 molar, compared to the 9.0 molar value that has been used previously. Ion exchange loading of cesium on Linde AW-400 based on the new waste composition is about double that observed for the old composition, and flow rates can also be doubled. Breakthrough of cesium to 1 percent was 50 column volumes at a flow rate of four column volumes per hour.

A 1:1 dilution of SX farm supernatant waste is used which brings the waste volume to 1000 gallons per ton of uranium. The 1 percent breakthrough for a synthetic waste of this concentration was found at 8, 17, 20 and 22 column volumes for clinoptilolite, AW-400, Duolite C-3, and AW-500, respectively, at a flow rate of two column volumes per hour. Ninety-five percent of the cesium was eluted from the column by 4 M  $(\text{NH}_4)_2\text{CO}_3$  with 4, 10 and 15 column volumes for Duolite C-3, clinoptilolite and AW-500, respectively.

Cesium loadings for S farm synthetic supernatant waste are similar to those for SX farm wastes except the Duolite C-3 shows the highest loadings.

1103120

Samples of 10 to 60 mesh beads of clinoptilolite received from Minerals and Chemicals Phillip Corporation were tested with a synthetic FTW waste. Cesium loading was about 20 percent below that of crushed particles of clinoptilolite, reflecting dilutions by the binder. Loading kinetics were good and about the same as 20 to 50 mesh particles. The mineral was not acid-treated before beading, and gassing with some fracturing occurred when the acid waste contacted the exchanger. This type of product should result in less attrition and fines production than crushed particles.

#### Fission Product Packaging

Samples of Linde 13X and AW-400 loaded with Cs-137 and 13X loaded with Sr-85 were heated at 440 C with continual air flow for 17.5 hours. This is the same temperature but a longer time than is estimated for drying the loaded storage columns. The amount of Cs-137 or Sr-85 vaporized during the heating period varied from 0.01 to 0.1 percent.

#### Storage of Cesium and Strontium on Zeolites

Preparations were completed during the month for full-level B-Cell testing of the Hanford Waste Management zeolite storage concept. Major areas of uncertainty which the hot-cell experiments are expected to resolve are (1) the extent and severity of the pressure build-up due to radiolytic gas production, and (2) the effect, if any, of radiation on efficiency of adsorption of the radioisotope and on ease of retrieval from the bed. In the first run, one mole of fission product strontium containing 7500 curies of strontium-90 will be adsorbed on an instrumented bed of Linde 4A approximately 2.5 inches in diameter by 14 inches high, the bed will be dried with heated dry air using the proposed plant drying cycle, then sealed and heated to the temperature (600 C) which decay heat will generate in the large units. This bed will be maintained at this temperature for a prolonged storage and observation period and then eluted. Later experiments will simulate failure of the air drying equipment or of the air heater (conceivable plant operating errors) and determine severity of the consequences.

Ex-cell testing of the experimental cartridges revealed several design deficiencies. Using an axial heater and 3/4-inch of external Tipersul insulation, the temperature drop (200 C) across the bed was excessive, suggesting either that the Tipersul is a poorer insulation than claimed by the manufacturer, and/or that the zeolite is a better insulator than expected. The latter, if true, would have an adverse effect on the allowable size of plant-scale containers.

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Unfortunately, the experimental arrangement used was not suitable for quantitative evaluation of the two materials. The design was modified to use an external copper block jacket heated with two Calrod heaters (with the entire assembly surrounded by 1-1/2 inches of Thermaasbestos pipe insulation). Temperature gradients at a bed temperature of 600 C were reduced to an acceptable 10 - 20 C.

## Denitration of Purex Waste

Tests were completed on the use of several Moses Lake crude beet sugar concentrates and Hawaiian cane sugar molasses as substitutes for refined sugar. All gave similar results and were fully equivalent to sucrose. Use of the molasses would reduce reagents costs by an estimated factor of 2.6. A possible disadvantage of the crude concentrates, if strontium-90 is to be recovered from the denitrated waste, is the presence of traces of calcium (which would reduce the capacity of the DZEHPA process). Trace calcium should, however, be readily removable from the syrups by fairly simple treatment with a cation exchange resin.

## Promethium Shielding Measurements

A lead shielding curve was determined experimentally by measuring the radiation dosage from a 1270 curie Pm-147 source through various thicknesses of lead. Results were in excellent agreement with calculation. Most of the observed dose was from the 0.9 millicuries of Pm-148 and 0.1 millicurie of Pm-146 contained in the two-year old material. The experiments will be repeated after an additional period of cooling.

In other experiments, the oxide formed by the 750 C ignition of promethium oxalate was shown, by weighing and titration, to be accurately represented by  $Pm_2O_3$ .

A paper, "Detection of Pm-146 Among the Products of Uranium Fission," by Roberts, Wheelwright and Van Tuyl has been submitted to the Journal of Inorganic and Nuclear Chemistry.

## EQUIPMENT AND MATERIALS

### Low Flow Metering Pumps

Design, fabrication and testing of two low flow metering pumps have been completed, and the pumps have been sent to the plutonium processing plant for installation. The pumps employ a "dipper" principle for removing precise quantities of liquid from a reservoir.

1103122

Control of rotational speed, tube size, and reservoir depth provides a range of flows from 28 to 65,000 ml per hour. Using direct contact adjustment, flow reproducibility of  $\pm 0.02$  percent was obtained. With remote control, the reproducibility was  $\pm 0.5$  percent. The simplicity of design and absence of wearing parts in the pump itself result in a very rugged metering device with a long life expectancy and virtually no maintenance. A current limitation of this type of pump is the fact that it does not deliver fluid against a back pressure. For many process uses this is not a requirement; however, some additional work is planned to develop a suitable modification allowing use of the pump in a greater variety of applications.

#### Anodic Passivation Studies

Studies on the anodic passivation of 304-L stainless steel in  $\text{NH}_4\text{F-NH}_4\text{NO}_3$  solutions show good promise that the technique can be applied to large scale equipment to reduce corrosion of 304-L during Zirflex decladding. Potential versus current curves were determined for 304-L in 4 M  $\text{NH}_4\text{F}$ -0.5 M  $\text{NH}_4\text{NO}_3$ . Active-passive behavior was shown with a critical (Flade) potential and current of -450 mv (vs. SCE) and 1 ma/sq.in., respectively. The passive film formed rapidly; about 0.01 coulombs/sq.in. was required to form the film. The film dissolved rapidly when current was interrupted.

Corrosion of 304-L (acting as a heat transfer surface) in boiling 4 M  $\text{NH}_4\text{F}$ -0.5 M  $\text{NH}_4\text{NO}_3$  was determined as a function of applied potential. Corrosion rates under anodic control were about one-tenth those in the absence of anodic control. Effective potential range was about 300 mv. No active-passive behavior was found with 304-L in  $\text{NH}_4\text{F}$  solution without  $\text{NH}_4\text{NO}_3$  present.

#### Non-Metallic Materials

Chemical compatibility tests were run on nine different commercially available rubbers (seven silicone, one nitrile, and one neoprene) to determine their stability in nine typical process solutions. These rubbers have been tested previously for radiation tolerance. One of the silicone rubbers had exceptionally good stability in all nine solutions. The other rubbers showed the usual pattern of good stability in some solutions and failure in others. Two heat insulating materials were tested in  $\text{HNO}_3$ -HF vapor at room temperature and at 50 C. Both showed good stability at both temperatures.

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PROCESS CONTROL DEVELOPMENT

Control System for New Plutonium Reclamation Facility

Specifications for control system components required to adapt the Gradient Control System to the new Plutonium Reclamation Facility were prepared and the equipment has been placed on order. Development of the control system for the air pulser is underway with emphasis on the evaluation of various methods of bi-directional flow measurement and the design of circuitry to reduce the average flow value to a signal compatible with conventional D.C. control system inputs. Results thus far indicate that the use of bi-directional strain gage differential pressure transducers will require a variable gain pre-amplifier. The pre-amplifier must have high common mode rejection, low drift and less than 10  $\mu$ V noise rms referred to the input in order to operate over the range of input signals with acceptable accuracy for low amplitude-frequency products.

Measurement of column density has been studied experimentally to determine how it is affected by pulse amplitude and frequency and by the net flow through the column. On the basis of the study a system is being designed to apply a correction to the observed column density such that control is maintained by a value more accurately analogous to dispersed phase holdup.

Plutonium Detection Test System

A plutonium detection test system has been devised and fabricated and is being installed in two adjacent hoods in the 222-S Building Laboratory. The purpose of the system is to flow plutonium solutions of known concentration through various in-line detectors to facilitate their evaluation and calibration. Two detectors have been designed and are included in the system for initial testing. The first device monitors the intensity of the 17 kev X-rays from plutonium; the second uses scintillating glass for detecting alpha particles. Initial emphasis is the in-line detection of low levels of plutonium (0.1 mg/liter and above) in process streams. Provisions have been made for decontaminating the detectors in place. Procurement of all necessary instrumentation has been completed.

Calorimeter Studies

Several calorimetric methods have been investigated to evaluate their potential for measuring heat generation rates in fission product solutions. The following minimum heat rates have been measured using various techniques: (1) temperature rate of rise in an insulated

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1103124

container, 0.2 watt per liter, (2) temperature difference between a bare and a covered temperature sensor in an insulated container, 0.4 watt per liter, (3) continuous-flow calorimeter, one watt per liter. The choice of any one technique will depend on the particular application. In these tests the objective was to find the lower limits and for this reason thermistors were used.

In the continuous flow calorimeter, temperatures were measured at the inlet and outlet of an insulated section of line while heat was added electrically to simulate the fission products. Heat rates of one watt per liter and above were measurable with the test apparatus used, and the sensitivity could be improved by enlarging the insulated volume being measured. In the range of one watt per liter to ten watts per liter the thermistors were flow sensitive and picked up any small temperature variations in the inlet stream, particularly when measuring heat rates near one watt per liter.

#### C-Column Mathematical Model

The second block of 16 runs was completed to provide experimental data for refinement of the C-Column mathematical model. Feed preparation and routine instrument maintenance was undertaken for the final block of the 16 runs. An integral part of the model is its dependency upon  $\epsilon$  (the organic volume fraction) as a function of distance up the column. This function is determined experimentally in the current series of runs. A method of smoothing the data for use in the model was necessary. It has been determined that an expression which is cubic polynomial in distance serves adequately to represent the data within the inherent experimental error. The polynomial has been fit by least squares to all the data obtained so far to obtain the desired smoothing.

#### ANALYTICAL AND INSTRUMENTAL CHEMISTRY

##### Determination of Traces of Thorium-230 in Natural Thorium

As little as 0.2 part of Th-230 per million parts of natural thorium was determined by alpha energy analysis using a low background silicon diode detector. The thorium was prepared for counting by electroplating approximately 100  $\mu\text{g}$  of thorium on platinum discs. The ratio of the Th-230 to Th-232 was calculated from the relative abundances of the 4.6 - 4.7 Mev alphas from Th-230 and the 3.95 - 4.0 Mev alphas from Th-232 and their known half-lives. The ratios found on the four samples tested ranged from  $0.17:10^6$  to  $83:10^6$ , grams Th-230:grams Th-232. The low ratios were beyond the range of current mass spectrometric equipment.

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## REACTOR DEVELOPMENT - O4 PROGRAM

### PLUTONIUM RECYCLE PROGRAM

#### Salt Cycle Process

Absorption Spectra of Plutonium in Molten Chloride Salt Solutions - Spectrophotometric studies have proven the existence of plutonyl(VI) in molten chloride salt solutions under certain conditions. Stable solutions whose spectra are in close agreement with published plutonyl(VI)-chloride spectra were obtained by sparging 1.4 LiCl-CsCl solutions containing 0.0103 M plutonium with 33 percent O<sub>2</sub> - 67 percent Cl<sub>2</sub> at 550 C. These solutions also contained a minor amount of plutonyl(V) but no detectable amounts of plutonium(III) or (IV). In a similar experiment at 700 C, a higher concentration of plutonyl(V) was formed and a significant portion of the plutonium precipitated (presumably as PuO<sub>2</sub>). The use of a 100 percent O<sub>2</sub> sparge at 550 C resulted in the formation of a precipitate (presumably PuO<sub>2</sub>) rather than the formation of a plutonyl(VI) solution.

On sparging plutonyl(VI) solutions with chlorine alone, no spectral changes were observed (in 20 minutes) at 400 C but, at 550 C, the concentrations of plutonium(V) and (VI) decreased and plutonium(IV) appeared in the solution. Helium sparging a plutonyl(VI) solution at 400 C resulted in a rapid decrease in plutonyl(VI) concentration and increase in plutonyl(V) concentration. A subsequent HCl sparge resulted in rapid conversion of the plutonyl(V) and (VI) to plutonium(IV).

The extent of conversion of plutonium(III) to plutonium(IV) on saturating 1.4 LiCl-CsCl solutions with chlorine was found to be at least 95 percent at 400 C, 75 percent at 550 C, and 40 percent at 700 C. These values are significantly higher than those measured previously in 1.4 LiCl-KCl solutions, as expected from the trend observed in uranium studies that the complexing power of molten chloride solutions increases with the size of the solvent cation.

The Synthesis of Uranium Sulfides in Chloride Salt Melts - Exploratory work on the synthesis of refractory uranium compounds in molten salt media has given promising results. A crystalline appearing material prepared by H<sub>2</sub>S-sparging a chloride salt melt containing uranium(IV) has been identified by X-ray diffraction as nearly pure UO<sub>2</sub>S. A similar material prepared by H<sub>2</sub>S-sparging a uranium(III-IV) melt proved to be a mixture of UO<sub>2</sub>S and US<sub>2</sub>. The parameters governing the composition of the products of this reaction are not yet identified.

1103126

Decladding-Oxidation Testing - Evaluation of vibratory decladding and  $UO_2$  to  $U_3O_8$  oxidation continued with decladding of ten 1/2-section fuel rods with a total of 30 pounds of  $UO_2$  being discharged to the receiver-oxidizer. The  $UO_2$  was subsequently oxidized to  $U_3O_8$ , and dumped from the oxidizer. The average losses of  $UO_2$  to the cladding from seven half-section cold swaged rods and three half-section vibratory compacted rods were 0.072 percent and 0.014 percent, respectively. Each half-rod required only one knock-out pass, and an average of two minutes were spent decladding each half-rod.

Precipitation Studies - Additional study of the precipitation of  $CeO_2$  from a 2.5 LiCl-KCl -5 to 10 w/o U melt has indicated that at least 50 percent of the  $CeO_2$  present in the salt solution can be recovered on a bed of quartz chips supported in a quartz container by circulating the fused salt through the bed with a  $Cl_2-O_2$  gas lift. Approximately 80 percent of the  $CeO_2$  was precipitated under conditions where plutonium precipitation of 90 percent and greater has been reported. Breakage caused by attack on the quartz during  $CeO_2$  precipitation makes it necessary to replace the filter apparatus after two runs.

Crushing of Electrolytic  $UO_2$  - The electrolytic  $UO_2$  or  $PuO_2-UO_2$  from the molten salt baths in the Salt Cycle Process must be crushed and sized to certain particle size fractions to be used as feed material to vibratory compaction. An approximate composition of 60 percent -4 +10 mesh, 20 percent -35 +65 mesh and 20 percent -200 mesh is the present goal in crushing experiments.

Crushing of electrolytic  $UO_2$  with parallel plates mounted in a hydraulic press has given products with the desired particle size distribution.

#### RADIOACTIVE RESIDUE PROCESS DEVELOPMENT

##### Eighteen-Inch Radiant-Heat Spray Calciner

Five runs were made in the Cold Semiworks radiant-heat spray calciner to explore capacity and mercury volatilization characteristics. The calciner contained a 13-3/4 inch diameter by 7-foot long induction tube and employed a cyclone and filter unit for primary off-gas treatment. The major findings include: (a) The majority of the mercury was volatilized from the calcine at temperatures above 450 C but re-deposited on the calcine in the off-gas train in zones with temperatures below about 300 to 350 C. No mercury containing deposits were visible on piping.

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(b) Using air as the atomizing gas, heating the feed stream to near boiling increased the water capacity of the unit by 40 to 50 percent over that measured with 18 C water. Since this increase is much greater than the sensible heat increase of the feed, the effect is probably caused by changes in the atomization characteristics or by an improvement in the convective heat transfer.

Following the aforementioned runs the off-gas train was modified by eliminating the cyclones. The filter unit was rebuilt reducing the number of filters to five and relocating the unit so that the calcine blown off the filters would fall into the single pot below the calciner proper. The blow back system allows each individual element to be blown back under programmed conditions. The programmer employs a modification of a circuit originally developed for filters in plutonium processing hoods. Modifications include incorporation of a variable time spacing between the blow back cycles and adapting the original circuit for use with a greater variety of output relays. The variable spacing between blow back cycles was obtained by actuating the cam timer with an added reset timer. A silicon controlled rectifier (SCR) was added to the time delay circuit and is used to pull the timed relay. The SCR makes the choice of an output relay much less critical with regard to sensitivity, coil resistance, and operating time.

The programmed air pulse is discharged into a venturi on the filter element. Each element is equipped with a venturi of a different design. In the one 10 GPH run made in the modified calciner adequate blow back of every filter element was obtained with an 0.2 second air pulse per element every 90 seconds. The air pressure was 40 psig.

#### Synthesis of a Clinoptilolite-Like Zeolite

Initial work was completed on the synthesis of a clinoptilolite-like zeolite. Starting materials included a ball-milled dry silica gel, ethyl orthosilicate, silicic acid and co-precipitated silica-alumina gels along with dried aluminum hydroxide, lithium hydroxide, sodium hydroxide and potassium hydroxide or calcined nitrates of aluminum, lithium, sodium and potassium.

The clinoptilolite-like phase was produced only in the high silica portion of the pure lithium systems. The source of silica did not affect the end product, i.e., the clinoptilolite-like phase was obtained with silicic acid and ethyl orthosilicate as well as with ball-milled silica gel. The clinoptilolite-like phase was not stable in systems containing potassium or sodium. Mordenite was the stable phase in mixed base cation systems.

The hydrothermal results support the conclusion that clinoptilolite is normally a metastable phase in most natural environments, and that the composition of clinoptilolite can vary from a silica to alumina ratio of 8 to at least 12. The substitution involved in the compositional change is  $Si^{+4}$  for  $NaAl^{+4}$ .

#### Condensate Treatment

The major equipment for demonstrating the decontamination of condensate streams was installed. The installation of the process and service pipes is in progress.

The decontamination ability of thin beds of ion-exchange materials continue to be of interest because of the possible high flow rates and low pressure drops. Steam-stripped Purex tank farm condensate was adjusted to pH 4 with nitric acid and passed through a bed of 100 to 250 mesh clinoptilolite at 5 gpm/ft<sup>2</sup> (10 cv/min). The bed was 5 cm in diameter and 2 cm thick.

The decontamination factors for cesium and strontium ranged from 100 at the beginning of the run to 400 at the end; a total of 4600 column volumes of waste were treated. Strontium-90 and Cs-137 concentrations in the effluent were less than their respective  $MFC_w$  values during the entire run. The pressure drop through the bed did not exceed 2 psi.

#### Ruthenium Removal from Low-Level Waste Streams

Scouting studies were initiated to evaluate possible methods for removal of ruthenium from actual plant condensate waste streams. Methods of removal which show promise are sulfide precipitation and adsorption on charcoal. Decontamination factors of 4 to 5 were obtained by sulfide precipitation on addition of 25 ppm  $S^{2-}$  and 50 ppm  $Cu^{++}$  at pH 2. The main difficulty with this procedure is precipitate aging and growth. Distribution coefficients of 100 to 300 were obtained with three types of activated charcoal for two waste streams.

#### Electrostatic Bubble Scrubber

The scrubber (1/6-ft<sup>2</sup> cross-sectional area) and associated equipment for testing electrostatic bubble scrubbing was received and assembled. Tests were made using uncharged fluorescent dye particles to evaluate the entrainment losses and to measure the inherent scrubber efficiency. These measurements will provide the background data necessary for the evaluation of the scrubber with charged particles. The liquid entrainment loss increased from 0.7 mg/min. at a gas flow

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1103129

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rate of 3 cfm/ft<sup>2</sup> to 3 mg/min. at 12 cfm/ft<sup>2</sup>. The scrubber efficiency for the uncharged dye particles (mean mass diameter  $\approx 0.2 \mu$ ) demonstrated the difficulties of efficient particle removal. With a gas flow rate of 6 cfm/ft<sup>2</sup>, the particle removal efficiencies decreased from 35 percent with 13 cm liquid depth to 6 percent with 0.7 cm liquid depth.

## Calcination of Radioactive Wastes

Analytical work was completed during the month on the spray calcination run with alkaline waste, a pot calcination run was made without additives, and construction and testing of a calorimeter was completed and the unit installed in the cell.

A spray calcination run with a slurry feed, prepared by adding excess caustic to Purex 1WW, was made last month and reported briefly in the February monthly report, HW-76596 C. Off-gas decontamination behavior is shown in the following table:

Off-Gas Behavior  
Spray Calcination Run with Alkaline Purex Waste

<u>Through:</u>	<u>Cumulative Decontamination Factors</u>			
	<u>Ce</u>	<u>Ru</u>	<u>Cs</u>	<u>Zr-Nb</u>
Calciner	$2 \times 10^3$	$2 \times 10^3$	$2 \times 10^3$	$2 \times 10^3$
Condenser	$3 \times 10^8$	$10^8$	$10^9$	$5 \times 10^8$
Electrostatic scrubber	$3 \times 10^8$	$10^8$	$10^9$	$5 \times 10^8$
Silica gel bed	$3 \times 10^8$	$5 \times 10^7$	$10^9$	$3 \times 10^8$
"Absolute" filters	$5 \times 10^8$	$2 \times 10^8$	$3 \times 10^9$	$3 \times 10^9$

It will be noted that the small fraction of the ruthenium escaping from the calciner is the same as that of the other fission products, suggesting entrainment rather than volatilization. Also, the condenser alone provides sufficient additional decontamination for discharge, and the additional units accomplish very little. The slight apparent decrease in decontamination across the silica gel bed probably represents desorption of ruthenium absorbed in a previous run. The traces of several fission products passing the "absolute" filters may be real or may represent cross contamination in handling the samples.

A pot run was made with sulfate-deficient, "as received," waste to determine the effect of residual nitrate on storage stability. In this run, the off-gas system was limited to the condenser,

1103130

electrostatic scrubber, and "absolute" filters, i.e., the caustic scrubber, demister, and silica gel bed were deleted. Although the run was apparently successful (no deposition of activity was seen on the cell exhaust filters), no analytical results are yet available.

A calorimeter, designed for remote reassembly in cell, was fabricated, tested, and installed. The "hottest" pot on hand was found to read 37 watts  $\pm$  6 percent, in reasonable agreement with a value of 44 watts calculated from radiochemical analysis of the feed.

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BIOLOGY AND MEDICINE - 06 PROGRAMTERRESTRIAL ECOLOGY - EARTH SCIENCESHydrology and Geology

Work on the determination of stream and path functions for three-dimensional flow through heterogeneous porous media was completed with the obtaining of equations having the desired properties for transient path functions. These derivations will permit the desirable analytical route to be followed in determining the in-place permeability distribution for electrical analog simulation of ground water flow and provide the basis of a method for later reconverting electrical measurements to meaningful hydrological results.

A 10 milligram Am-241 source was investigated as a means for measuring degree of laboratory soil column saturation for unsaturated flow studies. This long-lived radioisotope, which emits a 60 Kev photon, does not require the massive shielding that is required by other long-lived sources, e.g., Co-60, Cs-137. The following counting data were obtained, using a typical soil sample contained in a Lucite cylinder (2-inches I.D., 2.5-inches O.D.):

Three Minute Count

Dry soil	235,000
Wet soil	160,000
Background	400

The counting error was calculated to be about  $\pm 1.0$  percent for a three-minute count. Although some changes in the counting equipment and source arrangement are needed to permit traversing the length of a laboratory soil column, this method will be pursued for measuring degree of saturation.

Airborne magnetometer readings were plotted on profiles along existing geological cross sections. All known anticlinal ridges in the basalt south of Gable Mountain were confirmed. Locations differed at most only slightly from those previously indicated. North of Gable Mountain a complex of basalt structures was indicated that will require considerable further interpretation. A noticeable but minor increase eastward in the magnetic readings within about three miles of the Columbia River north of 300 Area coincides there with the appearance of the basalt flow overlying the Prosser bed.

RADIOLOGICAL AND HEALTH CHEMISTRYAnalytical Methods

A method for the determination of Pm-147 in urine was developed which is short, simple, and has a detection limit of less than 10 dpm for a 10 minute count. The method consists of wet ashing or muffling the sample, dissolving in dilute nitric acid, adjusting to a pH of 3.4 with sodium hydroxide solution, multiple extractions with scintillator solutions containing di-2-ethylhexyl phosphoric acid, and liquid scintillation counting.

Uranium Ore Inhalation Study

Measurements on a rat phantom indicate that 10 - 20 mg of the uranium ore used in these studies may be determined in vivo by gamma counting using the 184 kev gamma from U-235. It is estimated that these quantities of ore will deposit in the rat's lungs during the study and it will be important to the study to determine the actual amount within a few days after inhalation. The count can be made on the live rat placed in a large NaI(Tl) well crystal.

Environmental Studies

Cs-134 and Cs-137 concentrations were measured on an air filter which had been collecting air from early December, 1962, to March 1, 1963, at the 329 Building. The Cs-134 contributed 1.7 percent of the Cs-134 - Cs-137 total activity. This value agrees well with a previous measurement of 1.6 percent for an air filter sample taken during December, 1962, and January, 1963. This radionuclide concentration is larger than would be expected from the fission yields and probably arises as an activation product on material of the explosive device.

Radiation Chemistry

The presence of long-lived (half lives of the order of tens of seconds) organic free radicals formed by Co-60 irradiation of organic materials in aqueous solutions was demonstrated for the first time as far as can be determined. This finding adds new hope to the possibility of post-irradiation therapy and a new dimension to chemical protection studies. A  $3.6 \times 10^{-3}$  M p-nitroaniline solution was irradiated in oxygen-free alkaline solution and circulated through the electron spin resonance (ESR) cavity. The spectrum consists of three groups of 11 - 13 lines and is similar to that obtained when p-nitroaniline is

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reduced electrolytically. No signal is seen if oxygen is present; no signal is seen in acid or neutral solution; and addition of alcohol to its alkaline solution enhances the signal. These properties suggest that p-nitroaniline is reduced by hydrogen atoms derived from irradiated water and that the radical formed can be easily reoxidized by dissolved oxygen or hydroxyl radicals (unless the latter are scavenged by alcohol). The observed triplet splitting appears to be due to isotropic interaction between the unpaired electron and the nuclear moment of the nitrogen atom ( $I=1$ ) in the  $\text{NO}_2$  group, giving  $2I+1$  lines (further split by other interactions). The half-life of this radical is about 14 seconds at 25 C, increasing to 34 seconds at 5.5 C. Although this compound is of little biological significance, its radiation chemistry serves as a good starting place from which to attempt studies of more complicated biochemical radical species. From results such as described above and a study of the unpaired electron densities according to molecular orbital theory, we hope to evaluate the reactivities of biologically important radicals found during irradiation with biochemical substrates.

#### RADIOISOTOPES AS PARTICLES AND VOLATILES

##### Particle Deposition in Conduits

The statistical analyses of the deposition studies in the 58-foot long, 1-1/4-inch and 3-inch diameter tubes have been completed. The error in the reproducibility of the sampling method used to estimate entrance losses was found to be independent of the sampling rate and was an average of 1.7 percent for the 1-1/4-inch tube and 3.1 percent for the 3-inch tube. This loss was subtracted from the measured deposition.

Empirical equations for the deposition of ZnS particles (mean mass diameter  $\approx 3.7 \mu$ ) in the 1-1/4-inch diameter tube have been developed. The percent deposition equals

$$33.7 \ln (\text{cfm}) - 39.4$$

for flows from 5 to 25.6 cfm. Above 25.6 cfm the deposition remains approximately constant at 70 percent for flows up to 36 cfm.

This deposition is less than the predicted 89 percent and is thought to be due to re-entrainment. This conclusion is supported by the limited re-entrainment data available. Correlation of this data predicts re-entrainment to become significant at a flow rate of 18 cfm in the 1-1/4-inch diameter tube. This is approximately the flow rate at which the difference between the predicted and

experimental deposition becomes appreciable. Similarly, re-entrainment is predicted to occur about 2.5 cfm in the 1/2-inch tube and to be insignificant in the 3-inch tube at the flow rates studied. These results are qualitatively supported by the deposition data. It seems, then, that the velocity for serious re-entrainment can be estimated by available correlations.

Deposition in the 3-inch vertical tube was found to be constant at 6 percent for flow rates from 4 to 36 cfm. This deposition is much higher than that predicted and a new deposition model is being investigated.

### ISOTOPES DEVELOPMENT - O8 PROGRAM

#### Fission Product Encapsulation

Successful formation of strontium titanate by the Dynapak (high energy compaction) process was reported last month. Use of the technique was extended this month to include several other fission products and compound forms and to the formation of 4-inch cylinders of strontium titanate (versus 2-1/2 inches, largest previously produced). A new (Mark II) can design with rounded corners was successful in eliminating weldment cracks and wrinkled walls. Highlights of container design studies are as follows:

1. Container wall deformation mode varies with wall thickness to container height ratio: thin wall (0.025-inch) containers deform by buckling, whereas thick wall (0.125-inch) containers thicken, apparently by deforming in shear.
2. Failure of the joint between the cap and wall during compaction can be eliminated by providing a fillet transition and by locating the closure weld in the can wall rather than at the intersection between the cap and the can wall.

Monolithic strontium oxide compacts (of interest to the space program) of 96 percent theoretical density were produced by loading strontium carbonate into a can, outgassing at 1300 C, and compacting at 237,000 psi. Neodymium oxide, a stand-in for promethium oxide ( $Pm_2O_3$ ) was compacted to a crystalline, non-porous product with a density of 98.5 percent of theoretical, and ceric oxide ( $CeO_2$ ) to a density of 84 percent. The  $CeO_2$  product was not crystalline but rather a compacted powder. Two 4-inch right circular cylinders of strontium titanate, Inconel-X clad, were readily formed. Each contained 2100 grams of  $SrTiO_3$ , equivalent to over 2000 thermal watts, and had excellent densities of about 96 percent. These 4-inch sources are as large as would be required

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for any presently conceived thermoelectric generator; however, four inches is by no means the limit of the Dynapak technique.

Early tests will be aimed at extending the Dynapak process to cesium compacts (cesium poly glass and salts such as cesium chloride) and at development of a filter can concept to eliminate powder handling.

Shielding curves have been prepared relating dose rate from separated, purified individual fission products to thickness of shielding (for kilocurie point sources). These curves will be particularly beneficial in calculating the attenuation of Bremstrahlung.

*W. H. Reas*

Manager  
Chemical Laboratory

WH Reas:cf

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## BIOLOGY LABORATORY

## A. ORGANIZATION AND PERSONNEL

Dr. C. E. Breckinridge joined the Pharmacology Operation as a Biological Scientist on March 8, 1963.

## B. TECHNICAL ACTIVITIES

## FISSIONABLE MATERIALS - O2 PROGRAM

Effect of Reactor Effluent on Aquatic Organisms

Eight groups, each with 500 young chinook salmon, in their tenth week of chronic exposure to 3 and 6% effluent containing either dichromate or Quachrom Glucosate as the corrosion inhibitor show little difference in mortality. About 3% mortality is observed in all groups.

Growth depression caused by higher effluent concentration is strongly inferred by the statistically significant ( $P < 0.01$ ) smaller body weight of fish in 6% effluent. The over-all mean by treatment groups is summarized below. While Quachrom Glucosate introduces less chromium, no apparent difference in growth is observed between dichromate and Quachrom Glucosate effluent.

<u>Corrosion Inhibitor</u>	<u>Effluent Concentration</u>	
	<u>3%</u>	<u>6%</u>
Dichromate	0.641 g	0.615 g
Quachrom Glucosate	0.644 g	0.617 g

Columnaris

Bacto tryptone is known to serve as a basic nutrient for columnaris. In an attempt to further identify the specific nutrients required for growth, the tryptone was fractionated into acetone insoluble and soluble fractions at pH's of 4, 7, and 10. No growth was obtained from any of the insoluble fractions. Best growth was obtained from the pH 4 and 7 soluble fractions.

No evidence of a change in virulence was noted in isolates from trout held throughout the summer in troughs in spite of the fact that mortality had been largely limited to one two-week period early in the summer. This observation is consistent with the possibility that the fish developed a resistance to the columnaris organism.

Chinook fingerlings of the same chronological age but varying in size (one group 0.6 g, the other 0.9 g) were equally susceptible to acute death from columnaris. Apparently age, not size, is the determining factor.

## BIOLOGY AND MEDICINE - 06 PROGRAM

## METABOLISM, TOXICITY, AND TRANSFER OF RADIOACTIVE MATERIALS

Zinc

The importance of the gills of fish in the excretion of orally administered  $Zn^{65}$  was indicated by a material balance study using three-year-old trout with the anus ligated and the bladder cannulated. Under the assumption that the unaccounted fraction of dose is the amount excreted by the gills into the water, about 0.7 of dose is excreted by the gill tissue during seven days post-administration.

Early results indicated that younger fish have relatively greater retention of  $Zn^{65}$  a week after a single oral dose. Body burden on the average was 0.36 and 0.27 of dose for two-year-old trout and three-year-old trout, respectively. Exclusive of the gastrointestinal tract no apparent difference due to age was found in the distribution in the various tissues. The highest concentration was found in gill filaments in both age groups. Fractions retained in the gastrointestinal tract were 0.11 and 0.06 for two- and three-year-old trout, respectively.

Strontium

The skeletal retention of  $Ca^{45}$  and  $Sr^{90}$  ten days following a single oral dose was determined for miniature swine from 15 to 240 days of age. Suckling animals were administered the radionuclides in a milk solution via gastric intubation, older animals were given the  $Sr^{90}$  and  $Ca^{45}$  with an aliquot of feed. The highest retention of both radionuclides was ~80% and was observed in the youngest animals. With increasing age, the percentage retention decreased with the most marked change occurring in animals dosed between 15 and 45 days of age. The retention of  $Sr^{90}$  decreased more markedly with age than did the retention of  $Ca^{45}$ . As a result, the discrimination against  $Sr^{90}$  relative to  $Ca^{45}$  for transfer from diet to skeleton changed from near unity for the youngest animals to ~25% for the older animals.

One female miniature pig which had ingested high levels of  $Sr^{90}$  early in life and had survived, due to removal from  $Sr^{90}$  feeding, gave birth to apparently normal offspring. (This animal was born to and had suckled a dam ingesting 625  $\mu$ c  $Sr^{90}$ /day and had ingested 155  $\mu$ c  $Sr^{90}$ /day for 6 weeks. After 6 weeks, the radiation dose to this animal's bone was on the order of 100 rads/day.)

Iodine

The stable iodine content of the ration of two of the three cows fed 5  $\mu$ c of  $I^{131}$  per day was raised from 5 to 15 mg of  $I^{127}$  per day. After about two weeks on this increased stable iodine diet, the  $I^{131}$  concentration in the thyroid decreased about 40%, while the concentration of  $I^{131}$  in the milk showed a slight increase (this milk is being periodically drunk by volunteers).

Peak thyroid uptakes of  $I^{131}$  were 2-6%, five to eight days following the topical application of  $I^{125}$  and  $I^{131}$  over 50 and 100  $cm^2$  of skin area of adult male sheep. The uptake appeared to be independent of the area or application site.

Protein-bound iodine values manifest an increase at mid-gestation in ewes that are now showing various stages of hypothyroidism following administration of 3 mc of radioiodine 4 to 5 years ago.

Three female miniature goats fed 0.15  $\mu c$  of  $I^{131}$  per day are maintaining thyroid burdens of 3 to 4 times the daily dose, while the 5 nursing offspring have a thyroid burden of 1/4 to 1/2 that of the adults. The milk concentration in these goats reflects the blood concentrations and shows the highest concentration at about 4 hours after daily feeding and lowest values at about 24 hours post-feeding. A more than ten-fold change in milk concentration occurs in a 24-hour period. (The blood concentration in the offspring approximates that observed in the dams.)

#### Cerium and Promethium

Four animals were administered single oral doses of either  $Ce^{144}$  or  $Pm^{147}$  and killed ten days later to determine gastrointestinal absorption and tissue distribution. Samples of excreta were collected prior to the killing of the animals. (Radioanalysis is incomplete at this time.) Both radionuclides had been obtained as chlorides from Oak Ridge. Additional animals will be given  $Pm^{147}$  of Hanford origin.

#### Plutonium

Additional data on the oral effectiveness of TTHA in removing deposited plutonium confirm the preliminary results reported last month. TTHA is only very slightly more effective than DTPA. In chronic treatment studies, DTPA and TTHA were administered orally, daily, for nine days, at a treatment level of 1.5 mM/kg per day. Neither agent was significantly effective under these conditions.

#### Inhalation Studies

One dog died about 1200 days after a single exposure to  $Pu^{239}O_2$ , which resulted in deposition of an estimated 6  $\mu c$  of  $Pu^{239}$ . Prior to death, electro-cardiographs and thoracic radiographs showed cardiac failure and right ventricular heart enlargement causing dorsal deviation of the trachea. Blood  $CO_2$  levels were high and  $O_2$  levels were low. Anorexia, dehydration cyanosis, and 20 per cent weight loss occurred before death. At post mortem, lung lesions were extensive and the bronchial lymph nodes were small, fibrotic, and contained enough  $Pu^{239}$  to be detected with a "poppy". The right ventricle of the heart was enlarged, and a growth on the left a.v. valve of the heart nearly filled the left ventricle. It has not been determined whether the heart lesion was a consequence of the plutonium inhalation.

Thirty-six rats were exposed to an aerosol of pitchblende uranium ore, and are being monitored in a well-type scintillation counter detecting the 180 KEV gamma of U<sup>235</sup>. Preparations are being made for studying the excretion of tissue distribution of inhaled Pm<sup>147</sup> and Ru<sup>106</sup> in dogs.

Further tests were initiated to determine methods for removal of inhaled Pu<sup>239</sup>O<sub>2</sub> from rats. Dogs were exposed to I<sup>131</sup> vapor with and without I<sup>127</sup> to determine the effects of I<sup>127</sup> on radiiodine uptake in the thyroid.

#### Gastrointestinal Radiation Injury

The effectiveness of abdominal X-irradiation (1000 r) on bile salt absorption from the intestinal tract was studied in rats with biliary fistulae, injected intraduodenally with C<sup>14</sup>-labeled taurocholate and C<sup>14</sup>-labeled cholic acid. Appearance of the C<sup>14</sup> label in the excreted bile showed that irradiation did not decrease absorption of the bile salts from the intestine during the period from one to six days following irradiation.

Efforts to simulate the effects of bile salts in producing diarrhea in irradiated rats, by administering the surfactants Triton X100 and Duponol have thus far met with no success.

An ion exchange resin, cholestyramine, which is supposed to have a special affinity for bile salts, is being tested for its ability to prevent the diarrhea which results from abdominal irradiation. Results from these experiments are not yet available.

#### Secondary Disease Studies

Attempts to produce healthy chimeras by neonatal injection of homologous cells have been partially successful. A/He (Darwin) mice, neonatal treated with LAF mouse cells, have been successfully grafted (5/7) with skin from LAF mice. These animals will be used as donors of cells to be injected into irradiated LAF mice. Rats neonatally injected with mouse cells have not accepted mouse skin grafts. However, three out of eight rats which rejected the mouse skin grafts do not show anti-graft hemagglutinins, indicating, perhaps, some degree of tolerance.

Attempts are being made to fractionate chromatographically the antibodies found in rat-mouse chimeras. The rat anti-LAF hemagglutinin appears to be associated with the gamma globulin fraction.

#### Hepatic Tumor Study

An experiment was initiated to determine the effect of internal emitters on induction of hepatic tumors by the azo-dye carcinogen, n,N-dimethyl-p-phenylazoaniline (DMAB). The isotopes to be studied are Pu<sup>238</sup> and Ce<sup>144</sup>-Pr<sup>144</sup>. A single isotope injection was given intravenously. The DMAB is included in the diet at a level of 0.08 per cent. The time, degree, and type of tumor incidence will be determined.

Microbiology

In four yeast strains, each having characteristic carbohydrate utilizing enzymatic systems, D<sub>2</sub>O had little effect on growth, as measured by cell mass, but had pronounced effect on cell division. The effect on cell division with yeast is in accord with reported effects of D<sub>2</sub>O on cell division in animal cells.

More HTO was incorporated into yeast cells when the cells were suspended in normal medium than when they were suspended in distilled water. Likewise little label was lost from cells previously cultured in HTO when these cells were shaken in distilled water. From these limited data, it appears that water movement in yeast may be related to presence of either ions or organic materials.

Radiation Effects on Insects

Two more attempts to isolate proteins by electrophoresis in larval Ephestia haemolymph were unsuccessful. Techniques are being refined, and a study to determine changes in total protein between control and X-rayed Ephestia larval is planned.

The influence of freezing temperatures (5 C) on radiation damage (failure of adult development when larvae are X-rayed) in Ephestia is being investigated. Data, so far, indicate that controls kept as long as two weeks at 5 C will continued development when returned to 28 C.

In the progeny from X-rayed parental Tribolium, antennal characteristics appear most easily altered by radiation. Failure of antennal segmentation and branched antennal are found.

Plant Studies

The uptake of water and of I<sup>131</sup> by barley seedlings was measured at two-hour intervals over a 24-hour period. Extreme care was taken to avoid upsetting the diurnal rhythm by exposing plants to light during the normal eight-hour dark period. The accumulation of I<sup>131</sup> was three times greater during the night periods than during the light periods. Uptake of water was appreciable during light periods but almost nil during dark periods. Consequently the uptake of I<sup>131</sup> appears to have been nearly completely dissociated from the uptake of water. Thus it appears that the iodide ion cannot be absorbed as a consequence of water flow but instead must be taken into the plant by a specific metabolically driven process.

Leaf-soil ratios for the uptake of W<sup>185</sup> of the following magnitude were observed:

Ephrata soil	0.0017
Cinebar soil	0.0004
Milville soil	0.0033

These values were obtained with barley plants and comparable values for bean plants were approximately an order of magnitude less. The uptake of tungsten appears to differ from most other ions studied in that greater uptake was observed from alkaline than from acidic soils.

No internally deposited  $I^{131}$  was removed from leaves when they were exposed to high vacuum. Leaves had been removed from the plants at the time of exposure to vacuum.

#### Plant Ecology

Soil moisture studies in the field from February 20 to March 19, 1963, showed that sagebrush soil lost moisture more rapidly than greasewood soil. Moisture loss amounted to 9 g/dm<sup>2</sup> over a 27-day period of no measurable precipitation, in the greasewood soil as compared to a loss of 13 g/dm<sup>2</sup> in the sagebrush soil. Soil moisture loss early in the moisture depletion season is characteristic of the sagebrush habitat and appears to be an important feature restricting the distribution of greasewood.

#### Rattlesnake Springs Limnology

A periphyton sampler was placed in the creek to determine the composition of this community and to find out if it would be practical to use this group of organisms in uptake studies. Four slides were immersed and one examined each week. Gross examination revealed the presence of three diatom species, one of which comprised better than 99% of the population. One green algae species was present. Chironomid and simuliid larvae appeared on the slides after two weeks exposure.

#### Columbia River Limnology

Analysis of plankton samples from the 100-F Area was continued. A series of periphyton substrates were placed in the channel which receives thermally hot seepage water from the 100-F retention basin. Water temperatures at the three stations selected, upper end, mid-way, and lower end, were 45.1, 33.2, and 29.2 C, respectively. The temperature of the seepage water as it emerges from the ground is about 54.5 C. This area is well-suited to study the effects of temperature upon the distribution, biomass, and composition of aquatic communities.

#### Fallout

Radioiodine concentrations in North American deer thyroids decreased at approximately 15 days half-time and approached the detection limit at the month's end. Values at the various sampling locations were as follow:

<u>Location and Sample Type</u>	<u>Number of Animals</u>	<u>Picocuries I<sup>131</sup> per gram Wet Thyroid</u>		
		<u>Mean</u>	<u>+</u>	<u>One Std. Error</u>
Montana elk	15	650		70
Maryland deer	9	110		30
Alaska reindeer	5	72		6
New York deer	4	69		8
California deer	1	30		-

The longer half-time reported last month resulted from a plateau in thyroidal I<sup>131</sup> concentrations caused by a series of seven Russian atmospheric nuclear tests conducted during late December 1962. No effect was observed in thyroids as a result of underground U.S. tests conducted during February.

  
Manager  
BIOLOGY LABORATORY

HA Kornberg:es

TECHNICAL INTERCHANGE DATA  
BIOLOGY LABORATORY

I. Speeches Presented

a. Papers Presented at Society Meetings and Symposiums

None

b. Seminars (Off-Site and Local)

McClellan, R. O. Biological effects of Sr<sup>90</sup> and other bone-seeking radionuclides. Department of Zoology, Washington State University, Pullman, Washington (Exchange Seminar Program). March 4, 1963.

c. Seminars (Biology)

Brown, D. and E. Sheen, Chemical Laboratory, Hanford Laboratories. Recent trends in transistor electronics and some biological applications. March 5, 1963.

Fujihara, M. P. Observations of irradiation effects on cichlids. March 12, 1963.

Horstman, V. G. (presented by L. K. Bustad). Basis for extrapolating miniature swine data to man. March 12, 1963

Stephens, C. M., Department of Chemistry, Washington State University, Pullman, Washington. Biosynthesis of leucine in microorganisms. March 15, 1963.

Tombropoulos, E. G. Fatty acid synthesis. March 19, 1963.

Uyekl, E. M. Some immunological reasons for bone marrow rejection in radiation chimeras. March 19, 1963.

Mahlum, D. D. Np<sup>237</sup> and fatty liver production. March 26, 1963.

Park, J. F. Chronic toxicity of inhaled plutonium in dogs. March 26, 1963.

d. Miscellaneous

Stuart, B. O. Theories for mechanism of action of radiation. Radiation Biology Course, University of Washington Branch, Richland, Washington. March 20, 1963.

Hungate, F. P. Genetic effects. Radiation Biology Course, University of Washington Branch, Richland, Washington. March 27, 1963.

Hanson, W. C. Project Chariot. Kiwanis Club, Benton City, Wash. March 8, 1963.

1103144

UNCLASSIFIED

## II. Articles Published

### a. HW Documents

- Uyeki, E. M. Intracellular distribution of cations and protein in regenerating rat liver. Am. J. Physiol. 204: 257-261, 1963.
- McClellan, R. O., M. E. Kerr, and L. K. Bustad. 1963. Reproductive performance of female miniature swine ingesting strontium-90 daily. Nature 197: 670.

## III. Visits and Visitors

### a. Visits to Hanford

- 3-8-63 Dr. David C. England and Roy E. Fancher. Department of Animal Science, Oregon State University, Corvallis, Oregon. Discuss research with L. K. Bustad.
- 3-8-63 Bishop Hubbard, Episcopal Church, Richland, Wash. Tour of Biology facilities with L. A. Temple.
- 3-11 to 15 Michael Tierman, Health and Safety Division, AEC, Idaho Falls, Idaho. Review work in progress in Biology as part of a training program. Contacted Biology staff members.
- 3-11-63 Drs. Ralph Baltzo and Kenneth Jackson, University of Washington, Seattle, Washington. Tour Biology and discuss research with H. A. Kornberg.
- 3-14-63 Dr. Lauriston Taylor, Head, National Committee on Radiation Protection. Toured with H. M. Parker and H. A. Kornberg.
- 3-15-63 Dr. C. M. Stephens and M. G. Kalyanbur, Washington State University, Pullman, Washington. Dr. Stephens presented a seminar. They conferred on radiation problems with Biology staff members.
- 3-15-63 Dr. Glenn T. Seaborg, Senator H. M. Jackson, A. R. Luedecke, E. J. Bloch, Arnold Fritsch, J. T. Ramey, J. T. Conway, and L. R. Fink (GE Vice President), members of AEC, Washington and Joint Committee on Atomic Energy. Toured Biology facilities with H. M. Parker and H. A. Kornberg.
- 3-28-63 Washington State Power Planning Sub-Committee (19), from various cities in Washington. Toured Biology with R. F. Palmer.

## III. Visits and Visitors (Continued)

## b. Visits Off-Site

- 3/1 and 3/5 V. G. Horstman inspected feed at the Anderson Feed and Produce Company, Othello, Washington.
- 3/1-3 L. K. Bustad worked on his ionizing radiation study at the University of Washington, Seattle, with Dr. Ruch.
- 3/2-12 R. F. Keough attended the Conference on Analytical Chemistry and Applied Spectroscopy at Pittsburg, Pa., and discussed research techniques with Coleman, Blake, and Brown at Oak Ridge National Laboratory.
- 3/3 R. O. McClellan presented a seminar (Exchange Seminar Program) at Washington State University, Pullman, Washington.
- 3/8-9 E. G. Tombropoulos and W. J. Bair attended the AEC's Inhalation Toxicity Meeting at Wayne State University, Detroit, Mich. and discussed research with Vorwald and Willard.
- 3/9-16 W. J. Clarke conferred on establishment of a Pathology Registry with Hillberg at the Armed Forces Institute of Pathology in Washington, D. C.
- 3/17-20 H. A. Kornberg attended a sub-committee meeting of the BEAR Committee of the National Academy of Sciences to discuss a report on the treatment of radiation syndrome. Meeting was conducted by Shields Warren. He also toured the Radiological Laboratories of Columbia University in New York with Dr. H. Rossi.
- 3/13 R. O. McClellan inspected radiographic equipment at the Sacred Heart Hospital in Spokane.
- 3/20-22 D. G. Watson and R. E. Nakatani attended the Pacific Fisheries Biologists Meeting in Gearhart, Oregon.
- 3/19-22 R. O. McClellan attended the International Symposium on Bone Biodynamics in Detroit.
- 3/23-26 L. K. Bustad attended an informal meeting on I<sup>131</sup> at Argonne Cancer Hospital, Chicago. Contacted Dr. Bruner, AEC, Washington, DC and others attending.
- 3/29 R. O. McClellan visited Memorial Hospital in Yakima to discuss radiographic techniques with Dr. James.

IV. Achievements

No degrees were earned, nor did any professional licensing or certification occur.

V. Honors and Recognitions

None

VI. Professional Group or Organization Assignments

None

which apply to it, by allowing treatment of reactor component repairs while avoiding the sticky (and realistically speaking, impossible) problem of writing and solving something like 2,500 to 22,000 simultaneous differential equations.

Two programming problems must be solved before the Monte Carlo approach can be used. First, a computer method is needed for writing Boolean expressions given the pieces of equipment concerned and the degree of repair required of the equipment. Second, a computer problem must be written to determine, in the most efficient manner, the truth or falsity of a Boolean statement, given the truth or falsity of the subevents upon which the statement depends. A good start toward the solution of the second problem has been made; a few small networks have been treated by the computer.

Work continued on parameter estimation in the crack problem for NPR primary piping. A program was used to plot parameter spaces for a new function which confirmed least square minima in some cases and gave new points to use for initial points in a least square minimization program.

#### Irradiation Processing Department

A topical study was made of the measurement error structure for the non-destructive testers used in the evaluation of fuel quality. Primary emphasis was given an evaluation of the economic consequences of the existing measurement errors. It was concluded that immediate steps should be taken to eliminate biases, but that at present, there seems to be little incentive for improved precision. Concrete proposals for reduction of bias were presented. Results of this study were presented in two meetings and a topical report on this subject is currently being prepared.

In pursuance of the solution to the rail height specification problem for self-support K-reactor fuels, an expression was found relating minimum annulus between the fuel element and the process tube wall as a function of tube and fuel element radii, rail height, and angle subtended at the fuel center by the rails. This expression, which takes into account the fact that both rails supporting the underside of the fuel are more than likely not equal in height, is being used in a computer simulation which gives the distributions of minimum annulus as functions of input distribution for the independent characteristics. Results are useful in determining the in-reactor distribution of minimum annuli, which, in turn, are required when performing heat calculations. For this application, input distributions are truncated by inspection. As another application, it is possible to anticipate what would be the reject rate if the specification is changed from one on rail heights to one on minimum annulus with a revised type of gaging. Finally, an additional simulation is being performed to give the distribution of minimum circumscribed circle as a function of input distribution, including fuel ellipticity.

Also in connection with this self-support problem, additional calculations and analysis are being performed on "R" data from the ten-tube test using.

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APPLIED MATHEMATICS OPERATION

MONTHLY REPORT - MARCH, 1963

ORGANIZATION AND PERSONNEL

J. L. Jaech transferred to Vallecitos Atomic Laboratory as Consulting Statistician, effective April 5.

P. Riggle, a technical graduate, rotated out of the group on April 1 to take an assignment in the N-Reactor Department.

OPERATIONS RESEARCH ACTIVITIES

The current status of the Reactor Simulation Study for IPD was reviewed. A decision was reached to push the study along despite data difficulties. Analysis of reactor activity data has now been brought up to February, 1963. Missing data on maintenance staffing patterns is to be estimated by field interviews.

A sampling plan was developed for a proposed semimonthly survey of HAPO employees. The subsequent statistical computations will be performed by the computer as soon as programs can be developed.

STATISTICAL AND MATHEMATICAL ACTIVITIES FOR OTHER HAPO COMPONENTS

N-Reactor Department

Some help was given as to the type of statistical statements that can be made about the true percentage of good fuel elements from the N-Reactor fuel element manufacturing process, based on the limited number of fuel elements which have been tested to date.

Two meetings of N-Reactor personnel were attended to provide guidance regarding logical representation of reactor systems and operational ground rules.

The approach to the N-Reactor reliability study has been broadened to include an application of the Monte Carlo method for determining percentage of time spent by the reactor in any of the modes of operation being considered. This provides a very faithful model of the reactor system and the operating rules

1103148

DECLASSIFIED

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HW 7346

different rail heights. "R" data give a measure of temperature imbalance in a process tube. The computer routine developed for his application was also referred to IPD personnel monitoring the aluminum process tube corrosion problem, who routinely analyze large amounts of "R" data. A preliminary run was made on submitted data in connection with this application.

A statistical analysis was completed of postirradiation dimensional distortion data from IP-490 for fuel elements canned by the ALSi, the Hanford hot press, and the Sylcor hot press processes. A balanced incomplete block design was used in this test to insure that not all information would be lost in the event poor performance by one treatment necessitated early discharge.

A rough analysis was made of rupture data for the past two years to estimate what proportion of ruptures are "atypical". This estimate was found by examining the distribution of "multiple-failure" charging dates, confounded with "multiple-failure" lots, and was checked by observing the proportion of ruptures occurring at exposures less than 50 percent of goal. Under the existing rupture model, about three percent of the "normal" ruptures will occur at these low exposures.

Data from a test based on a second-order composite rotatable design to determine the best conditions for end bonding fuel elements canned under the experimental hot die sizing process are being analyzed as the different yield data are made available. To date the internal, external, cap and base bond counts as well as stud-pull values from five locations on each fuel element have been analyzed. Very useful and consistent results have indicated how the process variables affect these stud-pull values indicating possible regions of optimization for these variables. Dimensional measurements are presently being analyzed. Heretofore all the fuel elements have been canned with dingot cores, but this test makes a comparison of dingot with ingot material.

Data from several pilot plant tests were analyzed as requested. The conclusions were based on UT-4 and UE-1 data. Two of the tests compared "old" versus "new" lots and the normal F process versus a process using an ultrasonic deoxidizing cleaning procedure.

An extensive amount of dimensional measuring was performed on nine process tubes by two individuals using two types of measuring devices, each individual making repeat measurements over an interval of several days. The resulting data, which will give valuable information not only on measurement error, but also on process variation, are being analyzed using the MERCY routine.

In connection with the proposed audit program for C-Basin, some simulated data were submitted to the MERCY routine, and the resulting output were

1103149

further analyzed using methods of incomplete block analysis. This simulation was the basis of a report directed to C-Basin personnel to demonstrate the kinds of information which will result from a program of the type proposed.

An error analysis of a device for measuring tube displacement was started. The initial short term investigation did not produce any conclusions.

#### Chemical Processing Department

Recommendations are being prepared relative to possible revisions in the system for control of MJF's at Z-Plant.

A review is being made as requested of the current practice of using monthly bias corrections for analytical results based on monthly analyses of standards.

Continued analyses are being made on dimensional data for parts. Currently the underlying process distributions, free from the effects of measurement error, are under investigation.

In the final inspection of parts, yields at a given station are obscured in meaning, since once a part is rejected at a given station, it is not measured at ensuing stations. Consideration is being given ways of providing meaningful yield figures for each station.

Work continued on the refinement and programming of a mathematical model of spare parts and general inventory control.

#### STATISTICAL AND MATHEMATICAL ACTIVITIES WITHIN HL

##### 2000 Program

##### Pulse Column Facility

Data from a block of 16 pulse column runs, a portion of the factorial experiment to study the extraction characteristics of the column as a function of six independent variables, extracted stream temperature, extracted stream acid concentration, feed stream concentration, feed stream and extractant stream flow rates, and pulsing frequency, are being used to investigate the organic volume fraction profile in the column. A mathematical model expressing the organic volume fraction as a function of column position and these independent variables is needed to firm up the system of nonlinear differential equations expressing the mass transfer dynamics in the column. Previous attempts to fit the mass transfer model to experimental organic and aqueous concentration data have been hampered because such a mathematical form was not available. Preliminary analysis of the block of 16 runs indicates that the profile of the organic volume fraction is primarily a function of the distance from the flooding

1103150

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DECLASSIFIED

5-E

HW 1103

curve in the capacity-power space. Some second order dependence on other independent variables is also indicated.

Sets of data were submitted presenting isotopic activity as functions of distance along a process tube. These were analyzed using the GEORGE routine in which the logarithm of activity was expressed as a quadratic function of distance along the tube for each isotope. An addition was made to the GEORGE routine to calculate predicted accumulated activity at discrete small intervals along the tube.

Consulting assistance was provided in expressing minimum fuel cost as a function of price values assigned to the isotopes of plutonium, to americium<sup>243</sup>, and to curium<sup>244</sup> for each of several reactor cases and associated ground rules of operation. For the cases considered to date, very simple expressions were found.

#### 3000 Program

An analysis of a series of tests of shear-spinning preformed metal blanks into selected shapes indicated the desirability of possessing a means of designing such blanks which would be rapid, yet flexible to alternative selections of design parameters. An EDPM program, based on a modified uniform shear theory, and which exhibits the desired flexibility, has been written and placed in service.

The EDPM program which generates the magnetic tape input to the experimental  $\delta$ - $\omega$  lathe, has been slightly modified in an attempt to prevent non-uniformly spaced bits from jamming the pulse motors. A complete 1251 exterior contour tape has been generated using the modified techniques and is presently under study on the lathe.

#### 4000 Program

The EDPM program for obtaining numerical solutions to a simultaneous set of first order ordinary differential equations has been completed. A function comparison subroutine was written and added to the basic program. This program is now being used to study the NPR stack gas chemical reactions which are vital to the Zirconium-Graphite compatibility problem.

Progress has been made on the study of the geometry of spheres in cylindrical containers. In particular, a quantitative description of the observed phenomenon of spiraling has been developed for a limited range of sphere-cylinder diameter ratios.

An experimental model of a vibrationally compactified annular shaped fuel element was fabricated which achieved a density of 93.3 percent of theoretical. This sets a new density record.

1103151

1103151

### Swelling Studies

The modifications to the quantitative metallography FORTRAN program for estimating volume fraction and density of a spherical phase in a three-dimensional matrix from Zeiss particle size distributions were completed. Several debug runs indicate that the program is functioning properly.

### 5000 Program

#### Actinide Element Research

The second phase of the FORTRAN program for indexing hexagonal-tetragonal crystals was nearly completed in March. Test data with random error components were successfully run on the program. Acceptable criteria for the validity of solutions have been formulated.

A major addition to the program made during March was an alternative wavelength selector. All previous programs have used copper X rays with no provision for an alternative target such as iron or molybdenum. Rough draft work has started to properly document these programs.

During the month of March a new and unknown crystal was indexed from a powder pattern on an orthorhombic basis using the program being developed for this purpose. Subsequently, a single crystal of the compound was analyzed independently, which verified that the computer solution was correct. It is believed that this is the first time a comprehensive computer program has ever achieved the feat of indexing a complicated crystal, such as an orthorhombic one, exclusively from powder pattern data, and subsequently have the indexing proved out as correct.

#### Computation and Statistical Analysis

Data evaluation continued during March in preparation for an April staff meeting in Washington

#### Radiochemical Analysis

Many improvements were made and many errors corrected in the new IRA II system during the month of March. These improvements have significantly reduced the running time on several passes. Several successful processings of large amounts of program data were completed. Work continues on the specification of improvements to the system.

### 6000 Program

#### Biology and Medicine

An analysis of variance was started on a biology problem concerning irradiated tribolium. No results have been completed to date.

1103152

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Assistance was provided in connection with the estimation of crystal structure lattice constants. A procedure was outlined for performing a least squares fit of X-ray diffraction data to estimate precision lattice constants at 90 degrees using Nelson-Riley extrapolation.

Work resumed on a tabling program for radiation protection of density conversion factors.

*Carl A. Bennett*

Manager  
Applied Mathematics

CA Bennett:dgl

1103153

REACTOR DEVELOPMENT - O4 PROGRAMPLUTONIUM RECYCLE PROGRAMThe Economic Impact of Valuing Cm-244 and Other Special Isotopes

A part of the Cm-244 production study has been to assess the economic effect of setting various dollar values for Cm-244, Am-243, Cm-242, Pu-238, and other isotopes which are important in the production of isotope heat sources. This analysis has been carried out in a simulated pressurized water reactor operated at a specific power of 20 MW/T. Eight fueling schemes are considered, each with a variety of assumed isotope prices.

Table I contains results from two choices of Am-243 and Cm-244 prices as a demonstration of their effect on fuel cost. The plutonium prices are held constant at \$10 per gram fissile and the fuel element fabrication charge is \$40 per pound of fuel.

Examination of Table I shows the uranium enriched fuel cycle cost would be reduced about 0.1 mill. Whereas the plutonium in natural schemes, which produce four or five times as much Am-243 and Cm-244, would enjoy 0.3 to 0.5 mill reductions, making the plutonium schemes that are already attractive at the \$10 per gram price even cheaper.

The other fueling schemes considered (4, 5, 6, 7, and 8) are designed to increase the purity of Cm-244 in the Cm-242-Cm-244 batch formed during irradiation. The Cm-242 is formed when Am-241, which is a decay product of Pu-241, captures a neutron. Cm-244 is formed by successive neutron captures in Pu-242 and then Am-243. Thus the Cm-244 purity is enhanced by using plutonium with a high percentage of Pu-242 and by reducing the formation of new Pu-239, Pu-240, and Pu-241 atoms from U-238. A high Cm-244 purity is needed because of the high heat producing rate of Cm-242. (About 50 times greater than Cm-244.)

Schemes 4 through 8 obviously are not economically competitive without a high credit for Am-243 and Cm-244. With the credit used in this analysis, scheme number 4, with a high initial Pu-242 percentage, became very competitive. Schemes 5 and 6 receive proportionately smaller reductions in fuel cost due mostly to the smaller Pu-242 percentages. The last two schemes are much less dependent on Cm-244 and Am-243 price because of the low plutonium concentration (0.03 gm/cc). Although these schemes produce curium of much higher quality the latter schemes will be investigated with higher plutonium concentration in the fuel.

A program is being written to establish yields of pure Cm-244 as a function of decay time after reactor irradiation. The Physics codes are being arranged to allow simultaneous irradiation of plutonium batches of different composition. This should permit achieving Cm-244 purities approaching the purity achieved by schemes 7 and 8 of Table I but with acceptable fuel cost and with acceptable Cm-244 quantities.

TABLE I  
 MINIMIZED FUEL COST IN MILLS FOR A PRESSURIZED WATER REACTOR  
 (With and without curium credit.)

No.	Fueling Scheme	Am-243 = \$0/gm & Cm-244 = \$0/gm		Am-243 = \$350/gm & Cm-244 = \$700/gm	
		Batch	Graded	Batch	Graded
1	Uranium Enriched	1.92	1.56	1.82	1.34
2	Plutonium (93,7,0,0) in Natural Uranium	1.73	1.42	1.28	0.86
3	Plutonium (76,18,5,1) in Natural Uranium	1.67	1.41	0.87	0.60
4	Plutonium (76,18,5,1)	2.22	1.96	-0.66	-1.27
5	Plutonium (22,46,22,10)	2.27	2.29	1.26	1.18
6	Plutonium (93,7,0,0)	2.39	2.43	1.84	1.68
7*	0.03 gm/cc Plutonium (76,18,5,1) with U-235 Enrichment	3.91	3.06	2.78	2.89
8*	0.03 gm/cc Plutonium (22,46,22,10) with U-235 Enrichment	2.89	3.00	2.58	2.56

\* These schemes operate at high flux and produce purity Cm-244 by minimizing Pu-241 decay which leads to Cm-242 via Am-241 neutron capture in Am-241.

Plutonium-240 Utilization in Thermal Reactors

In the December 1962 Programming Monthly Report (HW-75925), it was reported that optimum economic use of Pu-240 in thermal reactors might depend on use of fuel elements where geometrical self-shielding of the Pu-240 resonance was reduced. Two possible methods that would reduce resonance self-shielding were suggested: (1) use of small rods or thin plates, and (2) increasing the average  $\Sigma_s$  (scattering cross section) of the fuel by use of a diluent of high  $\Sigma_s$ , such as beryllium oxide. The consequence of reducing resonance self-shielding is to increase the effective cross section of fuel isotopes that have low lying resonances. Although the effective cross sections of all the fuel isotopes are affected, the most pronounced changes are in the effective capture cross section of the Pu-240 isotope which has a 100,000 barn resonance at 1 ev.

It was shown previously that effective utilization of Pu-240 could result in a decrease in fuel cycle costs and an increase in plutonium value. The effective utilization is enhanced by the use of small rods or thin plates which also give an additional advantage; in that, higher fuel specific power should be possible because the ratio of cladding surface (heat transfer area) to fuel volume is increased. These two advantages (increase of the Pu-240 cross section and higher specific power) may be offset by the increased fuel element fabricating and jacketing (FEFJ) charges for small fuel rods.

Some calculations have been made to determine fuel costs and plutonium value for cases where FEFJ costs are increased simultaneously with an increase in fuel specific power and a decrease in resonance self-shielding. A factor that has been omitted in the study is the increased parasitic neutron capture of the cladding material occurring when cladding volume is increased relative to fuel volume. This could be an important factor if cladding were used that had a relatively high neutron capture cross section. However, neutron capture in the cladding would be of less importance with material such as Zircaloy, which was assumed for this study.

In the MELEAGER burnup code, which was used for this study, the resonance self-shielding is characterized by a parameter named SCA. Typically, this parameter varies from values of 0.5 to 2.0 for rod sizes from  $\frac{1}{2}$ -inch to  $\frac{1}{8}$ -inch in diameter and  $\Sigma_s$  values of 0.3 to 0.9. As used in MELEAGER, larger values of SCA correspond to reduced resonance self-shielding.

The definition of SCA used in MELEAGER is:

$$SCA = \sum_{\text{Fuel}} \sigma_s + \frac{A \times B}{A + B}$$

where

$$A = \frac{S_{\text{eff}}}{4V}$$

$$B = \frac{SDFV}{\xi} = \frac{\xi \sum_{\text{mod}} \frac{V_{\text{mod}}}{V_{\text{fuel}}}}{\xi}$$

this may be solved for A giving

$$A = \frac{S_{\text{eff}}}{4V} = \frac{\frac{SDFV}{\xi}}{\frac{SDFV}{\xi} + SCA - \sum_{\text{mod}} \sigma_s} - 1$$

$S_{\text{eff}}$  = fuel surface

$V$  = fuel volume

$\xi$  = logarithmic energy decrement of the moderator

$\sum_{\text{mod}} \sigma_s$  = macroscopic scattering cross section

$S_{\text{eff}}/4V = 1/D$  for a rod of diameter  $D$ , and approximately

$1/(2t)$  for a plate of thickness  $t$ .

If we assume that  $\sum_{\text{mod}} \sigma_s$  of the fuel can be varied by a proper choice of diluent material, then a range of values of SCA is possible for a given fuel rod diameter. For this discussion, we will assume that the neutron capture in diluent material is negligible in comparison to absorption in the fuel isotopes.

It can be seen from Table II that SCA values can range from 0.87 to 1.47 for  $\frac{1}{2}$ -inch diameter fuel rods. From Table II, it appears that to achieve an SCA value of 2 it would require use of a rod somewhat smaller than  $\frac{1}{4}$ -inch diameter, even with  $\Sigma_s$  values of 0.9 ( $\Sigma_s$  iron = 0.933,  $\Sigma_s$  beryllium = 0.865,  $\Sigma_s$  nickel = 1.60).

TABLE II

SCA\* FOR RODS OF SEVERAL SIZES AND THREE VALUES OF  $\Sigma_s$   
(MACROSCOPIC SCATTERING CROSS SECTION) OF THE FUEL REGION

$\Sigma_s$	Rod Size	SCA				
		1/10 in.	1/8 in.	1/4 in.	3/8 in.	1/2 in.
0.3	SCA =	1.63	1.52	1.183	0.99	0.87
0.5	SCA =	1.83	1.72	1.383	1.19	1.07
0.9	SCA =	2.23	2.12	1.783	1.59	1.47

calculated from the equation:

$$SCA = \Sigma_{s, \text{fuel}} + \frac{\left(\frac{1}{D}\right) \frac{SDPV}{\xi}}{\left(\frac{1}{D}\right) + \frac{SDPV}{\xi}}$$

D = fuel rod diameter

$$\frac{SDPV}{\xi} = 2$$

\* SCA is the resonance shielding index used in MELEAGER in calculating SCA,  $\Sigma_s$  is the macroscopic scattering cross section of the fuel + diluent.

Table III shows cladding cost estimates by H. E. Hanthorn from document HW-74304. This information indicates that reduction of fuel rod size from  $\frac{1}{2}$ -inch to  $\frac{1}{4}$ -inch diameter would approximately double fuel element fabricating and jacketing costs. Therefore, cases have been studied where the FEFJ costs were doubled for SCA = 2 in comparison with the SCA = 1 cases.

Suppose we choose a reactor with a water-to-metal ratio of about 2 (SDFV  $\approx$  2). Let us take, as the base case, a  $\frac{1}{2}$ -inch diameter fuel rod with SCA = 1,  $\Sigma_s$  is assumed to be between 0.3 and 0.5. If this fuel element is operated at a specific power of 15 MW/T, the plutonium value for 45, 40, 10, 5 plutonium is \$10.80 per gram fissile. The fuel cycle cost at the plutonium value point is 1.5 mills/kwh<sub>e</sub>.

This case is now compared with one in which the fuel rod diameter is  $\frac{1}{4}$ -inch. Table III shows that the power output for a  $\frac{1}{4}$ -inch diameter rod can be almost triple that for a  $\frac{1}{2}$ -inch rod. This would indicate that a fuel specific power of more than double would be reasonable; so a specific power of 30 MW/T was chosen. For the  $\frac{1}{4}$ -inch fuel element, we have chosen an SCA value of 2, which corresponds to a  $\Sigma_s$  of slightly over 0.9. Fuel element fabricating and jacketing costs are double that for the  $\frac{1}{2}$ -inch diameter rod. If the plutonium is priced at \$10.80 a gram (the plutonium value obtained by an indifference value solution for comparable U-235 enriched and plutonium enriched cases under the conditions mentioned in the previous paragraph) the fuel cost for the  $\frac{1}{4}$ -inch element operated at 30 MW/T specific power would be 1.43 mills/kwh<sub>e</sub> -- a reduction in fuel costs of about 0.1 of a mill, even though FEFJ has been doubled. We might also make a comparison between comparable  $\frac{1}{4}$ -inch (SCA = 2, FEFJ = 2X) U-235 and plutonium element operated at 30 MW/T. The increased FEFJ raises the fuel cost for the system to 1.6 mills per kilowatt-hour and the corresponding indifference value of the plutonium will be greater -- \$13 per gram fissile (Case 4, Table IV). At a plutonium price of \$10.80 per gram, however, the fuel cost for plutonium fueling is lowered to 1.42 MW/kwh<sub>e</sub> while the fuel cost for U-235 enrichment is raised to 1.68 for Case 4.

It should be noted that increasing the specific power has a greater effect than does reduction of the self-shielding in increasing plutonium value and reducing fuel cost (Table IV). Reduction of the self-shielding (corresponding to an SCA increase of 1 to 2) results in an increase in plutonium value of \$1.20 per gram fissile and a reduction in the fuel cycle cost of 0.1 of a mill (Cases 1 and 2, Table IV). If it were possible to increase the specific power of the  $\frac{1}{2}$ -inch rod (corresponding to the SDFV = 2, SCA = 1 case) other variables being held constant, then the fuel cycle cost would drop by 0.2 of a mill and the plutonium value would increase \$1.70 per gram fissile (Cases 1 and 3, Table IV). Note that in Case 2, the FEFJ is the same as in Case 1; so that the effect of reduced self-shielding can be observed without the introduction of other factors such as FEFJ. In reality, it may be necessary to double the FEFJ to obtain an SCA 2. In Case 4, the combined effects of increasing the specific power, reducing the self-shielding, and increasing the fuel element fabricating and jacketing cost are seen. It appears that the dominant effect is due to the increase in specific power and that the reduction in self-shielding of the Pu-240 resonance is of a lesser effect.

TABLE III

ZIRCALOY CLADDING COSTS FOR U-235 ENRICHED AND PLUTONIUM ENRICHED NATURAL URANIUM FUEL ELEMENTS(a)

	3	7	19(c)	27	37	61
$\eta^{(b)}$ =						
Tube I.D. in wall thickness	1.268 0.075	0.830 0.049	0.505 0.030	0.423 0.030	0.361 0.030	0.281 0.030
Heat Transfer area/ft of fuel element	1.1137	1.7006	2.8054	3.4141	4.0780	5.4457
Power Output kw/element	482	775	1380	1830	2386	3877
Fuel Element Cost (1) (2)	3157 3094	3560 3429	4352 4112	5182 4847	6069 5639	7954 7318
\$/kg (1) (2)	48.60 47.63	54.81 52.79	67.00 63.30	79.78 74.62	93.43 86.81	122.46 112.66
\$/lb (1) (2)	22.09 21.50	24.91 23.90	30.45 28.60	36.26 33.40	42.47 39.30	55.66 51.00
~\$/cc at normal oxide density (1) (2)	0.48 0.47	0.54 0.52	0.66 0.62	0.78 0.72	0.92 0.85	1.20 1.10
Packed uranium density =	0.354 lb/in <sup>3</sup>	90% TD				
=	9.81 g/cc	90% TD	TD = 10.9 g/cc			

(a) HW-74304, "Calculated Costs of Fabrication of Plutonium Enriched Fuel Elements," HE Hanthorn.  
 (b) Number of fuel rods in the fuel element cluster.  
 (c) A 19-rod cluster fuel element 10.06 feet long contains 16.1 pounds of UO<sub>2</sub> per foot.

(1) Plutonium enriched natural uranium.  
 (2) U-235 enriched uranium.

TABLE IV

FUEL CYCLE COSTS AND PLUTONIUM VALUES

<u>Case</u>	<u>SDFV</u>	<u>SCA</u>	<u>Specific Power MW/T</u>	<u>FEFJ \$/cc</u>	<u>Fuel Cycle Cost* mills/kwhe</u>	<u>Plutonium Value \$/gram fissile</u>
1	2	1	15	0.61	1.5	10.80
2	2	2	15	0.61	1.4	12.00
3	2	1	30	0.61	1.3	12.50
4	2	2	30	1.22	1.6	13.00

\* The fuel cycle cost that results for U-235 or plutonium enrichment at the plutonium value solution point.

Table V summarizes plutonium values obtained to date in the reduced uranium spatial concentration study. The effect of increased specific power and reduced resonance self-shielding may also be observed by perusal of the fuel costs and plutonium values in this table.

Recent information obtain from HL Physics group has indicated that alpha (the ratio of the capture cross section to fission cross section) for Pu-241, may be greater than previously supposed. If the investigations presently being made substantiate this, the benefits obtained from reduced self-shielding may be less; however, the benefits obtained from operating at increased specific power when large amounts of Pu-240 and Pu-241 are present should remain essentially unchanged.

Fuel Costs of U-233-U-238 Mixtures

Investigation of U-233 enriched natural and depleted uranium fuels was continued. As described in previous monthly reports, the merits of these fuels depend on their cost relative to that of slightly enriched uranium purchased from the diffusion cascade. The fuel costs of U-233 enriched fuels, however, can be based on either of two alternative schemes for the disposition of the spent fuel: (1) selling it for a price compatible with the cost of isotopically separating the U-233 from the U-238 in a diffusion cascade, or (2) purchasing additional U-233 for a recycle operation. The present part of the study examines the first of these choices. The recycle mode is assumed to be bracketed by this case and a U-233 in a pure U-238 case which is also included. (Other alternatives were previously reported and will be included in a summary document.)

TABLE V  
PLUTONIUM VALUES FOR PLUONIUM(1) IN TAILS URANIUM AT OPTIMUM DENSITY

Fuel Cost mills/kwh	Case	SDPV	SCA	Specific Power(3) MW/T	AEC Int %	FEFJ \$/lb	Uranium- Plutonium Density(4)	Comment on Fuel Density	Pu Value \$/gm
1.50	2	2	1	15	4.75	30	Optimum	100% (85-100% little change)	10.80
1.42	3	2	1	15	4.75	30	Optimum	33-50% (Higher densities failed to run)	12.00
1.60	4	3	2	15	4.75	30	Optimum	~66%	11.00
1.30	5	2	1	30	4.75	30	Optimum	~100%	12.70
1.34	6	2	1	30	4.75	30	33%	Note lowered value at 33% density	11.00
1.24	7	2	2	30	4.75	30	Optimum	~50%	12.40
3.06	8	2	1	15	12.50	120	100%		9.45
3.04	9	2	1	15	12.50	120	Optimum		10.40
2.98	10	2	2	15	12.50	120	Optimum	33%	11.80
1.62	11	3	2	15	4.75	30	100%	Density is too high (see Case 4)	10.40
3.16	12	3	2	15	12.50	120	100%	Density is too high	7.90
3.18	13	3	2	15	12.50	120	Optimum	35-60%	10.20
2.50	14	3	2	30	12.50	120	100%	~66% is optimum	11.15
2.49	15	3	2	30	12.50	120	Optimum	~66% is optimum	11.65
2.51	16	3	2	30	12.50	120	10%	~66% is optimum	10.15
1.55	17	3	2	30	4.75	30	Optimum	Optimum is ~85- ~100%	14.40
1.62	18	3	2	30	4.75	30	10%	Optimum is ~85- ~100%	10.80
1.59	19	3	2	30	4.75	30	33%	Optimum is ~85- ~100%	12.50
1.57	20	3	2	30	4.75	30	50%	Optimum is ~85- ~100%	13.60
1.55	21	3	2	30	4.75	30	66%	Optimum is ~85- ~100%	14.35
1.54	22	3	2	30	4.75	30	100%	Optimum is ~85- ~100%	14.35
2.30	23	2	2	30	12.50	120	Optimum	~33- ~50%	11.90

At normal uranium oxide density \$30/lb = \$0.61/cc; \$120/lb = \$2.44/cc.

(1) Plutonium isotopic composition is 45% Pu-239, 40% Pu-240, 10% Pu-241, 5% Pu-242.

(2) Fuel cost at the indifference point - the plutonium value point.

(3) Normalized to 100% fuel density. The specific power per unit volume was held constant as the density was varied.

(4) The fuel density was varied by changing the uranium spatial concentration while the plutonium spatial concentration was held constant. 100% density corresponds to normal uranium oxide density.

The present fuel cost computation is somewhat modified from that previously described. As before, the cost of the initial loading per gram of U-233 is independent of the enrichment level. However, the spent fuel credit is now based on the price that must be assigned to the feed to a diffusion cascade such that the cost of producing 99% pure U-233 is equal to the price initially assigned to the U-233. In previous work, the spent fuel price was such that the cost of producing an enrichment equal to the initial enrichment of the reactor was the same as that originally assumed. Therefore, the spent fuel credit has been diminished which has, in turn, increased the minimized total fuel costs by roughly 0.1 mill/kwh<sub>e</sub> while decreasing the fuel value by roughly \$1 per gram.

Table VI contains the minimized fuel costs calculated for batch irradiation in a pressurized water reactor simulation. Also shown is the "indifference U-233 value" which is the price that must be assigned to the U-233 for the fuel cost of the U-233 enriched fuel loading to equal that of the U-235 enriched loading. Only batch irradiation was considered because the fuel discharged from a graded irradiation is so depleted in U-233 that the two-isotope spent fuel assumption implicit in this analysis is extremely invalid in the graded cases.

The data presented in Table VI show that the fuel costs of all of the U-233 enriched fueling modes are quite sensitive to the assigned price of the U-233. The fueling mode has little importance when the price is low (as shown by the fuel costs for \$5 per gram U-233) but, when the price is increased, the low priced U-235 in natural or depleted uranium becomes more beneficial (as shown by the fuel costs for \$15 per gram U-233). The indifference values also reflect the advantage of the cheap U-235 contained in natural and depleted uranium as the reactor operator can afford to pay approximately \$1 per gram more to use U-233 in this kind of fueling mode. It is interesting to note that the indifference value for U-235 in U-238 and for U-233 in U-238 (modes 1 and 2) indicate that the performance of U-233 is about 25% superior in this reactor.

The importance of the lower separative duty required for the isotopic separation of U-233 from U-238 is shown in Table VII wherein fuel costs were calculated for separative duty costs of \$10.71 per kg and \$30 per kg. The indifference values associated with the latter figure were not calculated, but would appear to be reduced by about \$2 per gram. It should be noted that all of these calculations are optimistic because the figure of \$10.71 per kg ignores inventory and turnaround costs, and because an optimum tails composition is presumed. This is indicative, however, of the productivity of an isotopic separation scheme with low inventories such as a successful centrifuge development.

TABLE VI

MINIMIZED TOTAL FUEL COSTS AND CORRESPONDING U-233 VALUES FOR THE  
BATCH IRRADIATION OF U-233-U-238 MIXTURES IN THE SPWR<sup>(1)</sup>

Plutonium Credit = \$10.24 per gram fissile  
 FEFJ = \$40 per pound fuel  
 Depreciating Interest = 12.5 percent  
 Nondepreciating Interest = 4.75 percent  
 Cost of Separative Duty for  
 U-233-U-238 Separation = \$10.71 per kg

Fuel Loading	Fuel Costs, mills/kwh <sub>e</sub> , for Different Assigned U-233 Prices			Indifference Value \$/gm
	\$5/gm	\$10/gm	\$15/gm	
1. Cascade Enriched Uranium	2.21	2.21	2.21	8.00 (U-235)
2. U-233 Enriched U-238	1.35	2.17	2.93	10.25 (U-233)
3. U-233 Enriched Depleted Uranium <sup>(2)</sup>	1.34	2.04	2.71	11.25 (U-233)
4. U-233 Enriched Natural Uranium <sup>(3)</sup>	1.39	2.06	2.67	11.20 (U-233)

(1) Pressurized Water Reactor Simulation as Described in TID-8502.

(2) 0.4% U-235 Priced at \$3 per kg Uranium.

(3) Natural Uranium Priced at \$10 per kg Uranium.

TABLE VII

MINIMIZED TOTAL FUEL COSTS FOR BATCH IRRADIATION OF U-233-U-238  
MIXTURES IN THE SPWR<sup>(1)</sup> - U-233 PRICED AT \$10/gm

Plutonium Credit = \$10.24 per gram fissile  
 FEFJ = \$40 per pound fuel  
 Depreciating Interest = 12.5 percent  
 Nondepreciating Interest = 4.75 percent

<u>Fuel Loading</u>	Fuel Cost, mills/kwh <sub>e</sub> , for Different Separative Duty Costs Associated with the Re-Enrichment of the U-233 in the Spent Fuel	
	<u>\$10.71/kg</u>	<u>\$30/kg</u>
1. Cascade Enriched Uranium	2.21	2.21
2. U-233 Enriched U-238	2.17	2.32
3. U-233 Enriched Depleted Uranium <sup>(2)</sup>	2.04	2.20
4. U-233 Enriched Natural Uranium <sup>(3)</sup>	2.06	2.21

(1) Pressurized Water Reactor Simulation as Described in TID-8502.

(2) 0.4% U-235 Priced at \$3 per kg Uranium.

(3) Natural Uranium Priced at \$10 per kg Uranium.

The effect of the economic environment on the preceding computations is shown in Table VIII. This table shows the minimized total fuel costs for cascade enriched uranium and for the U-233 enrichment of depleted uranium at three price levels for six different economic environments. (A new environment is made by perturbing one of the values used in the base set.) The most striking conclusion shown by the data of Table VIII is that the U-233 indifference value is essentially independent of the economic environment.

TABLE VIII

MINIMIZED TOTAL FUEL COSTS AND CORRESPONDING U-233 VALUES FOR THE BATCH IRRADIATION OF A U-233-DEPLETED URANIUM MIXTURE IN THE SPWR(1)

Depleted Uranium (0.4%) at \$3 per kg  
 Plutonium Credit = \$10.24 per gram fissile  
 FEFJ = \$40 per pound fuel  
 Depreciating Interest = 12.5 percent  
 Nondepreciating Interest = 4.75 percent  
 except as noted

Perturbated Economic Parameter and Value	Fuel Costs, mills/kwh <sub>e</sub>				Indifference Value \$/gm
	For Enriched Uranium Fuel Loading	For Different Assigned U-233 Prices, \$/gm			
		5	10	15	
1. None	2.21	1.34	2.04	2.71	11.25 (U-233)
2. FEFJ = \$20/lb	1.86	1.06	1.74	2.40	10.90 (U-233)
3. FEFJ = \$60/lb	2.50	1.61	2.32	3.00	11.35 (U-233)
4. Nondepreciating Interest = 10%	2.57	1.48	2.34	3.17	11.35 (U-233)
5. Depreciating Interest = 10%	2.14	1.27	1.96	2.63	11.30 (U-233)
6. Depleted Uranium Price = \$6/kg	2.21	1.36	2.06	2.73	11.10 (U-233)

(1) Pressurized Water Reactor Simulated as Described in TID-8502.

Code DevelopmentUninteracted Graded Irradiation

Uninteracted graded burnup is a form of graded irradiation determined by the specific fuel and has a flux spectrum which varies with time accordingly. It is patterned on an extremely well moderated reactor in which none of the excess neutrons are lost to the control system. The isotope concentrations in adjacent fuels do not influence the neutron spectra seen by each fuel. (Hence, "noninteracted".)

Uninteracted graded calculations are made by the PROTEUS code by normalizing MELEAGER constant power variable flux burnup calculations to constant flux burnup. The proper flux level for graded operation had been considered to be the time average of the flux over the reactivity limited lifetime as determined by MELEAGER. This is not quite correct, but it is not greatly in error unless the flux changes during the constant power irradiation are very large as, for example, in the decay heat source production program.

The correct formulation is presently being programmed as the GEFLUX subroutine of PROTEUS. It is based as having an average specific power in graded irradiation that is exactly equal to the constant specific power of the MELEAGER case. This is possible only when the graded flux is the cross section weighted time average. That is, the weighting factor is the product of the macroscopic fission cross section and the energy of fission of each isotope summed over the entire fuel and is, therefore, a rather complicated function. However, the formula can be expressed in terms of the flux at constant power only in the form

$$\phi^* = \frac{T}{\int_0^T \frac{dt}{\phi(\tau)}}$$

where

$\phi^*$  = graded flux (i.e., constant over fuel lifetime)

$\phi$  = batch flux (i.e., variable over fuel lifetime)

$T$  = reactivity limited lifetime.

It can be shown that if  $\phi$  is constant or varies slowly, the above formula reduces the formula for the time average.

PROTEUS

In addition to converting MELEAGER batch irradiation data to uninteracted graded data, the PROTEUS code is often used to develop batch cases for different terminal  $k_{\infty}$  values. Although MELEAGER calculates data exactly on a six-month interval, the PROTEUS computation did not previously take advantage of this but, instead, was kept general. Because the new version of MELEAGER retains this feature, PROTEUS has been revised and will now check the data and, if six-month data is given, will accept it and curve fit only in the region of the terminal point. This feature will increase both the speed and accuracy of the batch computations.

MELEAGER

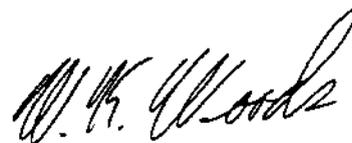
Refinements of the Runge Kutta analysis were completed and provisions for batch and graded end point calculations were incorporated in MELEAGER. Output format was provided for QUICK, PROTEUS, and PLOTTER codes that improved their usefulness. Compatible chain systems have been organized including the above codes. JASON is yet to be converted to the compatible systems.

Several MELEAGER cross section libraries have been assembled for use in current reactor studies.

PLOTTER

Considerable time has been spent on adding to, and improving PLOTTER sub-routines.

A program by which the line items of the Fuel Properties Table (a PLOTTER subroutine) will be selected, by group or by line designation, has been submitted for compilation. PLOTTER input routine has been revised to accept the DECAY plots programmed and being compiled by EDPO personnel.



Manager,  
Programming

WK Woods:jm

RADIATION PROTECTION OPERATION  
REPORT FOR THE MONTH OF MARCH 1963

A. ORGANIZATION AND PERSONNEL

Lena R. Smith was granted a medical leave of absence from Radiological Development and Calibrations. R. M. Joyce Bernard transferred from Internal Dosimetry to the Physics and Instrument Laboratory. Linda J. Dalton transferred from the Biology Laboratory to Internal Dosimetry.

B. ACTIVITIES

Occupational Exposure Experience

There were no new plutonium deposition cases confirmed during the month. The total number of plutonium deposition cases that have occurred at Hanford is 316 of which 229 are currently employed.

Four plutonium incidents required special bioassay sampling of personnel involved to determine if internal deposition occurred. The following is a brief description of each incident. Bioassay results are now pending.

A CPD employee received plutonium nasal contamination up to 200 d/m at the 234-5 Building on March 1, 1963, as a result of a glove rupture. Contamination on the skin ranged from 2,000 to 10,000 d/m and >40,000 d/m on his coveralls.

Plutonium contamination of 80,000 d/m was spread over ~12 square feet of floor in the 231 Building on March 1, 1963, when a HL employee transferred a bag of plutonium oxide into an airlock without surveying it. The employee received 20,000 d/m contamination on his coveralls; however, no nasal contamination was detected.

A CPD employee received a plutonium contaminated injury at the 234-5 Building March 11, 1963, while cutting apart a fluorinator tube in the maintenance hood. The initial survey of the wound with portable survey equipment indicated ~5,000 d/m plutonium contamination. Examination at the plutonium wound monitor indicated 0.014  $\mu\text{c}$  plutonium present at the wound site. After two excisions  $<1 \times 10^{-4} \mu\text{c}$  (<1% of the MPBB\*) remained in the wound.

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\*MPBB plutonium (bone as reference) = 0.04  $\mu\text{c}$ .

Nasal contamination ranging from 3,000 to 5,000 d/m was received by a CPD technician when he opened an unmarked ice cream carton containing plutonium oxide at the 234-5 Building on March 18, 1963. Three nasal irrigations were required to reduce nasal contamination to background. Contamination on his face, chest and clothing exceeded 40,000 d/m. A second employee received nasal contamination ranging from 212 to 364 d/m while assisting in the removal of clothing from the above technician. The plutonium oxide was spread in levels >40,000 d/m over ~300 square feet of floor. Very little contamination was found on other surfaces such as hoods or benches in the room.

Several other incidents of significance included the following:

A HL radiometallurgy technician was exposed to fission product contamination at the 327 Building on March 4, 1963, while dismantling a micro-hardness tester. A survey when the work was completed revealed general contamination on his skin from 10,000 to 20,000 c/m with a maximum of 25,000 c/m on his hair. Nasal contamination ranged from 4,000 to 10,000 c/m. The contaminant appeared to be large particles. When the 30-minute job was set up, no respiratory protection was specified since the piece of equipment had been cleaned by immersion in alcohol. Apparently the contamination came from the inside of the instrument. Examination at the whole body counter revealed a trace of Zr-Nb<sup>95</sup>. The estimated transient deposition is <<1% of the MPBB.\*

During the course of a personal survey of the PRTR on March 22, 1963, 20,000 c/m beta-gamma contamination was found inside the right shoe of a technologist. Since there was no apparent explanation as to the source or cause of the contamination, a home survey was scheduled. No contamination was found.

#### Environmental Experience

Concentrations of fallout materials in the air of the Pacific Northwest ranged from 4 to 7 pc/m<sup>3</sup> of air, a significant reduction from ~8 pc/m<sup>3</sup> noted during January and February.

Two aerial surveys were made. One flight followed standard pattern 5-S for background measurements. The second flight was made to measure river activity from Priest Rapids to the mouth of the Snake River. No activity above previously observed background was noted.

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\*MPBB Zr-Nb<sup>95</sup> (total body) = 20 µc.

The following 200 biological, produce, and food samples were obtained for radiochemical analysis:

Milk	68 samples	158 gallons
Oysters	2 samples	4 pounds
Ground Round	2 samples	4 pounds
Beef Thyroids	35 sets	
Fish	93 samples	

Liquid wastes from the Purex operation discharged to swamps have frequently contained abnormally high concentrations of radioactive materials since the first of the year. This situation was called to the attention of the operating component.

#### Studies and Improvements

The second phase of the  $I^{131}$  uptake study was initiated during the month. The first phase of this study was concerned with the uptake by the human thyroid of  $I^{131}$  in milk. The milk, provided for the study by Biology, was produced from dairy cows fed carrier-free  $I^{131}$ . Thyroid burden measurements were made on eight adult male employees who voluntarily drank the  $I^{131}$  labeled milk. Both chronic and single intakes were involved. These data are now being analyzed. The second phase of this study involves the effect of  $I^{127}$  on thyroid uptake of  $I^{131}$ . The milk in Phase I was produced by cows on a low stable iodine ( $I^{127}$ ) diet. The milk consumed this month was produced after increasing the  $I^{127}$  content in the cows' diet to 5 mg  $I^{127}$ /day. It is anticipated that single intakes by the volunteers will be sufficient to determine any difference in uptake of  $I^{131}$  caused by the increased level of stable iodine. Additional single intakes of  $I^{131}$  labeled milk at several increased levels of  $I^{127}$  are planned.

Appropriation requests were prepared for the remaining portable instrumentation required for river temperature and dye studies.

Additional aerial photos were received during the month so that complete coverage of the land area around the Plant perimeter within a radius of 50-60 miles is now available.

Emergency film processing materials are now being stored in the 747 Building. These materials include developer, fix and calibration film. The latter item will be changed on a regular four-week badge schedule. A densitometer, an X-ray coding machine and a payroll number punching machine will also be stored in this facility.

Thirty ORNL criticality dosimeters were transferred to the Albuquerque, New Mexico Operations Office.

The experimental apparatus for the determination of the dose to the various parts of the body (relative to the dose to personnel dosimeters) from a simulated reactor beam is being assembled in the 3745-B Building. The stand to support the phantom was completed and an actual gun barrel, tube, and nozzle assembly as used in the reactors was located.

Five Sandia Pulsed Reactor Facility irradiations were obtained on March 18 and 19 at Albuquerque, New Mexico. The purpose of these irradiations was to further test the performance of the Hanford Criticality Dosimeter neutron spectrum measuring foils and the personnel dosimeter neutron spectrum measuring foils. Table I gives the characteristics of the five bursts obtained.

TABLE I

SPRF Reactor Burst Characteristics

<u>Pulse Width (Half Maximum)</u>	<u>Total Number of Fissions</u>	<u>Neutron Dose Rate* rad/sec</u>	<u>Gamma Dose Rate* r/sec</u>
50 $\mu$ s	$2.0 \times 10^{16}$	$6 \times 10^6$ and $1 \times 10^7$	$1 \times 10^6$ and $2 \times 10^6$
110 $\mu$ s	$1.82 \times 10^{16}$	$2 \times 10^6$ and $5 \times 10^6$	$5 \times 10^5$ and $8 \times 10^5$
500 $\mu$ s	$1.42 \times 10^{16}$	$7 \times 10^5$ and $2 \times 10^6$	$2 \times 10^5$ and $3 \times 10^5$
1,200 $\mu$ s	$0.86 \times 10^{16}$	$3 \times 10^5$ and $5 \times 10^5$	$5 \times 10^4$ and $8 \times 10^4$
25,000 $\mu$ s	$0.42 \times 10^{16}$	$3 \times 10^4$ and $8 \times 10^4$	$2 \times 10^3$ and $5 \times 10^3$

The neutron spectrum measurements were accomplished satisfactorily and will be reported in detail later.

Performance tests were conducted on a small  $\text{BF}_3$  tube of reduced neutron sensitivity. The initial response indicates a sensitivity reduction of about 35 times compared to the standard  $\text{BF}_3$  tube now in use on the BFQ monitoring instruments. With the use of this low sensitivity tube, the maximum thermal neutron dose ranges on the BFQ instrument would be 70, 700, and 7,000 mrem/hr. Additional efforts are being directed toward reducing the sensitivity of the tube by a factor of 100.

The silicon diode neutron dosimeters were exposed to five bursts of radiation from the Sandia Pulsed Reactor. The bursts ranged from about  $4 \times 10^{15}$  fissions to  $2 \times 10^{16}$  fissions. Measurements of the silicon diode neutron sensitivity at various neutron energies also continued during the month.

One set of eight silicon diodes was exposed to a burst of  $2 \times 10^{16}$  fissions last January and exposed to a second burst of  $2 \times 10^{16}$  fissions this month. The results of both exposures are given in Table II.

\*At exposure positions.

TABLE II

Silicon Diode - Burst Exposure Results

<u>First Exposure</u>		<u>Second Exposure</u>	
<u>Expected Dose</u>	<u>Measured</u>	<u>Expected Dose</u>	<u>Measured</u>
<u>(rads)</u>	<u>Neutron Dose</u>	<u>(rads)</u>	<u>Neutron Dose</u>
	<u>(rads)</u>		<u>(rads)</u>
2715	2598	1370	390
2715	2629	1370	385
1341	1557	595	391
1341	1535	595	357
560	617	287	306
560	617	287	300
289	259	198	226
289	250	198	219

The diodes exposed to the largest doses in January could not accept all of the increased damage given during the second exposure. These diodes appear to have approached a saturation point somewhere near 2,000 rads. The diodes previously exposed to 617 rads were able to accurately measure another 287 rads, and those exposed to 289 rads previously were able to measure an additional 198 rads. The accuracy in the measurements appears to be about  $\pm 10\%$ , probably as good as the expected dose measurement.

The sensitivities of a sampling of each type of diode were measured using the Van de Graaff accelerator for neutron exposures. The results are shown in Table III.

TABLE III

Sensitivity of the Diodes

<u>Crystal Type</u>	<u>Resistivity</u>	<u>Conductivity</u>	<u>Base Width</u>	<u>Sensitivity*</u>
	<u>(ohm-cm)</u>	<u>Type</u>	<u>(in)</u>	<u>% Change/rad</u>
Float Zone	150	p	0.040	0.071
" "	150	p	0.070	0.077
" "	75	p	0.040	0.056
" "	75	p	0.070	0.048
" "	150	n	0.040	0.078
" "	150	n	0.070	0.200
" "	75	n	0.040	0.067
" "	75	n	0.070	0.162
Pulled	150	p	0.070	0.246
"	150	p	0.040	0.065
"	75	n	0.070	0.247
"	75	n	0.040	0.035

\*Average of several measurements.

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The "pulled" p type diodes were somewhat more sensitive than the float-zone p or n type diodes. All of the diodes with a base width of 0.040" have about the same sensitivity.

The Biology Laboratory has requested additional dose measurements of simulated bone material. The dose rate from organs removed from sheep which had been fed Cs<sup>137</sup> were measured through the use of ionization chamber pencils. Special dose rate measurements were requested on three animals which are to be sacrificed.

#### C. RELATIONS

Four suggestions were submitted by personnel of the Radiation Protection Operation during the month. Two suggestions were adopted; four were rejected. Two suggestions are pending evaluation.

Safety meetings were held throughout the Section during the month. Minor injury frequency, vehicle accident reports, emergency evacuation procedures, fire warning procedures, and "Physiological Hazards of D<sub>2</sub>O" were discussed. Two films entitled "Safety in the Chemical Laboratory" and "Safety Everywhere all the Time" were also shown.

A total of 13 radiation protection orientation lectures were presented to 150 personnel. One talk on the new Hanford film badge dosimeter was presented to Chemical Effluents Technology personnel. L. F. Kocher presented two talks on the new Hanford film badge dosimeter to the industrial medical nurses. A refresher talk was given to personnel in Line Maintenance of the Electrical Utility Operation.

#### D. SIGNIFICANT REPORTS

- HW-76525-2 - "Radiological Status of the Hanford Environs for February 1963" by R. F. Foster.
- HW-76638 - - "Radioactive Contamination in Liquid Wastes Discharged to Ground at the Separations Facilities through December 1962" by G. E. Backman.
- HW-76944 - - "The New Hanford Film Badge Dosimeter" by L. F. Kocher, P. E. Bramson, and C. M. Unruh.
- HW-77148 - - "Monthly Report for March 1963 - Radiation Monitoring Operation" by A. J. Stevens.

PERSONNEL DOSIMETRY AND RADIOLOGICAL RECORDS

<u>External Exposure Above Permissible Limits</u>	<u>March</u>	<u>1963</u>
Whole Body Penetrating	0	0
Whole Body Skin	0	0
Extremity	0	0
 <u>Hanford Pocket Dosimeters</u>		
Dosimeters Processed	7,876	18,978
 <u>Hanford Beta-Gamma Film Badge Dosimeters</u>		
Film Processed	9,559	27,579
Results - 100-300 mrad	156	487
- 300-500 mrad	14	61
- Over 500 mrad	1	15
Lost Results	24	85
Average Dose per Film Packet - mrad (ow)	5.74	6.08
- mr (s)	34.68	33.79
 <u>Hanford Neutron Film Badge Dosimeter</u>		
<u>Slow Neutron</u>		
Film Processed	1,665	4,873
Results - 50-100 mrem	1	4
- 100-300 mrem	1	1
- Over 300 mrem	0	0
Lost Results	13	42
 <u>Fast Neutron</u>		
Film Processed	421	1,195
Results - 50-100 mrem	87	149
- 100-300 mrem	30	336
- Over 300 mrem	0	4
Lost Results	10	21
 <u>Hand Checks</u>		
Checks Taken - Alpha	36,763	111,561
- Beta-Gamma	59,231	179,103
 <u>Skin Contamination</u>		
Plutonium	25	83
Fission Products	43	85
Uranium	0	0
Tritium	0	0

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1103175

Whole Body Counter

<u>Subject</u>	<u>Number of Examinations</u>			
	<u>747-A WBC</u>	<u>1963</u>	<u>Mobile WBC</u>	<u>1963</u>
GE Employees				
Regular	35	158		
Incident Cases	7	26		
Terminations	4	19		
New Hires	6	6		
Special Studies	48	75		
Non-Employees				
Children	0	1		
Visitors	1	3		
Environmental Studies	0	4		
	<u>101</u>	<u>292</u>		

Bioassay

<u>Analysis</u>	<u>Current Reporting Limit</u>	<u>Results Above Reporting Limit</u>		<u>Samples Assayed</u>	
		<u>March</u>	<u>1963</u>	<u>March</u>	<u>1963</u>
Plutonium	2.2x10 <sup>-8</sup> µc/sample	113	329	626	2,239
Fission Product	3.1x10 <sup>-5</sup> µc/sample	1	25	563	2,162
Strontium	3.1x10 <sup>-5</sup> µc/sample	0	0	0	0
Tritium	5.0 µc/l	47	383	176	751
Uranium	0.14 µgm/l	0	0	80	376
Special Studies		0	0	50	229

Calibrations

	<u>Number of Units Calibrated</u>	
	<u>March</u>	<u>1963</u>
Portable Instruments		
CP Meter	1,009	3,123
Juno	253	804
GM	543	1,676
Other	205	611
Audits	98	310
	<u>2,108</u>	<u>6,524</u>
Personnel Meters		
Badge Film	648	2,168
Pencils	165	570
Other	288	818
	<u>1,101</u>	<u>3,391</u>

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	<u>Number of Units Calibrated</u>	
	<u>March</u>	<u>1963</u>
Miscellaneous Special Services	1,315	2,853
Total Number of Calibrations	4,524	12,768

  
Manager  
RADIATION PROTECTION

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FINANCE AND ADMINISTRATIONACCOUNTINGCost Accounting

Adjustments in Operating Cost and Capital Equipment control budgets to reflect the most recent financial plan issued by RLOO-AEC show:

	Previous Control Budget	Increase (Decrease)	New Control Budget
<u>04 Program</u>			
Plutonium Recycle Program	\$6 500	\$ 515	\$7 015
Reactor Fuels & Materials	2 970	(165)	2 805
Plutonium Ceramics Research	200	35	235
DR-1 Loop	--	21	21
Euratom	5	1	6
Low & Inst. Waste Studies	125	65	190
High Level Waste Studies	403	(65)	338
Waste Calc. Demonstration Prototype	467	130	597
Environmental Studies	--	11	11
Capital Equipment	1 144	(116)	1 028
<u>05 Program</u>			
Plutonium Physical Met. Res.	80	(20)	60
Capital Equipment	120	20	140
<u>08 Program</u>			
Fission Product Prod. Study	100	(3)	97
Capital Equipment	--	3	3

A reduction of \$235,000 contained in the financial plan for the Gas-Cooled Power Reactor Program was not reflected in the new control budget because the AEC indicates intent to restore a major portion of these funds.

An authorization in the amount of \$57,600 to perform Plutonium Inhalation Studies for the U. S. Air Force during the period August 1, 1962 to August 1, 1963 was received during the month from RLOO-AEC.

Activities for which special accounting codes were established are described below:

- .3K Study of AEC Plutonium Power Reactor Fuel Capability. A work order was issued by RLOO-AEC for \$7,500 to cover travel, subsistence, salaries and overhead of R. D. Widrig, L. E. Mills, J. B. Burnham, E. A. Evans and F. W. Albaugh.

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1103178

- .3L Participation by J. M. Nielsen in Operation Roller Coaster. Several trips are anticipated. No dollar limitation has been established, but billing will be for salary, indirect costs, travel and subsistence.

A new program code (.16) was established during the month for "Environmental Studies" sponsored by the Division of Reactor Development.

The major effort in preparing Hanford Laboratories' Budget for FY 1965 and Revision of Budget for FY 1964 was completed with the publication of research and development proposals for the O2, O4, O5, O6 and O8 Programs. Review copies were provided HAPO management and RLOO-AEC. Remaining phases of the budget preparation are under way and will be completed during April.

#### General Accounting

Following is the status of letters seeking AEC concurrence to certain actions not directly covered by the Prime Contract:

AT-288	Participation in Standardizing Activities, ASTM Committee C-6 - Pyrolytic Materials (R. E. Nightingale)	Approved 3-13-63
AT-289	AEC Monograph on "Radiation Effects on Structural Materials" (S. H. Bush)	To AEC 3-25-63

Work authorized by letter under Agreement AT-6 was as follows:

	<u>Date Accepted</u>
Burst Test of Irradiated Zircaloy Pressure Tubes for AECL	3-5-63
Study of AEC Plutonium Power Reactor Fuel Capability	3-4-63
Participation in Operation Roller Coaster - J. M. Nielsen	3-25-63

The upward trend in travel activity continues:

<u>Number of Trips Started</u>	<u>March</u>	<u>FY to Date</u>
FY 1962	117	836
FY 1963	134	969

In every month this fiscal year except one, travel has exceeded the corresponding period of the previous year.

Revised OPGs issued were:

<u>OPG No.</u>	<u>Title</u>
1.1	Organization and Policy Guide System
44.7	Design Review Councils
55.4.6	Printing and Duplicating
22.3.1 (pp. 5, 6)	Approval Authorizations
7.8 (pp. 11, 12, 13, 14)	Control of Documents Classified Secret and Confidential

During the month \$201,719 were transferred to Plant and Equipment accounts from Work In Progress accounts.

Hanford Laboratories' material investment at March 1, 1963 totaled \$26.6 million as detailed below:

	(In Thousands)
SS Material	\$24 694
Reactor and Other Special Materials	1 614
Spare Parts	256-1)
	<u>\$26 564</u>

(1- Includes a reserve of \$79,340)

The value of nuclear material consumed in research this fiscal year to March 1, 1963 by Hanford Laboratories is \$3.3 million comprised as follows:

2000 Program	\$ 995
3000 Program	894
4000 Program	1 395
	<u>\$ 3 284</u>

Results reported during the month on the physical inventory of movable catalogued equipment in the custody of Finance and Administration Operation and Physics and Instruments Laboratory are presented below:

	<u>Finance &amp; Admin.</u>		<u>Physics &amp; Instruments</u>	
	<u>Quantity</u>	<u>Value</u>	<u>Quantity</u>	<u>Value</u>
Balance Prior to Inventory	1 352	\$1 330 542	2 614	\$2 407 349
Reconciled Adjustment	(127)	(20 086)	(25)	(11 367)
Adjusted Book Balance	1 225	1 310 456	2 589	2 395 982
Physical Inventory	1 221	1 309 491	2 583	2 392 368
Missing Equipment	<u>4</u>	<u>\$ 965</u>	<u>6</u>	<u>\$ 3 614</u>
% of Missing Equipment to Adjusted Book Balance	0.32%	0.07%	0.23%	0.15%

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1103180

Certification inventory reports for the quarter ending March 31, 1963 were prepared and forwarded to Hanford Laboratories' holders of Other Special Materials for completion and reconciliation with Hanford Laboratories' Property Accounting records.

The heavy water inventory at the end of March shows a loss of 1,185 pounds amounting to \$16,365. Heavy water scrap valued at approximately \$1,400 was generated during the month.

Laboratory Storage Pool activity is summarized below:

<u>Equipment</u>	<u>Current Month</u>		<u>FY to Date</u>	
	<u>Quantity</u>	<u>Value</u>	<u>Quantity</u>	<u>Value</u>
Beginning Balance	1 075	\$720 809	1 081	\$ 562 200
Items Received	183	70 963	1 089	611 357
Items Reclaimed by Custodians	(38)	(51 266)	(133)	(175 440)
Equipment Transfers	(29)	(11 812)	(182)	(62 946)
Items Disposed of by PDR	(6)	(1 041)	(122)	(16 850)
Items Disposed of by Excess	(5)	(10 982)	(553)	(160 385)
Adjustment				(41 265)
	<u>1 180</u>	<u>\$716 671</u>	<u>1 180</u>	<u>\$ 716 671 -1)</u>

(1- Includes 110 items valued at \$81,620 on loan at March 31.

During the month, 43 items valued at \$18,565 were loaned and/or transferred in lieu of purchases. A total of 278 items valued at \$134,480 have been redirected to useful purposes this fiscal year. Operating cost for the same period was \$12,778 indicating a net saving of \$121,702 this fiscal year.

Total investment in equipment and materials in the Laboratory Storage Pool at March 31, 1963 was \$1.3 million including Reactor and Special Materials valued at \$326,438 and other materials valued at \$241,603.

Action as indicated occurred during the month on the following projects:

New Money Authorized Hanford Laboratories

CAH-962	Low Level Radiochemistry Building	\$(13 500)*
CAH-995	309 Building Air Conditioning Modification	18 000

\*Transfer of detail design of special hoods and radiation monitoring devices from General Electric Company to Architect Engineer.

The following contracts were processed during the month:

CA-374 F. I. Badgley  
CA-375 J. L. Powell

CA-383 Rutherford Aris  
 DDR-164 Speer Carbon Company  
 DDR-165 Van's Metal Spinning Company  
 SA-273 Union Carbide Corporation  
 SA-274 Great Lakes Carbon Corporation  
 SA-275 Speer Carbon Company  
 MRO- 58 Beckman Instruments, Inc., Spinco Division  
 MRO- 59 Beckman Instruments, Inc., Scientific and Process  
 Instrument Division

During the month, \$2,500 Assistance to Hanford funds were transferred to Hanford Laboratories from Chemical Processing Department; concurrently, authorization ATH-HL-1-63-A, Consultations Service by Dr. Poritsky, was increased \$2,000 for a total authorization of \$7,000.

Accounting for expense project CAH-822 - Pressurized Gas-Cooled Loop Facility was transferred from CE&UO to Hanford Laboratories during the month.

#### Personnel Accounting

The following employees will retire on April 1, 1963:

B. L. Beaver	Normal Retirement
S. E. Irving	Normal Retirement
A. C. Reddell	Optional Retirement

F. B. Quinlan received a patent award, HWIR 1486, for "A Method of Preparing a Metallic Surface to Prevent Galling During a Cold Forming Operation."

Personnel statistics follow:

#### Number of Hanford Laboratories Employees

##### Changes During Month

	<u>Total</u>	<u>Exempt</u>	<u>Nonexempt</u>
Employees on payroll at beginning of month	1 639	698	941
Additions and transfers in	36	7	29
Removals and transfers out	28	6	22
Employees on Payroll at end of month	<u>1 647</u>	<u>699</u>	<u>948</u>

#### Overtime Payments During Month

	<u>March</u>	<u>February</u>
Exempt	\$ 8 145	\$ 7 746
Nonexempt	28 631	29 701
Total	<u>\$ 36 776</u>	<u>\$ 37 447</u>

#### Gross Payroll Paid During Month

Exempt	\$ 667 024	\$ 667 463
Nonexempt	654 180	522 803
Total	<u>\$1 321 204</u>	<u>\$1 190 266</u>

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1103182

<u>Participation in Employee Benefit Plans at Month End</u>	<u>March</u>		<u>February</u>	
	<u>Number</u>	<u>Percent</u>	<u>Number</u>	<u>Percent</u>
Pension	1 475	99.4	1 461	99.4
Insurance Plan - Personal	401		397	
- Dependent	1 241	99.9	1 235	99.8
U. S. Savings Bonds				
Stock Bonus Plan	155	42.6	157	43.4
Savings Plan	67	4.1	69	4.2
Savings and Security Plan	1 141	88.7	1 136	88.7
Good Neighbor Fund	1 178	71.4	1 178	71.7

Insurance Claims

<u>Employee Benefits</u>	<u>Number</u>	<u>Amount</u>	<u>Number</u>	<u>Amount</u>
Life Insurance	0	\$ -0-	1	\$16 800
Weekly Sickness and Accident	11	629	10	843
Comprehensive Medical	72	3 922	81	4 422
<u>Dependent Benefits</u>				
Comprehensive Medical	<u>120</u>	<u>10 433</u>	<u>137</u>	<u>11 938</u>
Total	<u>203</u>	<u>\$14 984</u>	<u>229</u>	<u>\$34 003</u>

TECHNICAL ADMINISTRATIONEmployee Relations

Thirty nonexempt requisitions were filled during the month; 22 remain to be filled.

Suggestion plan activity showed 55 suggestions received, 22 adopted, 16 rejected and 174 in process at month end.

Information and Presentations

Visitors Center activity is summarized below:

March attendance	1 533
Average per day open (March)	69.6
Cumulative attendance since 6-13-62	40 025
Conducted groups	6 (totaling 220 people)

Plant tour activity totaled:

	<u>Number</u>	<u>Total People</u>
General Public Relations Tours	12	278
Special Tours	4	51
	<u>16</u>	<u>329</u>

Thirty-two locations aside from the Visitors Center have been delineated and cleared for touring. Personnel in all but five components involved (Biology, Shielded Analytical Lab, High Level Cells, Van de Graaff Generator, and Exponential Pile) wish to handle their own presentations. Presentations have been written for the first three.

In addition to the above, tour stops in the 700 Area have been arranged for commercial and stenographic high school students. Examples include Central Duplicating, Data Processing, Mail Room and Stenographic Practices.

Document information flow during the month was comprised of 1,646 titles (10,129 copies) received at Hanford and 55 titles (5,175 copies) sent off-site.

The AEC Division of Technical Information Extension has requested use of the Hanford prepared bibliography "Review of Power and Heat Reactor Designs, Domestic and Foreign," for AEC information displays in Bogota, Columbia, and Vienna, Austria.

#### Professional Placement

Advanced Degree - Nine Ph.D. applicants visited HAPO for employment interviews. Five offers were extended; one acceptance and no rejections were received. Seven offers are currently open.

BS/MS (Direct Placement) - Nine offers were extended. Three acceptances and one rejection were received. Twelve offers are currently open.

BS/MS (Program) - Sixty-one offers were extended. Twenty-two acceptances and 16 rejections were received. Current open offers total 162.

Technical Graduate Program - Five technical graduates were placed on permanent assignment. Two new members were added to the roll and two members terminated. Current program strength is 41.

#### FACILITIES ENGINEERING

##### Projects

At month's end Facilities Engineering Operation was responsible for nine active projects having total authorized funds in the amount of \$6,300,500. The total estimated cost of these projects is \$7,745,000. Expenditures through February 28, 1963 were \$650,000.

The following summarizes project activity in March:

Number of authorized projects at month's end -----	9
Number of new projects authorized -----	1
CAH-995 - Air Conditioning Modifications - 309 Building	
Projects completed -----	0
New projects submitted to the AEC -----	0
Projects awaiting AEC authorization -----	2
CAH-985 - Addition to the 222-U Building	
CAH-986 - 300 Area Retention Waste System Expansion	
Project proposals complete or nearing completion -----	4
Heat Transfer Apparatus for Model Studies	
High Temperature Lattice Testing Reactor	
Plutonium Recycle Critical Facility Conversion to Light Water	
FRTR Storage Basin and Experimental Facilities Modifications	

The current status of projects authorized or awaiting approval is:

CAH-916 - Fuels Recycle Pilot Plant - Design is complete. The bid package for construction was issued March 1, 1963, and bid opening is scheduled for April 10, 1963. Soil bearing tests are currently being performed at the construction site.

CAH-922 - Burst Test Facility for Irradiated Zirconium Tubes - Construction work was started on February 27, 1963. All work to be performed by the CPFF Construction Services Contractor prior to start of work by the Fixed-Price Contractor has been completed. Tie-ins have been made to the stainless steel waste crib line and to a sanitary sewer line. Concrete walls for a loading dock and stairs have been poured. Procurement by GE was started February 27, 1963.

CAH-936 - Coolant Systems Development Laboratory - Construction is complete with minor exceptions. Project will be closed out April 1, 1963.

CAH-958 - Plutonium Fuels Testing and Evaluation Laboratories, 308 Building - All design drawings were submitted to the Commission March 8, 1963. A revised project proposal requesting an extension of directive completion date and authorization of procurement funds has been submitted to the AEC. To date no action has been taken.

CAH-962 - Low Level Radiochemistry Building - The Commission has negotiated a design contract with Leo A. Daly, Architects of Seattle, Washington for design of this facility.

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1103185

CGH-974 - Analog Simulation Facility - The project proposal was returned unapproved by the Commission on March 4, 1963.

CAH-977 - Facilities for Radioactive Inhalation Studies - The design criteria was approved by the Commission on February 25, 1963. No further action has been taken.

CAH-982 - Addition to the Radionuclide Facilities, 141-C Building - The criteria is essentially complete and an estimate prepared based on the criteria.

CAE-985 - Addition to the 222-U Building - The Commission has indicated acceptance of this project. It will be funded from FY 1964 General Plant Project funds unless FY 1963 funds become available later this fiscal year.

CAH-986 - 300 Area Retention Waste System Expansion - The project is awaiting authorization by AEC.

CGH-992 - Additional Fuel Loading Equipment - 308 Building - All design has been completed. The design and design criteria were approved by the AEC with minor exceptions on March 25, 1963. The AEC has requested a check estimate on this project; this estimate is presently being made by CE&UO. Procurement is continuing for this facility. Work on removal of the remaining equipment will resume shortly after April 1.

CAH-995 - Air Conditioning Modifications, 309 Building - A Directive authorizing the project was issued on March 13, 1963. The AEC Work Authority authorizing \$18,000 to the General Electric Company was received March 26, 1963. Requisitions for engineered equipment have been issued.

#### Engineering Services

Engineering service work was provided to research and development personnel as requested. Principal accomplishments were: (1) design of power supply for gas loop in 314 building, (2) design of control circuits for irradiation studies loop, (3) trouble-shooting on 108-F radiation source handling equipment, and (4) continuation of design and supervision of fabrication of C-1 loop.

#### Pressure Systems

Engineering review and inspection of pressure systems continued. The Travelers Insurance inspector witnessed the pressure test and inspected the EDEL-I. Inspection was made of the 308 building autoclaves and recommendations were made. An audit of the piping systems in 325 building was begun.

Plant Engineering

Consulting service was provided to maintenance and operating forces. Other work performed included: (1) completion of emergency power study and load survey, (2) initiation of procedure for complete operating test of 300 Area emergency power system, (3) analysis of 325 building "B" unit heater transformer to correct failure fault and replacement of transformer, (4) electrical design for third floor offices, 328 building, (5) review of bids on retention waste monitoring system, (6) completion and testing of two of three dual filter boxes for 325-A cells, (7) installation of 308 building supply air back draft dampers, and (8) consultation with Effluents Technology personnel on 747 building fume disposal.

Facilities Operation

Costs for February were \$241,741, which is 134% of the forecast for the month. The total cost to date is \$1,290,836, which is 94% of the predicted. During this month (1) improvement maintenance was \$35,870 as compared with \$15,000 forecast, (2) steam cost was \$56,794 as compared to \$40,000 predicted, and (3) engineering cost \$26,400 against an estimated \$14,000. Present expenditure forecasts indicate budgeted funds will be adequate.

Waste disposal operations during February, compared to the previous month, are summarized below:

	<u>January</u>	<u>February</u>
Concrete barrels disposed	8	20
Crib waste gallons	290,000	220,000
Loadluggers of dry waste	29	29

Liquid waste disposed from the 321 building to the 216 BC trench was 3,500 gallons.

Physical testing has been completed on waste trailer No. 5473. No significant wear was noted.

The floor between 308 building's supply fans 3 and 4 has been opened in an attempt to detect a leak in the lines buried underneath the floor.

The #4 process water well pump casing was broken Monday, March 25. The cause was unknown but the parts were fixed and service restored by March 28.

Drafting

The equivalent of 90 drawings were completed during the month for an average of 36 man-hours per drawing.

Major jobs in progress are: (1) PRTR as-builts, (2) PRTR shim rod control, (3) PRTR cladding cutter assembly, (4) PRTR process tube and fuel handling carriage, (5) 108-F inhalation studies hood, (6) equipment for salt-cycle process, 325-A, (7) capillary loading glove box, 308, (8) as-built drawings, PRP critical facility, (9) as-built drawings, control panel critical mass laboratory, (10) vacuum welder, 306, and (11) fuel element molds, 306.

Work performed by CE&UO and Bovay Engineers during the month was 40 man-hours and 100 man-hours respectively. Work assigned was 76 man-hours to Bovay.

#### Construction Supervision

Activity during the month on construction work (J. A. Jones Company) being performed for Hanford Laboratories components is given below:

	<u>Unexpended Balance</u>	<u>Waste Calcination (Job 7005)</u>
Orders outstanding beginning of month	\$149 322	\$314 368
Issued during the month (inc. suppl. and adj.)	123 083	
J. A. Jones expenditures during month (incl. C.O. costs)	121 306	38 968
Balance at month's end	151 099	275 400
Orders closed during month	87 209	

In addition, work on seven maintenance work orders having a total face value of \$15,864, issued to plant forces, was supervised.

Major nonproject jobs in progress are: (1) propane piping replacement, 108-F, (2) installation of plant growth room, 108-F, (3) refurbishing laboratories 402 and 403, 108-F, (4) building modifications, 146-FR, (5) construction of waste trench - 216 BC, (6) installation of intercom system, 231-Z, (7) emergency repairs to meteorology towers, (8) construction of offices, 306, (9) roof replacement, 306, (10) installation of gas loop heater, 309, (11) installation of alarm and annunciator system, 309, (12) construct hatchway in 321 roof, (13) construct room for computer and controls, 321, (14) duct and filter box repair, 325, (15) construct exterior stair, 325, (16) modify fuel inspection development facility, 326, and (17) waste calciner for 324.

Fourteen purchase requisitions totaling \$61,000 were processed during the month. Total value of equipment being purchased is \$100,000.

  
Manager  
Finance and Administration

W Sale:wim

O4 PROGRAM - REACTOR DEVELOPMENTPLUTONIUM RECYCLE PROGRAMPlutonium Recycle Test ReactorOperation

Reactor output for March was 963 MWD for an experimental time efficiency of 63% and a plant efficiency of 57.6%. There were six operating periods during the month, one of which was terminated by a scram as a result of pressurizer high pressure following malfunction of a pressure control valve. Two shutdowns were made because of high D<sub>2</sub>O collection in the recovery system, two were a result of personnel errors, and one was required when both high pressure helium compressors failed to function properly.

The core loading on March 1, consisted of 22 UO<sub>2</sub> elements, 22 Al-Pu elements and 41 mixed-oxide elements. At month-end the core consisted of 19 UO<sub>2</sub> elements, 22 Al-Pu elements and 44 mixed oxide elements. Fuel exposure history at month-end was:

Maximum UO <sub>2</sub> exposure/element	3030 MWD/TU
Average UO <sub>2</sub> exposure/element	2124 MWD/TU
Maximum Al-Pu exposure/element	85.8 MWD
Average Al-Pu exposure/element	69.8 MWD
Maximum Moxtyl exposure/element	49.1 MWD (~982 MWD/TU)
Average Moxtyl exposure/element	32.0 MWD (~640 MWD/TU)

The status of various test elements on March 31, 1963, is shown below. Those test elements which had reached their assigned goal exposure or had been permanently discharged for other reasons prior to March 1, 1963, have been deleted from this table.

PRTR Test No.	Channel Location	Fuel Element Number	Description	Date Initial Charge	Date Dis- charged	Approximate Accumulated MWD
10	Basin	1082	UO <sub>2</sub> -Hot Swage	11/3/61	3/26/63	84.8
10	Basin	1067	UO <sub>2</sub> -Vipac	11/3/63	3/26/63	87.9
13	1853	5094	Al-Pu Physics	12/3/63	--	85.8
14	1956	5097	Moxtyl-Swaged	4/2/62	--	21.8
14	1356	5098	Moxtyl-Vipac	5/8/62	-- Repad	46.8
14	1758	5099	Moxtyl-Vipac	5/8/62	-- Repad	41.2
37	1449	1096	UO <sub>2</sub> Physics	5/12/62	--	53.1
37	1649	1097	UO <sub>2</sub> Physics	5/12/62	--	52.2
37	1552	1098	UO <sub>2</sub> Physics	5/12/62	--	48.8
37	1548	1099	UO <sub>2</sub> Physics	5/12/62	--	50.6
37	1651	1100	UO <sub>2</sub> Physics	5/12/62	--	43.7

PRTR Test No.	Channel Location	Fuel Element Number	Description	Date Initial Charge	Date Dis- charged	Approximate Accumulated MWD
48	1243	5150	Moxtyl ( $\frac{1}{2} \times \frac{1}{2}$ " pads)	8/1/62	--	32.8
54	1948	5116	Moxtyl (clip-on pads)	5/8/62	-- 33.6	(27.5 w/clip)
54	1443	5117	Moxtyl (clip-on pads)	5/8/62	-- 49.1	(34.1 w/clip)
54	1459	5118	Moxtyl (clip-on pads)	5/8/62	-- 47.5	(32.5 w/clip)

A total of 5.9 kilograms of plutonium were recovered at the Redox plant, from 32 irradiated Al-Pu fuel elements. Composition of the final Redox product was as follows:

Pu-239	74.678
Pu-240	21.468
Pu-241	3.418
Pu-242	0.440

D<sub>2</sub>O and Helium losses for the month were 1,175 pounds and 112,642 scf, respectively.

#### Equipment Experience

A total of 10<sup>4</sup> outage hours were charged to repair work. The majority, 62 hours, was required for high pressure helium compressor repairs, 16 hours were required for gasket replacement, and 18 hours were required for helium valve repairs to eliminate in-line leaks. Compressor difficulties were traced to severe wearing of piston rings. Four tube-to-nozzle and 7 jumper-to-tube gaskets were replaced. Air inleakage resulting in high O<sub>2</sub> content in the systems was traced to the shaft of the core blanket blower and inlet fittings of one low pressure helium compressor. Corrective work and elimination of O<sub>2</sub> required 14 chargeable hours of outage time.

Repair of one irradiated shim rod assembly (originally considered impractical) was completed in the fuel element examination facility and repair of another was partially completed.

Preventive maintenance required 273 manhours or 5.6% of the total maintenance effort.

#### Improvement Work Status (Significant Items)

##### Work Completed:

Fuel transfer hoist replacement  
Core blanket blower seal modification

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Work Partially Completed:

Enlarge Chemical Feed System  
Reactor Core Level Indicator  
Primary Loop Drain and Flush Valve Modifications  
Interlock Between C-D Machine Shroud Seat and Discharge Hoist  
Shim Rod Connector Modifications  
Recording Ammeter for Primary Pumps  
Control Room Criticality Alarm  
Shim Well Rotameter Modification

Design Work Completed:

Pressurizer Vent Valve Relocation to Minimize D<sub>2</sub>O Loss  
Compressed Air Supply Modifications  
384 Emergency Power to Primary Pumps  
Low Pressure Light Water Injection Control Valve  
Actuator with Hydraulic Snubber for Bottom Blowdown Line  
Flash Tank Modification

Design Work Partially Completed:

Additional Fuel Storage and Examination  
Install Vibration Snubbers for Earthquake Protection  
Effluent Activity Monitor Replacement

Process Engineering and Reactor Physics

PRTR Test 61 was prepared to irradiate six mixed-oxide elements which have cobalt-zirconium wire wraps and which have three rods in each element containing highly homogenized sintered pellets of UO<sub>2</sub>-PuO<sub>2</sub>, for physics burnup studies.

Vibration data were obtained at the outlet nozzle of a few selected process tubes as the primary pump combinations were changed. The #2 and 3 pump combinations indicated a pulsing effect which occurred at approximately 5-second intervals. Pump combinations 1 and 2, and 1 and 3 appeared to be stable.

Additional analyses of functional testing of emergency backup systems were completed. A report has been issued.

A study of fuel element "allowable time without cooling" with respect to the maximum jacket temperature which will still permit reuse of the fuel element was completed. A draft of the report has been prepared.

Additional analyses on the light water injection system were completed.

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Procedures

Revised Operating Procedures Issued		6
Revised Operating Standards Issued		11
Temporary Deviations to Operating Standards Issued		5
Revised Process Specifications accepted for use		1
Maintenance Manuals and Procedures Issued		0

Drawing As-Built Status:	<u>March</u>	<u>Total</u>
Approved for as-built	27	937
Ready for approval		6
In Drafting		27
Voided		79
		<u>1 049</u>
Scheduled for review		406 (34 dwgs added)
		<u>1 455</u>

Personnel Training		
Qualification Subjects		322 manhours
Specifications, Standards, Procedures		26
Emergency Procedures		10
Maintenance Procedures		123
		<u>481</u>

Status of Qualified Personnel at Month-End		
Qualified Reactor Engineers		8
Qualified Technicians		6
Qualified Technologists		16
Provisionally Qualified Technologists		1

Plutonium Recycle Critical Facility

Nineteen uninterrupted days of beneficial use were realized. The initial loading was established. Power calibrations, rod calibrations, and moderator level sensitivity measurements were performed as planned. Expected experimental results were accomplished. The initial criticality milestone occurred on March 21. As an indicator of experimental quantity, the 19 days of experimental time yielded 22 period measurements, 5 flux traverses, 2 foil irradiations, and 127 sets of multiplication data (each set comprising 3 - 5 count rate records for up to 8 neutron counters).

Biological shielding integrity has been checked and found to be adequate for present operating conditions with "cold" fuel.

Design was completed for a moderator heater. The safety rod "In" limit light circuit was converted to 24 VAC. The scope of requirements for conversion to H<sub>2</sub>O moderation was completed.

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Fuel Element Rupture Test Facility (Project CAH-867)

Project Status

Mechanical design testing was 90% complete. A 24-hour test at 2000 psig and 600°F was completed on 3-24-63. Work status of major elements of work follows:

- Sampling system - 60% complete.
- Leakage collection system - 10% complete.
- B Cell test section installation - 30% complete (fabrication - complete)
- Shielding installation - 85% complete.
- Revised relief systems - 95% complete.
- Emergency depressurizing valve - complete.
- Inlet piping shutoff valve - complete.

Operation

The operating group participated in design testing. One hundred and sixty manhours were devoted to training.

GAS COOLED POWER REACTOR PROGRAM

Pressurized Gas-Cooled Loop Facility (Project CAH-822)

Project Status

Project status remains at 94% complete. Blower delivery was again delayed due to a second thrust plate failure. The vendor was advised to ship these blowers by mid-April.

Helium shroud coolant system tests were conducted with some difficulties but satisfactorily dependent upon final analysis.

Operation

The operating group participated in design testing. One hundred and twenty manhours were devoted to training.

TECHNICAL SHOPS OPERATION

Total productive time for the period was 23,194 hours. This includes 15,557 hours performed in Technical Shops, 4,809 hours assigned to Minor Construction, 3,702 hours assigned to off-site vendors, and 126 hours to other project shops. Total shop backlog is 28,679 hours, of which 70% is required in the current month with the remainder distributed over a three-month period. Overtime hours worked during the month was 6.2% (1,160) of the total available hours.

Distribution of time was as follows:

	<u>Man-Hours</u>	<u>% of Total</u>
N-Reactor Department	3,845	16.58
Irradiation Processing Department	3,964	17.09
Chemical Processing Department	617	2.66
Hanford Laboratories	14,768	63.67
Construction Engineering and Utilities	0	0

Requests for emergency service decreased slightly requiring an overtime ratio of 6.2% versus 8.1% for the previous month.

#### LABORATORY MAINTENANCE OPERATION

Manpower utilization for March is summarized as follows: Total productive time realized was 18,100 hours of a possible 19,100 hours potentially available. Of the total productive time realized, 87% was expended in support of Hanford Laboratories components, with the remaining 13% directed toward providing service for other HAPO organizations. Overtime worked during the month was 2.6% of total available hours.

A. Shop Work		3 200 hours
B. Maintenance		7 600 hours
1. Preventive Maintenance	1 900 hours	
2. Emergency or Unscheduled Maintenance	1 900 hours	
3. Normal scheduled maintenance	3 800 hours	
4. Overtime	500 hours	
C. R&D Assistance		7 300 hours

*WD Richmond*  
 Manager  
 Test Reactor and Auxiliaries

WD Richmond:bk

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INVENTIONS OR DISCOVERIES

All persons engaged in work that might reasonably be expected to result in inventions or discoveries advise that, to the best of their knowledge and belief, no inventions or discoveries were made in the course of their work during the period covered by this report except as listed below. Such persons further advise that, for the period therein covered by this report, notebook records, if any, kept in the course of their work have been examined for possible inventions or discoveries.

<u>INVENTOR</u>	<u>TITLE OF INVENTION OR DISCOVERY</u>
G. F. Garlick	HWIR-1611, A Technique for Measurement of Neutron Dose
E. M. Sheen	HWIR-1612, A Novel Count-Rate Circuit
W. W. Schulz	A Process for Dissolving Irradiated Pu-Al Alloy Fuel
G. E. Benedict and L. K. Mudge	Preparation of Certain Metal Oxides and Mixed Oxides by Electrodeposition from Molten Chloride Salt Solutions (HW-76828)
G. E. Benedict	Precipitation of $UO_2$ - $PuO_2$ and $UO_2$ - $ThO_2$ Solid Solutions by Hydrolysis of Their Chlorides in Molten Chloride Salt Solutions (HW-76827)
F. A. Scott and H. J. Anderson	Method of Preparing UOS and Other Uranium Sulfides (HW-76963)
Walter B. Jackson	HWIR-1602, Controlled Rotation Cylinder

  
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Manager, Hanford Laboratories

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