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RICHLAND, WASHINGTON

HANFORD ATOMIC PRODUCTS OPERATION



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FEBRUARY 14, 1964

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HANFORD LABORATORIES

MONTHLY ACTIVITIES

REPORT

JANUARY, 1964

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This document consists of 214 pages.

HANFORD LABORATORIES MONTHLY ACTIVITIES REPORT JANUARY 1964

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Compiled by Section Managers

February 14, 1964

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HANFORD ATOMIC PRODUCTS OPERATION RICHLAND, WASHINGTON

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 I - Hanford Laboratories Force Report

	Date: January 31, 1964					
	At Beginni Exempt	ing of Month Salaried	At Close Exempt	e of Month Salaried	Total	LCCATT
Chemical Laboratory	146	133	147	130	277	H L H
Reactor & Fuels Laboratory	204	198	207	197	404	
Physics & Instruments Laboratory	102	76	102	75	177	
Biology Laboratory	43	64	44	66	110	
Applied Mathematics Operation	19	5	21	5	26	
Radiation Protection Operation	42	99	41	95	136	ΔT
Finance & Administration Operation	151	119	149	118	267	
Programming Operation	17	4	18	4	22	
Test Reactor & Auxiliaries Operation	64	317	65	315	380	
General	<u> </u>	4	<u> </u>	5	9	
TOTAL	792	1019	<u> 798 </u>	1010	1808	

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BUDGET AND COST SUMMARY

January operating costs totaled \$2,893,000, an increase of \$327,000 over the previous month; fiscal year to date costs are \$19,027,000 or 58% of the \$32,597,000 control budget. Hanford Laboratories' research and development costs for January compared with last month and Financial Plan No. 4, dated January 22, 1964, are shown below:

		COST			
(Dollars in thousands)	Current Month	Previous Month	<u>To Date</u>	Budget	% Spent
HL Programs					
02	\$ 98	\$67	\$ 559	\$ 1 180	47
03	8	12	231	250	92
04	1 331	1 096	8 383	13 754	61
05	109	105	833	1 403	59
06	275	258	1 897	3 352	57
08	28	11	99	240	41
	1 849	1 549	12 002	20 179	59
Sponsored by					
NRD	175	168	1 149	1 798	64
IPD	31	29	354	490	72
CPD	166	147	904	1 668	54
Total	\$2 221	\$1 893	\$14 409	\$24 135	60%

RESEARCH AND DEVELOPMENT

1. Reactor and Fuels

In support of radiation hazard studies, inner component specimens from N-Reactor fuel have been pulse annealed at 980 C. Fuel volume increases of 19-38% were observed, but there was no evidence of clad necking and failure and, thus, no evidence that activity release or massive corrosion would result should these fuel temperatures be encountered for short periods.

Initial destructive examination of a prototypic inner component of N-Reactor fuel irradiated to 2500 Mwd/ton indicates excellent component performance.

A capsule irradiation test comparing irradiation performance of uranium containing submicron particles of uranium carbide standard ingot uranium has achieved an exposure of 0.05 at% burnup and will be irradiated to 0.3 at% burnup.

Eight Li-Al target elements have been discharged after 124 days at full power. The elements appear unchanged as a result of irradiation.

A full KER loading of target components has been fabricated. The target material contains 0.56% Li enriched to 63% Li⁶.

Postirradiation examination of Zircaloy-2 clad uranium rods containing known cladding defects have shown strain concentration at the defect varying from 15% to 40% to complete cladding failure. Results from examination of these rods should complete the picture of cladding instability in progressive stages from first indications to ultimate rupture.

Two inner tubes from N-Reactor fuel elements with fluted outer surfaces have accumulated 1800 Mwd/ton exposure and continue to show excellent visual appearance.

In the study of effects of altered fuel composition on irradiation behavior, single tube fuel elements containing uranium to which 400 ppm iron and 800 ppm aluminum have been added have been completed. These elements will be irradiated in a comparison test with standard N-Reactor fuel compositions.

A second generation N-Reactor fuel support has been fabricated by a single step hot forging of Zircaloy wire. This support has been designed to meet a much higher strength requirement than the first generation support.

In an IPD production test, hot-die sized fuel elements increased in outer and inner diameter more rapidly than did the Al-Si controls. Destructive examination has shown no evidence of grain boundary tearing. Steps in the canning process which might cause texture changes sufficient to account for this behavior are under study.





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The corrosion of aluminum coupons in heated process water was reduced by factors of 5 to 10 by coating the surface with either of two commercial marking inks.

Using NH₄OH to adjust the pH to 10 water quality continues to be satisfactory in the operation of the KER Loops. The radiolysis of ammonia during reactor operation is greater at low than at high coolant temperatures. Change of KER-2 from LiOH to NH₄OH resulted in a reduction in O_2 , demonstrating the effectiveness of radiolytic hydrazine in scavenging oxygen.

Samples of intergranularly attacked tubing from N-Reactor steam generator 4A with pinhole defects were tested at 520 and 550 F and pressures of 1720 and 1340 psig, respectively. Two of the samples split at the pinhole defect within a few hours. However, this failure is thought to be a result of the prior application of 3000 psi pressure to produce the defects. Eight decontamination cycles of this tubing with sulfamic acid widened the attack at the grain boundaries in the areas which had been intergranularly attacked, and completely removed the grains.

Refined experiments related to radiation-induced reaction of carbon monoxide with water vapor indicate no significant thermal reaction below 600 C.

The oxidation of TSX graphite in carbon dioxide and carbon monoxide mixtures was measured and found greater than in corresponding reactions of CSF graphite.

The rejuvenation fuel element successfully completed its third cycle of irradiation.

The new Vipac facility was used to fabricate f $\cdot e$ fuel elements for the PRTR. One of these uses a new end cap design to eliminate the crevice of the old design. Three other elements were made by the swage compaction technique.

New tubing for swage compacted elements allows an increase in reactivity by using more fuel per unit length.





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Tubular extensions of aluminum alloys containing 6 and 10% silicon have been successfully fabricated for evaluation as cladding components.

A pyrometallurgical method of decontaminating U^{233} is being evaluated using natural uranium which had been stored for a period of a year. Liquidation of the metal in Zirconia crucibles followed by casting into a graphite mold reduced the alpha activity of the cast uranium 98.3%.

Three massive tubular Zircaloy-2 and Th-U fuel elements have achieved an integration exposure of 1.5×10^{20} fissions/ac (4200 Mwd/ton), the elements continuing under irradiation.

The effect of specific fuel composition on the irradiation and corrosion behavior of Th-U alloys is under study. Nine additional alloys have been double vacuum arc-melted. The resulting ingots will be coextruded to Zircaloy-2 clad rods by defect corrosion testing.

Preliminary efforts to investigate the validity of existing two-phase steam-water critical discharge theories at high pressures have produced unexpected results. Laboratory experiments, performed on a short piece of pipe (L:D = 20) with upstream pressures approaching 2000 psia, showed correspondence with the more advanced theories only when two-phase mixtures entered the pipe. When compressed water entered the pipe, the location of the primary choking occurred at the entrance rather than the discharge of the pipe section, and very high flow rates resulted.

Temperature behavior was approximately as expected for an N-Reactor fuel surface changing from film to nucleate boiling, but that resistance to flow was less than expected. This information is of importance in analyses of loss -of-coolant incidents in N-Reactor.

A formal report giving two new cross section libraries for MAC shielding code was completed and submitted for publication.

A defected N-Reactor fuel element with a bonded tapered end cap, irradiated to 1200 Mwd/ton, was ruptured in IRP at 300 C. Rupture started after 162 min of incubation; the end cap came completely loose.



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Information about defect propagation and contamination of coolant was obtained during 406 hr of irradiation of a defected Al-Pu, PRTR spike element in the FERTF.

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Anticipated cladding failures of PRTR fuel elements containing fuel suspected of being contaminated with oil supplied strong evidence that such contamination was the cause of other recent failures.

Irradiation testing of prototypic fuel capsules and fuel rods progressed through charging of several test capsules into MTR-ETR, and by hydraulic analyses of a fuel element design for irradiating EBWR fuel rods in PRTR.

An eddy current detector fuel element was completed for use in EDEL-1 facility evaluation of fretting corrosion.

Evaluation of older stocks of PRTR (0.505 in. ID) Zircaloy-2 cladding by improved methods indicates a 20-25% rejection rate.

The south vibrational compaction unit in the 308 Building was adapted to PRTR fuel element fabrication.

Calculations were performed to determine heat transfer conditions for higher power densities in the PRTR. The five different instances of power density increases which were studied involved decreasing the number of fuel channels and/or decreasing the fuel length. It was found that by decreasing the number of tubes from 85 to 43, shortening the fuel from 88 to 48 in., and providing adequate subcooling, the maximum heat flux could be increased from 221,000 to 801,000 Btu/hr-sq ft without relaxation of the reactor's boiling burnout safety margin.

Zirconium concentrations in the PRTR primary coolant were significantly above normal for 3 days during the past month. Some fretting is possible, severe fretting is unlikely.

The laboratory for plutonium decontamination studies was completed, and tests of plutonium oxide dissolution were started.

Three irradiation test capsules containing solid solution UN-20 wt%PuN pellets were prepared for irradiation in the MTR.

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ThN was synthesized for use in preparation of (Th-Pu)N compounds for phase and irradiation studies.

Four thorium metal-plutonium oxide cermets were fabricated by the Nupac process.

Using uranium as a stand-in for plutonium, mockup tests were made of metallography procedures with the recently installed research metallograph.

A deliberately defected, vibrationally compacted thoria element was irradiated in the MTR GEH-4 loop and coolant activity level exceeded operating safety limits. The element is being returned to Hanford to evaluate extent of fuel washout.

Single crystal UO_2 containing metallic uranium has been examined by optical transmission microscopy to 700 C.

Uranium monosulfide has been irradiated for 154.5 effective full power days to 3.5 x 10^{20} fissions/cm³.

Preliminary results indicate plutonium and some fission products relocate in UO_2 under an applied potential gradient.

Brittle fracture tests have been continued at room temperature on a series of N-R actor process tube specimens containing 275 ppm, 180 ppm, and 90 ppm hydrogen. The reduction in burst pressure due to the presence of the hydride was 14.6% at 90 ppm, 18.4% at 180 ppm, and 29.2% at 275 ppm.

Progress on research and development in support of the ATR gas loop was reviewed with representatives from Babcock & Wilcox Company, Ebasco Services, Phillips Petroleum Company, and the Atomic Energy Commission.

Comparisons of the corrosion resistance of weldments of Haynes 25, Hastelloy X to themselves and each other in 1200 C air contaminated helium





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for 7 days indicate severe oxidation to complete destruction of the Haynes 25 alone or coupled. The Hastelloy X retains its integrity and a tenacious oxide film.

The tube failure in the PRTR rupture monitor heat exchanger resulted from wear against a support plate. Pits up to 20 mils in depth, of unexplained origin, were observed on the ID of the tube.

All of the equipment for the Project CAH-922 Irradiated Burst Test Facility is on hand. The major portion of the work remaining to be done involves mechanical and electrical connection of the instrument panels.

Tensile tests have been performed on irradiated stainless steel specimens at 300 C (572 F), the temperature at which they were irradiated in the ETR G-7 hot water loop. Ductility at 300 C was found to be 50% less that the room temperature value.

At the present time four creep capsules have been assembled and are ready for charging into the reactor. Three of these capsules contain AISI 304 SS specimens. The fourth contains a final 20% cold worked Zircaloy-2 specimen.

A temperature effect was observed in the tensile properties of Inconel 625 and Hastelloy N irradiated at 50 \odot (122 F) and 280 C (536 F) to an exposure of 1 x 10²⁰ nvt. Specimens of both alloys tested at room temperature showed that the increase in yield strength and decrease in total elongation were substantially greater in the case of 50 C irradiations.

Samples (AIS) 348 SS) exposed 19 days in 280 C water of the ETR G-7 loop have been stripped of oxide and metal losses determined. Corrosion of the irradiated samples was somewhat less than that of unirradiated controls.

Rates of hydrogen absorption and desorption at 750 C (1562 F) in the Zr-H system have been measured over the range $ZrH_{0.2}$ to $ZrH_{1.18}$ in a closed system. The time required for 90% saturation varied from 3 to 30 min, the longer times apparently the result of oxygen contamination. The time required for 90% desorption varied from 2 to 9 min.

Initial results of the quantitative metallographic study of second phase particles in dilute alloys of uranium have been obtained. Data on 9 of the 30 specimens representing different compositions and heat treatments indicate that the population density and average particle size of the second phase are significantly affected by both composition and heat treatment.

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Two swelling capsules which were previously brought to operating conditions at 625 C (1157 F) and 425 C (797 F), respectively, are operating satisfactorily.

Studies of quench-hardening in nickel have shown that the amount of hardening achieved increases with decreasing impurity content. This is direct evidence that impurity atoms are effective trapping sites for vacancies. Aging at a temperature of 500 K (437 F) results in a significant increase in the yield stress.

Strain-rate cycling experiments on unirradiated polycrystalline molybdenum have shown that the effective activation volume is approximately 10^{-19} cm³ at room temperature. This unexpectedly high value has been tentatively interpreted in terms of a mechanism involving cross-slip of screw dislocations.

High-purity, electrorefined plutonium has been received from the Los Alamos Scientific Laboratory. It is currently being used for basic research studies and its properties are being compared with those of selected high-quality, as-reduced, and vacuum-cast metal. Metallographic observation revealed that both types of metal have very few inclusions and approximately the same amount and distribution of microcracks. The beta-toalpha reaction rates of both types of metal are comparable.

The steady state strain rate of plutonium in the beta and delta phases has been found to be proportional to the fifth power of the applied compressive stress over a wide range of stresses. For plutonium in the gamma phase, this proportionality is valid over only a limited stress range.





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Detectors to measure the energy distribution of neutron, have been calibrated and are currently being used to monitor the neutron experiment in the N-Reactor core during startup tests.

Neutron flux monitoring tests indicated the degree of consistency to be expected in such measurements is probably about 15%.

Irradiation experiments with systems involving carbon, carbon monoxide, and carbon dioxide showed (1) CO disproportionated to yield carbon dioxide and black solids, (2) graphite and carbon dioxide yielded carbon monoxide, the ratio CO_2 :CO being 0.86, and (3) thermal reaction between carbon dioxide and graphite was negligible below 675 C; reaction was rapid in the range 1200 to 1400 C, and the CO_2 :CO ratio was 0.5.

Further work on the oxidation of TSX graphite by water vapor indicated a higher activation energy for large samples than was previously reported. This is in better accord with earlier experimental data and theoretical projections.

The long term irradiation of boronated graphite is proceeding satisfactorily. The high and low temperature sections are operating respectively at about 900 F and 725 F.

Photomicrographs of gray boronated graphite containing 7% boron indicate almost complete recrystallization of the structure. It is expected that this material will have quite different response to radiation than the black boronated graphite which resembles polycrystalline graphite.

Additional studies on the Military Compact Reactor (MCR) were completed, and an informal document has been prepared summarizing the findings of the study.

The second generation shim rod assembly has been successfully tested in the environmental mockup.

The wedged-type PRTR fuel element designed to prevent fretting corrosion at the end brackets was tested at PRTR operating conditions in EDEL-I for 3 weeks. Tube examination showed fretting of 1/2-mil depth at the two supports opposite the wedges at the top and bottom of the fuel element. However, the carbon steel spring used to maintain tension on the wedges had relaxed from high temperature, so that the fuel element was probably not "locked" during testing as intended.

The "excessive vibration" detector on the PRTR operated satisfactorily for 2 weeks. Increased vibration levels in PRTR were indicated during scrams, power level changes, and changes in pump combinations.

The HTLTR heating element test mockup was completed and considerable water was removed during heating of the insulating material. A heating element surface temperature of 1020 C was achieved. The heating element subsequently failed, presumably due to oxidation of the graphite heater.

More than 400 EBWR rods have been fabricated by the Vipac process.

A number of improvements were made in the process for pneumatic impaction of UO_2 -PuO₂ fuel material to provide consistently high density and reproducible vibrational compaction characteristics.

A relationship between tube volume and average diameter was found to exist for EBWR tubes and variations in tube volume are believed not likely to cause difficulties in the vibratory compaction process.

EBWR fuel element fabrication histories were better for the three of six examined lots that showed better particle bonding, highest densities, and O:U ratios closest to the value of the starting material.

Preliminary data obtained from recent sieve analyses indicate surprisingly little breakup of the particles within a lot of the formulated mesh composition, even after 40 min of vibrational compaction.





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Developments in the process for pneumatic impaction of UO_2 -PuO₂ fuel material provided improved control of fuel particle density. It was established that AISI 310 SS can be used in inside impaction containers without appreciably altering UO_2 stoichiometry during heating, but that UO_2 heated in AISI 304 SS cans is subjected to reduction in O:U ratio, because of oxygen gettering by the can. An O:U ratio of approximately 2.01 or more is required for maximum density and adequate particle bonding. Since UO_2 heated in AISI 304 SS cans might be reduced below an O.U ratio of 2.01, the use of this material was discontinued. It was also found that material near the center of the impaction cans was not uniformly dense. Heating time was increased from 55 min to 80 min to correct this.

In the study of tungsten clad cermet fuels, a method of forming fuel plates confined in a cylindrical tube has been demonstrated. A tube containing seven 0.045 in. thick tungsten plates was fabricated by coextruding a W-Mo assembly with subsequent chemical removal of the molybdenum.

A W-UO₂ cermet full material containing uniformly dispersed submicron UO₂ particles is under development. The coprecipitation of uranium and tungsten has been demonstrated, although optimum precipitation conditions have not yet been established. Reasting and sintering conditions required to reduce the precipitation to W-UO₂ and to densify the resulting material without serious agglomeration of the UO₂ particles are under investigation.

Postirradiation examination of a second NASA $W-UO_2$ fuel specimen was completed. Successful operation at approximately 3000 C was inferred from tungsten grain structure.

Nonfueled grids are being assembled for fuel element design studies.

Photomicrographs of sections of $W-UO_2$ impacted cermet platelets revealed good bonding between the tungsten powder of the cermet and the tungsten foil cladding.



An intensive investigation of the accelerated fuel loss resulting from thermal cycling has led to significant improvement in the fuel loss characteristics of W-UO₂ cermet fuel plates.

A hexagonal honeycomb grid with 1/8 in. hexagonal coolant channels was fabricated by pneumatic impaction. The 80 vol% W-UO₂ grid was initially loaded by vibratory compaction and the mild steel mandrels were removed solely by chemical techniques.

Spalling of $W-UO_2$ cermets in a plasma is being investigated.

Two $W-UO_2$ cermet grids have been vibrated to failure at room temperature and indicate the possibility of microcrack formation in the tungsten matrix during vibration.

Bonding was achieved on the longitudinal joint of a 1 in. wide section of a 2 in. OD x 0.020 in. wall tungsten cylinder formed by shaping of rolled plate.

Approximately 1 ton (632 fuel elements) of low uniform bulk density $(65.5-67.3\% \text{ TD})^{-6}$ Sol-Gel¹⁰ thoria was canned Experimental work to achieve a high density thoria mixture that utilizes a maximum percentage of the material as received from offsite produced encouraging initial results. A density of 82.5% was obtained using a low power vibrating table.

2. Physics and Instruments

The cold reactor physics tests for N-Reactor startup were completed and the data are now being reduced to physics parameters of interest. Preparations for the hot reactor tests are nearly complete.

In other N-Reactor supports, assistance was provided in checkout of the fuel rupture monitor systems. Rupture detection instrumentation for the PRTR Fuel Rupture Loop operated satisfactorily during the first defected fuel test; and preparations were made for a combined simulation of the primary loop and steam generator systems on the new analog computer.

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Emergency testing service was provided N-Reactor Department to locate the position and determine the extent of damaged Graphite Heat Exchanger tubes. Breaks in the tubing wall were successfully located; surprisingly, the defects were in each case located in the same relative position.

Seven more critical mass experiments were conducted with PuO_2 -polystyrene compacts in rectangular prism configurations. Plutonium concentration was 1.12 g/cc (2.2% Pu^{240}), with a H:Pu atomic ratio of ~15. Various reflector combinations involving Lucite were used to determine the effect of cadmium on reflector savings. Additional data were obtained with complex reflectors for evaluating the effect of neutron poison sheets or absorbing materials interposed between the reactor core and its reflector. Absorbing materials consisted of steel plate in several thicknesses, one of which contained ~0.3 wt% gadolinium.

Several major revisions and improvements were made in the GAMTEC Code (a neutron slowing down and thermalization code which provides multigroup constants for heterogeneous systems for use in multigroup diffusion calculations). The changes reduce the amount of input data and improve the theoretical accuracy of the code.

SMC, a Monte Carlo code, was written for studying neutron slowing down and diffusion in multiregion finite assemblies. SMC will be used to compute criticality in spherical geometries containing one-to-three material regions.

A study was made of a possible method for determining the neutron spectrum which in theory appears promising. The method consists in making comparisons between calculated and measured activity ratios for various foils whose energy dependent cross sections are known.

Work has proceeded on the design of an on-line reactor noise analysis system for the Critical Mass Laboratory.



Nuclear safety consulting services were provided to HL, CPD, and NRD. The Course B, Group II lecture series in nuclear safety (eight sessions) for CPD was completed January 23. The next session, Group III, will begin in February.

Modifications to the triple-axis spectrometer at 105-KE have been substantially completed. Modifications will allow the use of the adjacent beam hole for time-of-flight studies of slow-neutron scattering.

The complete reduction of data on the inelastic scattering of neutrons from 95 C H_2O reveals internal inconsistencies which will require further measurements.

The abundance sensitivity of a heavy-element mass spectrometer has been restored to its previous value by modifying the entrance port of the newly installed ion detector.

Phoenix fuel studies have been extended to beryllium-moderated systems. Beryllium provides an energy flux distribution more conducive to epithermal capture in Pu^{240} than does water and, therefore, increases the relative capture in that isotope. This increased capture could improve the Phoenix action in some reactors. Other Phoenix investigations indicate that the results of analysis are sensitive to the burnup-time steps used in the analysis. Planning of Phoenix fuel type critical experiments continues.

Approach-to-critical experiments were initiated during the month using EBWR PuO_2-UO_2 fuel rods. The first lattice being studied is equivalent to the EBWR fuel-to-moderator ratio, and has a lattice spacing of 0.71 inch. Approximately 400 of the estimated 500 rods needed for criticality have been loaded. The experiment will be completed as soon as sufficient fuel is available.

The conversion of the PRCF for light water experiments is essentially complete. The EBWR fuel rods will be loaded in that facility as soon as the





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first approach-to-critical experiment is completed. A report, HW-80092, has been issued describing the planned light water experiments in the PRCF.

Analysis of data taken on D_2O -moderated experiments in the PRCF continues. Moderator level coefficients and the ratio of delayed neutron yield to the neutron lifetime were determined during the month.

Neutron flux distributions produced by the modified heavy gas equation for homogeneous light water-moderated systems were compared to those produced by a more detailed thermalization model. The modified gas equation adequately describes the flux distribution of water-uranium and water-plutonium systems encountered in thermal reactors. Previous studies gave similar results for graphite systems.

In computer code development, analysis of the PRTR 19-rod cluster is being performed in final checkout of the RBU code. The current version of GE-HL Program S-XII, which has major improvements over previous versions, has been sent to Argonne National Laboratory Code Center for industry-wide distribution. The HRG code has been modified so that it provides punch cards directly in the Program S format. Improvements have been made in the Physics Chain that will allow multiple passes through GAM and TEMPEST. The ZODIAC burnup code has been expanded from 10 to 18 energy groups.

Extensive analysis of the three sets of Pu-Al, light water-moderated critical experiments continues. These have been analyzed using the GAM, HFN, and THERMOS codes. Thermal parameters using the latter code were determined employing the Brown-St. John and Nelkin scattering models. Neither model yielded consistently good agreement with experiments. Interestingly, the difficulties apparently lie in the spatial flux distribution in the moderator and not in the energy-space distribution in the fuel as one might expect. The poorest results were obtained using the

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Nelkin model in analyzing the 5 wt% experiments. The Nelkin kernel also gave poorer agreement when used in analyzing the UO_2 Yankee criticals. These studies will continue.

Destructive analyses have been completed on one of three H_x Pu-Al fuel elements irradiated in the PRTR. Assistance provided by Chemical Research Operation in the analysis procedures has improved the quality of data obtained. The PRTR Gamma Scan Facility has been redesigned to improve operation. Irradiated fuel rods are scanned in this facility for determining fission distributions. The ALTHAEA one-dimensional burnup code has been used successfully in analyzing the L_x Pu-Al burnup experiments performed in the PRTR.

An HTLTR graphite heater element was tested up to temperatures of 960 C for 24 hr, but failed when exposed to air that leaked in around a sight tube. A new heating element has been constructed and installed in the preliminary reactor mockup assembly for further tests. The construction of the outer shell of the larger mockup has been completed, but work has been stopped because of lack of funds. High temperature tests on samples of various materials in nitrogen continued during the month.

Four uranium samples irradiated at KE-Reactor under the Neutron Flux Monitor program have been discharged and analyzed by mass spectrometer. If these data confirm theoretical predictions, as expected, this phase of the program will be concluded. Laboratory efforts have suggested methods of circumventing difficulties encountered with the B-11 detectors, and these have been incorporated in new units now being fabricated. A production test has been approved for testing of the microwave system at KW-Reactor, test loops and plasma capsules are being made ready for these tests.

A technique which promises to be a major advancement in practical use of the eddy current multiparameter tester is undergoing laboratory checkout. In essence, the technique significantly simplifies calibration procedures by locating all adjustments associated with a particular test





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parameter at a single control. By preliminary estimates, this procedure will reduce average setup and calibration time from several hours to a few minutes.

Infrared studies for reducing the effect of surface emissivity variations have reached a milestone; the experimental demonstration of the delayed signal concept. Applied to l&E fuel elements having clean or dirty and bumpy surfaces, the test proved to be markedly less sensitive to surface condition. These experiments also revealed an interesting and significant effect in that a fractured bond, apparently caused when the fuel surface was purposely dented, increased in size with each successive heating of the aluminum jacket.

Successful laboratory tests have been performed on sensors and associated electronic circuitry that are to be used for the remote measurement of animal temperature, pulse rate, and respiration rate. The implanted sensors will telemeter the data to a remote station.

This year, a field party completed the first of a number of planned trips to Anaktuvuk Pass, Alaska to measure the Cs^{137} body burdens of Eskimos with the shadow-shield whole body counter.

Other radiological physics work included: comparison of the Hanford precision long counter response with S(n, p) reaction data to establish relationships with S(n, p) reaction calibrations by other U. S. laboratories; progress on reduction of the background for the plutonium counter; and development of a new neutron spectrometer and a new radiation calorimeter.

The mathematical model for the lateral growth of a diffusing plume of material dispersed to the atmosphere that was derived from Hanford ground source data was shown to also apply for some elevated source releases. Independent verification of the model was made using data from 16 releases at 200 ft height made during 1955-1956. The wind variability factor was determined from wind velocity measurements at the release height. Agreement between the new model and the data was far superior to the power function used earlier.



3. Chemistry

Gas and liquid chromatographic studies were performed on a sample of used Purex solvent which exhibited unusually high ruthenium retention. No evidence of a volatile ruthenium species was revealed by the gas chromatographic work. Liquid chromatography, on a silica gel column prepared with Soltrol, was much less effective in removing fission product complexing agents from the TBP-Soltrol solvent than it has been found to be with Soltrol itself.

Laboratory testing of the trilauryl amine extraction of neptunium and plutonium from actual Purex 1WW was successfully performed.

Initial studies of the Redox prototype countercurrent anion exchange column have demonstrated satisfactory performance using thorium as a stand-in for plutonium.

The control loops associated with the Gradient Control System and pulse control system for the Plutonium Reclamation Facility were given a final and satisfactory calibration check preparatory to plant delivery.

A plutonium detection test loop has been operated with a wide range of plutonium concentrations using two in-line detectors, a 17 kev X-ray crystal and an alpha-sensitive glass scintillator. The X-ray device proved best.

The first of a planned series of exploratory hot cell studies of the dissolution and processing of irradiated thorium was completed without incident.

Studies are continuing on preparation of target grade thoria by a modified Sol Gel process. In addition, a brief study was made of radiant heat spray calcination for this purpose. Thoria of high purity and low moisture and nitrate content was readily produced by spray calcination. The tap density was low (3 g/ce) but the powder was readily pressed to 65-70% of theoretical density.



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Batch contact studies have established suitable extraction conditions for adequate extraction of uranium and thorium recovery from scrap containing plutonium, uranium, and thorium.

The containment controls required for bismuth recycle (from bismuth irradiation and processing for Po^{210} recovery) have been studied. It appears that glove box techniques will be required unless polonium DF's in excess of 10^4 are secured.

Planned pilot plant work on the CSREX process has been completed with definition of operating characteristics of the solvent extraction columns.

In continuing studies related to the recovery of fission products from high level wastes, it was determined that synthetic zeolite Linde 13X is a selective sorbent for Ce^{+3} ions.

Laboratory studies to define the flowsheet to be used for final purification and reduction to metal of the one-kg quantity of CPD-recovered technetium are continuing.

Six ion exchange runs were made during the month to explore the effect of variables on the acid side (HEDTA) of the promethium purification process. This process continues to look very promising.

The following compounds of interest as isotopic heat source materials were successfully prepared: strontium fluoride, neodymium borate, cerium borate, strontium titanate, and neodymium metal (a stand-in for promethium metal).

Operations of the Salt Cycle Process in C-Cell of the HLRCF using "cold" U-Pu have been terminated after four electrolytic depositions. The equipment is being readied for processing irradiated PRTR fuel elements.

In supporting laboratory studies, equilibrium conditions and reaction rates were measured for plutonium and uranium reactions with O_2-Cl_2 and HCl in equimolar LiCl-KCl, under conditions representative of those employed in the C-Cell demonstration runs.







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Computer economic studies of close coupled fuel reprocessing feasibility have continued. The results indicate economic incentive for the close coupled plant (in comparison to a centralized plant) for a 1000 Mw_e reactor complex when fuel exposures are in the range of 10,000 to 20,000 Mwd/ton.

Hot cell studies of high level waste calcination were continued with a successful spray calcination run in which cold fission product stand-ins were added to Purex waste to simulate the chemical composition of waste from 10,000 Mwd/ton power fuels.

In cold pilot plant work five very successful melt runs were made in the continuous melter for spray calcination product. Recent evidence points to the segregation of melt constituents in the melter.

Laboratory glass-making studies confirmed the large chemical effect of fission products, at levels expected in power fuels, on the formation and properties of glasses proposed for waste fixation.

Promising results were obtained in efforts to alleviate the solids problem which has hampered complete demonstration of an integrated process for decontamination of alkaline Purex waste condensate. Hypochlorite added to the condensate collection tank appears to alleviate the bacterial problem.

An irradiated N-Reactor inner fuel element was inductively heated to 980 C successively in three positions along its length to characterize the swelling or other changes in an element when water is lost from the coolant channel and the element overheats. Swelling was pronounced but the cladding did not rupture.

A study is underway to define more accurately the ground-water travel time from the 1301-N Crib to the Columbia River. Preliminary evaluations indicate that a large safety factor is incorporated into the 11-day estimate previously made.

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An 81-node, three-dimensional resistance network, assembled and tested to check errors associated with resistor tolerances and continuum simulation with discrete resistors (related to ground-water regime simulation), provided information supporting earlier studies with two-dimensional networks.

A recent sediment core sample from the McNary reservoir was analyzed for sediment deposition rate by the isotope ratio technique. In the most recent 6-month period, the deposition rate was several times faster than the normal measured rate for older (deeper) sections. No immediately apparent reason seems to exist for such an increase.

The removal of methyl iodide from an air stream by several liquid scrubbing solutions was determined to be very inefficient by the usual scrubbing solutions, but highly efficient by a small amount of silver nitrate in 95% ethyl alcohol.

Spectrofluorometer studies of uranium in aqueous solution showed that as little as 2×10^{-9} grams could be detected by this means, giving promise that this method can become a more useful technique than the standard process.

An effort to improve the reliability of present methods for analyzing reactor fuel burn-up has increased the precision of analysis by a factor of about two. The precision is now sufficiently good that definite biases between alternate methods have been detected. Efforts to determine the cause(s) are underway.

4 Biology

The incidence of fish infected with columnaris when the Columbia River water is cold does not seem to be markedly smaller than when the water is warm.

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By using Sr^{90} it appears that deposition of calcium in the skeleton of crayfish occurs, contrary to established belief, in the exoskeleton before the molt of the crayfish.

The skeletal retention of Sr^{85} from SrTiO_3 was observed to be 0.15% of the administered dose 10 days after oral administration to miniature swine. (When given as the chloride, the absorption of Sr is about 50 times greater.)

Pig skin exposed to vapor of I^{131} showed absorption and peak deposition in the thyroid of 0.9% of the original skin activity. Earlier work with aqueous solutions of NaI showed about 3-4% uptake.

Young female miniature swine were found to develop severe nephritis from the injection of radium more readily than older animals. There is a possibility that this is not strictly an age dependent effect since equivalent doses per kg of body weight were administered, and the older animals had more fat.

Intestinal perfusion studies using rats are being performed to find the effectiveness of various chelating agents in removing plutonium from the animal body. DTPA increases the excretion of plutonium into the GI tract by way of the bile by about 10%. Urinary excretion increases by 20%. However, the greatest amount of plutonium is still excreted via the intestinal tract.

Two samples of $Pu^{239}O_2$ dusts were received from the British Atomic Energy Authority, Aldermaston, England. One was prepared by oxidation of the delta metal at 120 C and the other at 450 C. Both dusts were carefully sized and characterized. Comparisons will be made with locally prepared dusts.

Continued delay in obtaining the new dog runs has begun to sericusly affect the inhalation research program. Temporary runs are being constructed that are designed to serve for a few months.

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Diethyldithiocarbamate, administered to rats directly before wholebody radiation, was found to have no radioprotective effect. In fact, it appeared that there may have been some synergism.

Male rats irradiated with 150 rads of neutrons showed no loss of reproductive capacity. Litter sizes were normal. However, when irradiated females were mated with nonirradiated males, fecundity and litter size decreased.

Radioiodine gas deposited on one surface of the leaf was found to diffuse through the leaf at the rate of 0.1 cm per hour. Since the leaf (coleus) has no stomatal openings on the upper surface, the passage is presumed to occur through the waxy outer cuticle of the upper exposed surface. This tends to question reports from other laboratories that ascribe the penetration of iodine through the leaves as occurring through stomatal openings.

To further define the loss of I¹³¹ deposited on plants in the field, leaves containing radioiodine were exposed to different temperatures in the laboratory. Accelerated removal occurred, but lasted for only 24 hours.

A body temperature telemetering system developed by the Nucleonics Instrumentation group was calibrated and tested in sheep. Accuracy of the system was found to be within acceptable clinical standards.

5. Programming

Calculations indicate interesting benefits from mixing $U^{238}O_2$ and ThO_2 in the same fuel channel in a soft-spectrum reactor. Since both U^{238} and thorium cross sections are highly dependent on resonance absorptions, and since their resonances do not overlap significantly, the resonance captures in each material can occur without reducing those in the other. This effect might be exploited by increasing the fertility of the reactor, reducing the resonance escape probability without having to adjust the lattice. and reducing fuel cycle cost through extending the fuel reactivity lifetime.

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Idealized evaluations of seed blanket reactor systems suggest that a neutron multiplication factor, k, approaching unity may be necessary to develop the full potential of the seed blanket concept.

TECHNICAL AND OTHER SERVICES

There were eight new plutonium deposition cases confirmed by special bioassay analysis during the month. Six resulted from inhalation of contaminated air during the fire in the Redox Final Products Concentration Building (233-S), and one case resulted from a contaminated injury received in the Weapons Manufacturing Building (234-5). All seven were estimated to be depositions of less than 1% of the maximum permissible body burden (MPBB plutonium, with bone as reference, is $0.04 \mu c$). The eighth case, estimated to be a deposition of 3% of the MPBB, was detected by routine bioassay sampling. A re-evaluation of a deposition case originally estimated to be less than 1% of the MPBB resulted in its removal from the list of confirmed deposition cases. The total number of individuals who have received internal plutonium deposition at Hanford is 334 of which 242 are currently employed.

Concentrations of fallout materials in the air of the Pacific Northwest continued to decrease during January. The monthly average of 0.6 pc g/m^3 was the lowest recorded since the Fall of 1961. Averages for the months of November and December, 1963, were 1.0 and 0.9 pc g/m^3 , respectively.

Meetings and consultations were held with AEC-TAB personnel, and others from NRDL and Rand, in connection with a new project on postnuclear attack economic-biological interactions.

In connection with current AEC-GE efforts at economic diversification for the Tri-Cities, work has begun extending previous Tri-City Area economic modeling and analyses.

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A statistical analysis is being performed for IPD on a test of the effect of annuli thickness (of spire-core and can-core) on AlSi bonding integrity measurements.

The minimum variance inventory method for the control of Z-Plant component MUF's is now being applied by NMM.

Under the 2000 Program, discussions were held regarding the possibility of developing a technique for studying flow in heterogeneous porous media analogous to the hydrodynamical concept of "streak functions."

Under the 4000 Program, additional understanding of the complex process of particle packing has been gained by the successful analysis of the geometry of multiple spirals.

Statistical analysis was begun of data relating to second phase particles in dilutely alloyed dingot uranium. Results should help determine dependency of density volume fraction of such particles on alloy composition, time, and temperature of both solutionizing and precipitation anneals.

On 6000 Program, final data obtained from EDPM "triangular" diffusion model have been submitted for evaluation by program originator.

Statistical analysis was started of data from a study to investigate beetle populations of selected communities.

SUPPORTING FUNCTIONS

PRTR output for January was 1482 Mwd, for an experimental time efficiency of 78% and a plant efficiency of 68%. There were five operating periods during the month, two of which were terminated manually and three were terminated by scrams. A summary of the fuel irradiation program as of January 31, 1964, follows.

	Al-Pu		UO2		PuO2-UO2		Other		Program Totals	
	No	Mwd	No.	Mwd	<u>No.</u>	Mwd	<u>No.</u>	Mwd	<u>No.</u>	Mwd
In-Core Maximum Average	С		6	982.2 251.7 163.7	79	$9 \ 349.8 \\ 221.3 \\ 118.4$			85	10 332.0
In-Basin	43	3 726.4	26	2 882.1	24	1 150.2			93	7 758.7
Buried							1	7.3	1	7.3
Chemical Processing	<u>32</u>	2 309.3	<u>35</u>	1 965.8					_67	4 275.1
Program Totals	75	6 035.7	67	5 830.1	103	10 500.0	1	7.3	246	22 373.1
Note: (Mwd	l/Elemen	t) x 2	$20 = \sim Mw$	d/tor	${}^{\rm h}_{\rm U}$ for UO $_2$	and P	uO ₂ -U	0 ₂ .	

A total of 59 reactor outage hours were charged to repair work. Main items were:

Instruments	32	hr
Valves	11	hr
Process Tube to Nozzle Gaskets	6	hr

The January heavy water inventory indicates a loss of 700 lb for the month.

Startup work for the H_2O moderated PRCF continued during the month, which included preoperational testing, completion of project exceptions, modifications to the facility, administrative preparations and some repair work. At month-end the fuel follower sections for the rods are the last major items still needed.

FERTF test 4-0, using a defective Al-Pu element, was completed. Considerable difficulty with airborne radioactive contamination was encountered in the loop annex cell, equipment room and tunnel. Air activity problems were evaluated while FERTF test 4-1, utilizing another defective

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Al-Pu element, was being performed. As a result, vent lines to the pump hoods were installed upon completion of FERTF test 4-1, thereby eliminating the air activity problems.

A preliminary schedule was prepared for the chemical processing of 35 Al-Pu fuel elements in March.

Total technical shops operation productive time for the period was 20, 130 hr. Distribution of time was as follows:

	<u>Man hr</u>	% of Total
N-Reactor Department	2 681	13.3
Irradiation Processing Department	3 602	17.9
Chemical Processing Department	482	2.4
Hanford Laboratories	13 365	66.4
Hanford Utilities and Purchasing Department	-	-

Total productive time for Laboratory Maintenance was 23,300 hr of 24,600 hr potentially available. Of the total productive time, 91% was expended in support of Hanford Laboratories components, with the remaining 9% directed toward providing service for other HAPO organizations. Manpower utilization (in hours) for January was:

Α.	Shop Work		2 400
В.	Maintenance		8 400
	1. Preventive Maintenance	2 500	
	2. Emergency or Unscheduled Maintenance	1 700	
	3. Normal Scheduled Maintenance	4 200	
С.	R&D Assistance		12 500

The heavy water inventory at the end of January 1964, showed a loss of 701 lb (\$9623) for the PRTR. Heavy water scrap generated during the month amounted to 2127 lb, resulting in a \$2574 charge to operating cost. Heavy water accumulated at month end for return to SROO amounted to 11,006 lb, valued at \$137,818. Fifty-five drums (27,445 lb) of heavy water were received during the month, valued at \$375,448.



Cumulative data on Hanford visits:

	Number of Visitors				
	In January	Since June 13, 1962			
Visitors Center	1 034	61 319			
Plant Tours	38	n.a.			

HAPO professional recruiting activity this month is summarized below:

	Plant Visits	Offers Extended	Acceptances Received	Rejections Received	Open Offers at Month End
Ph. D. BS/MS (Direct	8	5	2	1	4
Placement)		4	4	0	1
BS/MS (Program)		1	6	17	27

Seven technical graduates were placed on permanent assignment. Four new members were added to the roll and none terminated. Current Program total is 67.

Authorized funds for nine active projects total \$7,592,500. The total estimated cost of these projects is \$10,839,000. Expenditures on them through December 31, 1963 were \$2,038,000.

Manager, Hanford Laboratories

HM Parker: JEB:dph

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REACTOR AND FUELS LABORATORY MONTHLY REPORT

JANUARY 1964

TECHNICAL ACTIVITIES

A. FISSIONABLE MATERIALS - 02 PROGRAM

1. Metallic Fuel Development

N-Reactor Fuel Evaluation. Examination of an N-inner fuel component irradiated to 2500 MWD/T in KER-4 has started. This is the companion piece to a 3100 MWD/T outer tube that has been previously examined and reported upon. Superficially, the component performed well with no indications of warp, surface bumping or rippling, clad striations, crud deposits or corrosion effects, and the brazed end closure and inner bore appear unchanged. Destructive examination indicates no deficiencies in the performance of the closure or the associated heataffected zones in the fuel, clad, and bond. Some micro and macro cracks have been observed in the fuel, but these are believed to have been produced during post-irradiation handling of the component.

Specimens of an inner tube irradiated to 3100 MWD/T have been pulse annealed to 980 C in support of reactor hazards studees. Fuel volume increases of 19, 26, and 38% were obtained for times at temperatures of 0, 5, and 30 minutes, respectively. Metallographic examination of the pulse annealed specimens shows the development of a large population of gaseous voids in the fuel matrix that correlates with the measured volume increases. The fuel component showed no evidence of localized clad strain or failure, and no evidence of release of activity.

Irradiation of Fine Carbide Uranium Fuel. In a test to investigate the ability of a submicron dispersion of uranium carbide to reduce uranium swelling, three NaK-filled capsules will be irradiated in the ETR to evaluate the performance of fuel rods produced from chillcast uranium shot. Two of these capsules each contain two fuel rods identical in uranium composition, but with one having a uranium carbide size of 2-5 microns produced from arc-melted uranium and the other a carbide size of less than 0.5 micron produced from the uranium shot. The third capsule contains two fuel rods with the fine carbide. One capsule, GEH-14-609, has been irradiated for one cycle in the ETR and has accumulated an exposure of 0.05 at% burnup. Goal exposure for this capsule is 0.3 at% burnup. During the first cycle of irradiation the capsule operated at a maximum core temperature of about







420 C. The capsule will be moved to another location in the reactor for the second cycle in order to attain the test design fuel temperature of 600-625 C.

Target Element Development. Irradiation testing of eight experimental lithium-aluminum target elements in KER Loop 2 was completed after the elements had accumulated 124 days of exposure at full reactor power. Examination of the targets in KER Basin was made and the elements appeared unchanged as a result of irradiation.

Four additional target elements have been prepared for irradiation in the KER Loops. Two of these elements are of advanced design and contain a nominal 3.2% natural Li in Al, and the target cores have been bonded to the Al can. In addition, the axial hole in the target core has been omitted. The other two elements are of identical design to those previously irradiated. One of these has a composition of 2 w/o Li (0.20 wt% Li⁰) and will serve as a reference element while the other contains a nominal 8 wt% natural Li.

Co-producer Target Elements. Target elements for the co-producer production test PT-645-D were completed on schedule. These elements have an aluminum-lithium core, 0.440-inch diameter containing 0.56% Li enriched with 63% Li⁶. The target cores are double clad. The first cladding is coextruded aluminum over the fertile core to prevent tritium diffusion into the Zircaloy jacket. Aluminum end plugs are welded into the counterbored ends to complete the gas seal, then X-rayed to assure weld integrity. The aluminum clad elements were then coated with dry graphite and inserted into Zircaloy sleeves. These sleeves had the supports pre-welded in position to assure that no aluminum contamination of the weld would occur. After assembly the target elements were evacuated, back filled with helium, then end plugs were welded in position. Each element was checked for helium leakage by means of a helium mass spectrometer leak tester, x-rayed for weld defects, then autoclaved for 72 hours in 300 C water. Fifteen target elements, 26 inches long, were delivered to NRD for final assembly with the driver elements.

Weld Development - Target Elements. The welding of 18 lithiumaluminum target elements clad in aluminum and Zircaloy with supports can be divided into three phases.

Welding of aluminum end caps into the coextruded aluminum clad, $\frac{1}{2}$ -inch diameter rods was done manually by the TIG process using AC and argon shielding gas. Some porosity was encountered when one group of end caps were handled with a leather glove, but this was eliminated when

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tongs were used to place the end caps in position. The welds were checked by x-ray.

Second, attaching of Zircaloy supports to the Zircaloy cladding tubes was done by resistance projection welding prior to loading the aluminum clad cores into the Zircaloy sleeves. A copper mandrel was used. Welds were checked by destructive tests on an identical tube and supports before, during, and at the end of the group used in the actual tests.

Third, welding of Zircaloy end caps into the Zircaloy tubes after they had been loaded with the aluminum clad cores was done in the 308 Building under the direction of Lloyd Lemon. Welding was done in a helium filled glove box using the TIG process. Prior to welding the glove box was evacuated to insure that the elements would contain helium as a heat transfer medium between the aluminum and the Zircaloy clad. After welding, the elements were helium leak checked and x-rayed.

Corrosion Rates of Aluminum-Lithium Alloys. A series of corrosion tests have been designed to determine the danger of exposure of lithium containing aluminum alloys to reactor water. Short term corrosion data in 100 C water up to 110 hours indicates that in the first 20 hours annealed alloys containing up to $3\frac{1}{27}$ Li corrode twice as fast as the cold worked alloys. However, the 1100 aluminum standard and the 0.5% Li alloys behave exactly the opposite with the higher weight gains in the cold worked condition. Corrosion rates of the 0.5% Li alloys and 1100 aluminum were the same, whereas the other compositions showed an increased corrosion rate with increasing lithium content. After 30 hours there were no further weight gains for any of the alloys and all observable surface reaction had stopped, indicating that the lithium contents had been depleted to the point where only the aluminum corrosion rates were the main contribution.

Fluted Fuel Element Irradiation. The two N-single tube fuel elements with fluted outer surfaces which are being irradiated in the ETR have successfully completed nine cycles of irradiation in the M-3 pressurized loop for an accumulated exposure of about 1800 MND/T. After the eighth reactor cycle (1600 MND/T), the elements had undergone a volume increase of 0.9%. The elements have been operating at a maximum specific power of about 130 kw/ft with a corresponding core temperature of 520 C. A Zircaloy-2 basket has been prepared as a replacement for the stainless steel basket which is currently being used for holding the elements in place in the loop. This replacement should result in an increase in the power level for the remainder of the irradiation.



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Cladding Deformation Studies. Post-irradiation examination continued on Zircaloy-2 clad uranium rods from capsule irradiations to determine the strain capabilities of Zircaloy-2 cladding as a function of cladding thickness, cladding thickness uniformity, and temperature. Results from visual examination and diameter measurements from the 94 specimens in this test and the correlation of cladding strain limits with cladding thickness uniformity have been reported previously.

Detailed measurements are being obtained on transverse sections of some of these rods to assess the plastic strain behavior of the cladding at striations of known pre-irradiation geometry. On specimens examined to date, about 15% of the total cladding strain occurred in the striations of two specimens. About 40% of the total strain occurred in the striations of a third, and in this specimen the necking occurred from both the inside and outside surface of the cladding. Metallography is being completed on a fourth rod in which the cladding necked severely and then ruptured. Results from this rod should complete the picture of the progressive stages from the iirst indications of the plastic instability to the ultimate rupture.

Alternate Uranium Composition. Studies are in progress to determine the effects of altered fuel compositions upon fuel element fabrication, corrosion behavior, and irradiation swelling resistance.

The comparative irradiation behavior of single tube fuel elements (1.790" OD x 0.975" ID) fabricated from standard N-fuel composition and an alloy containing 400 ppm Fe and 800 ppm Al will be determined from high temperature loop irradiations. The major impurities in these materials are: 510 ppm C, 137 Fe, 6 Al, 127 Si for the standard composition and 537 ppm C, 386 Fe, 837 Al, and 47 Si for the altered fuel. The production test fuel elements were beta heat treated and structural characterization of the ingots, primary extrusions, coextruded fuel, and completed fuel elements is in progress. Major differences are observed in grain size, second phase particle distribution, and hot hardness between the two materials.

Second Generation N-Outer Support. The hot forging method of making a stronger N-outer fuel support has been successfully demonstrated. It was necessary to set up the tooling in the 600 KVA resistance welder to obtain forging pressures of 12 tons. The primary qualifications for the new support are permanent set characteristics. The new support exceeds the permanent set criteria.

Uranium Alloy Investigation. Evaluation of a series of binary and ternary alloys of uranium with up to 5% total nicbium and/or zirconium



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is continuing. Alloy buttons were prepared by arc melting. Hardness measurements were taken and metallography is being completed on the as-cast buttons. During this month the alloy buttons were hot rolled into sheet which will be cut into corrosion coupons and given various heat treatments. The prepared coupons will be autoclaved under varying conditions of temperature and pH.

It was originally proposed to study the corrosion rates in cold worked material as well as heat treated material. However, several of the alloys are sufficiently brittle to preclude giving them an appreciable amount of cold work. The compositions of these alloys are 2 and 3 wt% Nb, 4 and 5 wt% Zr, and 2 through 5 wt% combined Zr and Nb. The hardness of these alloys can be attributed to solid solution hardening.

Uranium Sulfur Alloys. Samples of uranium-sulfur alloys (O-1 wt% S) heated for 24 hours (450-1100 C), and water quenched, have been under investigation. This study has been completed, and a report on the work done to date is in preparation. Metallographic studies indicate little or no gamma solid solubility of sulfur in uranium. No eutectic information was observed in the O-1 wt% sulfur range.

Hot Headed Closure Studies. Macroscopic examination of 65 hot-headed and machined fuel element sections revealed a number of small edge defects. These edge defects, rounded edges and laminations due to the back-extrusions process, would tend to generate defects in the projection weld and tend to promote crevice formation.

After the elements were remachined, inspection revealed that 19 were satisfactory, 32 had one to two small defects remaining, and 16 were total rejects. It is believed that most of the 32 can be used after remachining although there will be variation in the width of the exposed uranium. The ring projection spacing on the caps can be varied to accommodate the exposed uranium width variation.

Brazing Alloy Corrosion Rates. Earlier corrosion data indicated that catastrophic weight losses occurred in the 1.1-2.0% Be range of Zr-Be alloys after 500 hours in pH 10, 360 C water. These same losses were not observed in pH 7, 360 C water. Now, after 2500 hours of exposure, increased weight losses have begun to appear in the same concentration range in the pH 7, 360 C water test. In the meantime the weight loss range in pH 10 has spread to 0.8 to 2.2% Be after 1500 hours. These alloys appear to be very composition sensitive as well as structure sensitive. Zircaloy-2 containing uranium up to 1% and tested at both pH levels compares very favorably with pure Zircaloy-2.



Corrosion of N-Reactor Fuel Materials by High Temperature Salt. Short time corrosion rates of Zr-2 clad uranium in high temperature salt were measured to determine the feasibility of some special heat treatments of irradiated fuels in salt. Samples of Zircaloy-2 and uranium were immersed in a salt mixture of 90% barium chloride and 10% sodium chloride at 980 C (1800 F) for two periods of five minutes each. Samples of Zircaloy-2 plus 5% beryllium brazing alloy were given the same treatment, except at a temperature of 900 C (1650 F). Corrosion, although fairly rapid, was uniform and noncatastrophic.

<u>Creep Penetration</u>. An apparatus for determining the hot hardness of materials as a function of time, temperature or load has been built and installed. Load (100-1000 lbs) is applied to a 10 mm tungsten carbide ball in contact with two samples while temperature is controlled by immersion of test capsules in a nitrate salt bath. Final indention diameter is a measure of hot hardness and creep resistance. Samples of Zircaloy-2 and uranium have been investigated with this apparatus. Further investigation to obtain values over a range of temperatures and loads is now in progress. A report on "Creep Penetration Testing Procedure" will be available by the end of January.

Extrusion of Silicon Cladding Alloys. A tes quantity of two aluminum-silicon alloys were extruded for IPD Production Fuels Section to evaluate the feasibility of using the alloys for cladding material in the hot die sizing program. The two alloys were Al, 10% Si, 1% Ni, 0.5% Fe and Al. 6% Si, 1% Ni, 0.5% Fe, 1% Mg which are believed to have good corrosion performance. Both alloys appeared to extrude satisfactorily at a 20 to 1 reduction when heated to 300 to 400 C. Considerable extrusion die pickup was experienced in the beginning resulting in rough extrusion surfaces. This condition was corrected by altering the extrusion lubricant and speed.

The material was all extruded into small tubes for use as ID cladding components. Silicon alloy tubes for OD cladding components will be extruded in the near future.

Examination of Hot Die Size Diffusion Bonded Fuel Elements. Fuel elements fabricated by the hot die size process and irradiated under production test IP-546-A increased in both cuter and inner diameters to a greater extent than AlSi canned control fuel elements. Length changes are not available as yet.

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Metallographic examination was made of one transverse section from the conter region of a hot die sized element irradiated in a center position of the reactor column. No evidence of porosity or grain boundary tearing was found. The dimensional changes that occurred are believed to be due to anisotropic growth resulting from crystallographic texture introduced during the canning of these fuels.

Comparison of the uranium microstructures of hot die sized and Al-Si canned elements indicates that the temperatures and mechanical loads present during the hot die sizing process were sufficient to cause observable structure differences. Further examination of elements from several steps in the canning process is in progress.

Stainless Steel Clad Niobium Tube Fabrication. A 316 stainless steel clad Nb-1% Zr tube was produced by a combination of hot extrusion and cold swaging. 1020 steel was used as a sacrificial mandrel. To expedite removal of the steel, a steel tube was utilized and filled with solid graphite. The graphite could be later removed by drilling, permitting full length access of acid for steel removal. To insure a bond all the internal surfaces of this billet were cathodically cleaned and sealed by electron beam 'slding.

The extrusion billet was preheated to 1150 C and extruded from the $2\frac{1}{4}$ " extrusion container in a 0.030" thick graphite sleeve to 3/4" diameter. The extrusion was then cold swaged to $\frac{1}{2}$ " diameter, the graphite drilled out and the 1020 steel chemically removed, leaving the 0.020" thick stainless clad 0.040" thick Nb tube.

A diffusion layer between the stainless and Nb was just discernible when examined at a magnification of 750%. Samples of the extrusion were prepared and heat treated at temperatures of 900, 1000, and 1100 C for times of one and three hours at each temperature. Examination of the samples indicated that the diffusion width increased in all six samples, the amount of growth being proportional to the time and temperature. Peel tests to determine the ease of separating the stainless from the Nb at the interface indicated that the diffusion bond was very brittle as the samples separated considerably easier as the width of the diffusion layer increased. The asextruded material was by far the most difficult to separate.

Further work along this line will require the use of a material at the interface to promote bonding and prevent the formation of a britcle compound layer.

Pyrometallurgical Decontamination of Uranium. The U-238 isotope decays by a series of radioactive decay steps to produce a number



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of high energy alpha emitting elements of which the principal one is Th-230. Uranium more than six months since separation will have reached equilibrium in Th-230 and U-234 contents. A series of pyrometallurgical decontaminations were made by melting available stored uranium (3 years or more old) in zirconia crucibles and holding for three hours at 1400 C, then pasting into a graphite mold. Results were very encouraging. On the basis of alpha counting 1.7% of the original activity remains in the cast uranium, the balance being held in the slag crust on the crucible. Additions of powdered zirconia maintained the alpha activity at 1.7% of the original; however, UO₂ additions lowered the alpha activity to 1.4% of the original. When 0.1% natural thorium was added to the uranium, and run t³ rough the normal melting procedures, 5.5% alpha activity remained, but when melted with 10% UO₂, the activity was lowered to 4.1%.

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Thoria Fuels Development. Approximately one ton (632 fuel elements) of low uniform bulk density (65.5-67.3% TD) "Sol-Gel" thoria was canned. This material was shipped to the Irradiation Processing Department on January 17, 1964. Work was begun on the second ton of low density "Sol-Gel" thoria using new equipment recently installed.

Experimental work to achieve a high density thoria mixture that utilizes a maximum percentage of the material as delivered produced encouraging results. A density of 82.5% was obtained using a low power vibrating table. It is anticipated that both pneumatic and electronically controlled vibrators will substantially increase this figure. Installation of electronically controlled vibration equipment was started. Installation of a pneumatic impaction vibrator was completed.

Rupture Testing of Irradiated Fuel Elements in IRP. One N-Reactor inner fuel element, irradiated to 1200 MWD/T in KER, was rupture tested in the IRP. This element had a bonded tapered end cap, and it was defected with a 0.025" hole at the end cap. After 162 minutes of incubation at 300 C, the fuel element began rupturing. The water was cooled at the current N-Reactor shutdown rate. Examination of the fuel element showed the end cap had come completely loose and the rupture extended about an inch down the fuel element for the entire circumference.

2. Corrosion and Water Quality Studies

Protective Coatings for Aluminum. Two commercial marking inks were found to reduce the corrosion of aluminum by factors of 5 to 10.





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Coated samples of aluminum alloy X-8001 were exposed at the downstream end of process channels in F-Reactor to process water (treated river water) at pH 6.6 containing 1.8 ppm sodium dichromate.

Stress Corrosion Cracking of H-Reactor Crossheader. Tests were conducted to determine whether reaming the pigtail-connection fittings of the stainless steel H-Reactor crossheaders would result in susceptibility for stress corrosion cracking. Full scale portions of the crossheader, which included one or two pigtail fittings welded in place, were exposed in boiling 42% MgCl₂ solution for periods of 65 to 150 hours. Significant cracking was produced in each of the crossheader pieces, both opposite the fittings and at the welds. Longitudinal cracks were also produced in the bevel-thread region of the fittings on only the samples exposed for 150 hours. Thus, the reamed fittings were somewhat less susceptible to cracking than unmodified portions of the crossheader.

Testing of NH4OH in KER Loops. The first pair of instrumented UO2 crud detectors (thermocouple elements) was discharged from KER-1 after going to goal exposure. Their performance was satisfactory during the entire test. No increase in cladding temperature occurred during the test, which was with ammoniated coolant at pH 10. Visual inspection showed little or no deposit of crud on the discharged elements. A second identical pair of crud detectors, one upstream and one downstream, was charged into KER-1.

Final design was completed for a cermet (UO_2-MO) core crud detector which will be capable of longer exposures and higher heat flux than the present detectors. Two such elements are now being fabricated.

Operation of KER-1 (carbon steel) with ammonium hydroxide was continued during the month. During a seven-day period at low temperature with a pH of 10.1, the rate of loss of ammonia was 0.1 lb/day, which is higher than the 0.03 lb/day observed at high temperature. The total gas concentration was also several times greater than at high temperature; this is attributed to increased radiolysis of ammonia. Hydrazine was observed as a steady radiolysis product and apparently inhibited the buildup of radiolytic O_2 and H_2O_2 , as no gross formation of either occurred during this low temperature operation.

Water quality in KER-1 remained good during both high and low temperature operation at pH 10. Turbidity was 0.5 to 1.0 ppm, total solids 0.5 to 4.5 ppm.



A test was run in KER-1 to determine the crudding characteristics of ammoniated coolant at pH below 10. The pH was held at 9.5 for seven days, during which no gross crud buildup or release was indicated by the thermocouple elements, activity bursts, or crud concentration in the coolant. The pH was then reduced to 8.0 for four days; crud data for this period are not yet complete.

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Operation of KER-4 (stainless steel) continued at pH 10.1 using NH4OH. The rate of loss of ammonia was 0.18 lb/day at low temperature and 0.05 lb/day at high temperature. Water quality remained good. Turbidity was 0.5 to 1.0 ppm; total solids 0.5 to 2.5 ppm.

Loop KER-2 (stainless steel) was converted from LiOH to NH_4OH . Prior to the conversion at pH 10 with LiOH, the total dissolved gas and O₂ concentrations were high due to radiolysis of the coolant (about 60 cc/kg total gas and 18 ppm O₂). During low temperature operation at pH 8.5 with NH4OH, these concentrations decreased significantly (to about 12 cc/kg total gas and 6 ppm O₂) after three days. Coolant pH was then increased to 9.3-9.5 and total gas increased (to about 20 cc/kg) while O₂ disappeared. These results demonstrate that even at pH 8.5, NH4OH conditioned coolants inhibit radiolytic oxygen formation. Coolant quality was acceptable during this period, although total solids concentrations increased somewhat. Ion exchange cleanup operation is being employed for both coolant purification and pH control.

N-Reactor Steam Generator Tubing. A section of intergranularly attacked 304 stainless steel tubing from N-Reactor steam generator 4A was tested in TF-7 with 520 F, 1700 psig, pH 10 lithiated water for 43 days to determine the rate of propagation of a pinhole leak. The leak rate varied between operating periods from 5 to 20 ml/hour except for one three-day period at about 30 days of exposure, when the leak rate was 300 ml/hr. Metallographic examination revealed there was not any wire drawing or caustic embrittlement. Two additional samples were similarly tested in TF-7 and failed within a few hours due to an increase in size of the pinhole defect by cracking.

All the samples had been previously exposed at 300 C. 3000 psig until a pinhole developed at the intergranularly attacked areas. It is felt that this drastic method of producing the pinhole leak was probably responsible for the failures observed. Attempts to make pinhole defects by drilling a 0.003-inch diameter hole from the OD of the tubing into the intergranular attack areas were not successful.

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A test employing tubing defected by pre-exposure to 300 C, 3000 psig water is in progress in TF-2 in 550 F, 1340 psig, pH 10 ammoniated water. The leak rate remained constant at 9-14 ml/min for 18 days, then went to zero after a shutdown period. After a second shutdown, the rate increased to 54 ml/hr for one day, then returned to zero.

Samples of intergranularly attacked N-Reactor steam generator tubing were removed from TF-4 after 2, 4, 6 and 8 cycles to a sulfamic acid decontamination process followed by recirculation of high temperature water. The decontaminations progressively widened the attacked area at each grain boundary until by the eighth cycle all the grains in the attacked area had fallen out. Areas which had not been previously intergranularly attacked were not corroded. The areas which had been intergranularly attacked were not increased in width or depth.

Effect of Cleaning Solution on N Fuel Elements. Chemical cleaning of the N-Reactor primary system is being considered to remove heavy rust deposits. A test was performed in the TF-8 carbon steel loop to determine the corrosion resistance of N-Reactor fuel elements to a proprietary ammonia-EDTA cleaning solution. A 4% solution was used for 24 hours at 250 F. These conditions are more severe than those planned for use in N-Reactor in order to accelerate any adverse effects of the cleaning solution. The system was possivated following the cleaning with $\frac{1}{2}$ oz/gal of a 50:50 mixture of NaOH-NaNO₂.

Examination of the fuel elements revealed no attack to the Zircaloy-2 components. However, the elements had remained in stagnant water for two days following the cleaning which caused 2-5-mil deep pits on the carbon steel support shoes. Also, the cleaning solution had loosened magnetite from the loop walls and this had plated out on the fuel elements to form a thick, black, slippery coating. Since N-Reactor piping does not contain a magnetite film yet, this plating out is probably not indicative of what will happen in N.

Following the cleaning, the fuel elements were exposed two weeks to recirculating 500 F deionized water at pH 10 (NH_4OH). The elements were in good condition following this exposure. Corrosion samples of Zr-2, silver plated steel, copper tubing, and 17-4 pH stainless steel were also exposed to the entire test and none showed any adverse effects of the chemical cleaning.

Fretting of N Fuel Supports. Fretting corrosion studies on N-Reactor fuel elements in flowing, high temperature, pH 10 water with applied external vibrations were continued. A range of induced vibrations from 50 to 90 cps at 5 cps increments was studied. The inner fuel



element supports had failed during a previous test at these conditions but remained in excellent condition during this test. The test is being repeated.

Stability of Valve Packing. Some of the valves in the N-Reactor primary system have Teflon-impregnated asbestos packing rings, which are contacted by the fluid passing through the valve. Samples of this packing were tested at 535 F in pH 10 ammoniated coolant. The weight loss rate for the packing was about 0.1% per week. No measurable buildup of chloride or fluoride was observed in the coolant, and no significant change in pH occurred during a one-week test. Additional tests are being conducted at 200 F.

Anodic Passivation Studies of Corrosion by Decontaminants. Studies on the anodic passivation characteristics of carbon steel were continued. Scanning tests with aerated dibasic ammonium citrate were completed. It was found that there was a passive region at room temperature in the concentration range of 0.1 M to 1.0 M dibasic ammonium citrate. At elevated temperatures there was greater corrosion and narrower passive regions.

Preliminary results from testing on solutions of dibasic ammonium citrate and 0.01 <u>M EDTA</u> are encouraging. The addition of EDTA to the ammonium citrate solutions broadens the passive region and reduces the passive region current density. This indicates that EDTA acts as an inhibitor.

Chemonuclear Studies. The H-1 Loop was decontaminated to reduce activity and to obtain a clean system for testing of the chemonuclear production of hydrazine from ammonia. An alkaline permanganate-sulfamic acid procedure was used, and a DF of about 10 was obtained.

3. Gas-Atmosphere Studies

Kinetics of the Reaction $CO_2 + H_2 \rightarrow H_2O + CO_3$. To improve the reliability of the experimental data, the equipment for studying the water-gas reaction was modified to include (1) enclosed thermocouple wells in the reaction zone in place of wax seals (2) an infrared analyzer having a carbon monoxide sensitivity range of 0 to 2% CO, and (3) an electrolytic hygrometer for moisture analyses in the range 0 to 1%. Experiments made with the above improvements have given data which are consistent and considered reliable.

Eight runs were made at 905 ± 5 C with varying carbon dioxide but constant hydrogen concentrations. The scatter of rate constants







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calculated from an empirical equation was less than 8% and the average value agreed almost exactly with the best literature value. Continuing work stresses the effect of carbon monoxide and hydrogen on the reaction kinetics.

Radiolysis of Carbon Monoxide-Water Vapor Mixtures. Previous results on the radiolysis of carbon monoxide and water-vapor mixtures indicated that an Arrhenius plot of the 100-ev yields of hydrogen and carbon dioxide was linear to approximately 350 C, but above 350 C the 100-ev yields were much larger than expected. These large values were attributed to the thermal reaction becoming important above 350 C.

Experiments in the absence of a radiation field, however, showed that the thermal reaction is not significant below 500 C and contributes only slightly even at 600 C. The experiments in a radiation-field were very carefully repeated and revealed that the 100-ev yields (G values) indeed were not unusually high until above 500 C.

It is believed that the earlier results arose because of localized high temperatures in the ampoules due to location relative to the heaters. Additional experiments to determine the effect of helium gas on the radiation chemistry of the CO_2 -H₂O reaction indicate that the energy absorbed by the helium is approximately as effective as the energy absorbed by the carbon monoxide. The uncertainty in the data prevents a more quantitative statement at the present time.

Rate Constants in the Carbon Dioxide-Graphite System. Oxidation rates have been measured for TSX graphite in flowing carbon dioxide and in a carbon dioxide - 4.8% carbon monchide mixture. From data obtained at 800 to 900 C, specific rate constants were calculated. Comparison with data on CSF graphite was made; the CSF being purer is found to oxidize at a lower rate, as expected. Of interest is that the apparent inhibition of the burnout by carbon monoxide is generally less in the case of TSX graphite until a temperature of about 900 C is reached.

The ratio of over-all oxidation rates R_{TSX}/R_{CSF} for 100% carbon dioxide and for a mixture of 95.2% carbon dioxide and 4.8% carbon monoxide was determined to range from 2 to 15.

Thick Film Effects on the Hydriding of Zircaloy-2. Previous experiments have indicated that as the ZrO_2 film on Zircaloy-2 grows in thickness, it becomes permeable to hydrogen gas even in the presence of an oxidizing agent. A second experiment has been started which has confirmed earlier work. Zircaloy-2 coupons with preformed oxide films of 0, 50, 100, 200 and 500 mg/dm² were exposed to helium



contaminated with 2% H₂, 2% CO, and 0.15% H₂O at temperatures of 350 and 450 C (662 and 842 F). After 24 days, samples with 200 or 500 mg/dm² pre-films showed evidence of gas phase hydriding at both 350 and 450 C, whereas samples with 0 and 50 mg/dm² were normal with no gas phase hydriding. For the 100 mg/dm² samples, the increase in hydrogen content was greater than normal but could be accounted for by hydrogen pickup associated with the corrosion process. These results show there is an upper weight gain limit for safe exposure of Zircaloy-2 in a hydrogen containing atmosphere. Data to date predict that as the weight gain and/or temperature increases, the H₂O/H₂ ratio of the N stack gas must also increase to prevent hydriding of the Zr-2 pressure tubes.

Zirconium Graphite Compatibility Loop. The zirconium graphite compatibility loop is now operational. The first experimental use of the loop involved the measurement of the rate of transport of H₂O through an actual N-Reactor tube block, 24 inches long. These data are required to determine the dew point in the reactor gas required to supply H₂O to a process tube at rates sufficient to inhibit hydriding. Diffusion rates are measured by circulating helium of constant known moisture content around the loop, and weighing all the water which diffuses through the graphite on a magnesium perchlorate bed suspended from a recording balance. Measurements made at 400 and 700 C (752 and 1292 F) show the bar to be quite permeable to water vapor with the diffusion rate essentially temperature independent between 400 and 700 C. Initial results indicate that a moisture content of 50 ppm (-54 F dew point) would meet the oxidation demands of the process tube.

4. Process Tube Development

Process Tube Testing. Brittle fracture tests have been performed at room temperature on a series of hydrided N-Reactor process tube specimens containing 275 ppm. 180 ppm. and 90 ppm hydrogen. The hydrided tubes were defected by milled slots, 38 mm (1½ inches) long, and their burst pressures compared with that of a similarly defected unhydrided specimen. The reduction in burst pressure due to the presence of the hydride was 14.6% at 90 ppm, 18.4% at 180 ppm, and 29.2% at 275 ppm. All specimens failed with a full length fracture very brittle in character with little or no evidence of shear. The effect of precipitation under stress on the orientation of hydride platelets in Zircaloy tubing has also been examined. Specimens containing 90 ppm hydrogen were heated to 300 C (572 F) to dissolve the hydrides and allowed to cool slowly in the furnace while internal pressure was maintained. Hoop stresses of 50,000 psi and 25,000 psi were investigated. At both stress levels a strong orientation of hydrides in the radial direction was noted.



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Project CAH-922 Irradiated Burst Test. All of the equipment for this facility is on hand. The major portion of work remaining involves electrical and mechanical connections between the vessels and the instrument panels. The prototype vessel will be decontaminated and modified to fit the project arrangement. Completion is estimated at 93% versus a scheduled 98%. A 30-day extension of the project has been requested.

N-Reactor Steam Generator Tubing. The evaluation of tubing with massive flaw areas in excess of 50% of the tube wall is currently under way. Half of the specimens (three) have failed after a period of 840 hours of exposure to test conditions of 3000 psi and 300 C (572 F).

<u>Crush Tests of N-Reactor Fuel and Dummy Elements</u>. Axial compression tests have been performed upon carbon steel dummy spacers and short fuel elements individually and in tandem while confined in a length of N-Reactor Zircaloy-2 pressure tube. The purpose was to determine the energy absorption capacity that would be available in a full N-Reactor pressure tube in the eve: t the fuel-dummy charge was subjected to axial forces. The details of this report are published in a report entitled "Crush Tests of N-Reactor Fuel and Dummy Elements." HW-80436.

5. Thermal Hydraulic Studies

N-Reactor Studies. Laboratory data obtained in the heat transfer and fluid flow experiments with the full-scale, electrically-heated model of an N-Reactor fuel column with prototypic outlet piping were processed by means of the digital computer program described last month. This program was designed, primarily, for the conversion of instrument readings into engineering parameters, development of flow-pressure drop relationships, and comparisons of powers and flows calculated from different sets of readings to determine errors and uncertainties in the calculated values.

Liquid-phase flow versus pressure-drop relationships were developed from experimental data obtained at 1200 psig for all portions of the system. These relationships are being used in a modified program to develop two-phase to single-phase pressure drop ratios. For various portions of the system, pressure drops were proportional to some power of the flow, the powers falling in the range of 1.8 to 2.0, as would be expected in a system having both frictional and expansion-contraction losses.





Flow rates calculated from two independent sets of instruments showed good agreement; generally within 1%. Agreement between the calculated powers was not as good as that obtained for flow comparisons. Powers calculated from water flow rates and temperature rises were generally about 5% lower than those calculated from electric current and voltage measurements. Calculation methods, assumptions, and correction factors are being examined in an effort to improve the agreement.

Modifications to the computer program to allow calculations of twophase to liquid-phase pressure-drop ratios were completed. This modified program will also calculate boiling lengths in each of the three flow channels in the fuel column model and will separate the total pressure drop into frictional and momentum change losses.

Emergency Cooling of N-Reactor Fuel. Laboratory experiments were performed to investigate surface temperatures and rates of steam generation in N-Reactor fuel elements under certain conditions of emergency cooling. The conditions studied were for the case where emergency cooling would cease for a short period of time, allowing the temperature of the fuel to increase to approximately 1500 F, and then be re-established. The problem is of interest since the steam that would be generated could not be vented to the atmosphere and would cause a pressure buildup within the reactor building.

The test section consisted of two, 6-foot long, steel tubes, one placed within the other to form a 0.160-inch flow annulus. An electrical heating element was placed inside the inner tube to bring the test section to the desired temperature before the water coolant was introduced to the test section. Thermocouples were attached along the lengths of both tubes and were fed to fast response recorders for continuous readout.

The test procedure, consisted of heating the test section while it was dry. A water source was then valved in that would produce a certain flow under cold conditions. Pressures, temperatures, and flow were recorded while the test section lost its stored heat and returned to the temperature of the water. Tests were run at flow rates of 6 and 2 gpm.

The data thus obtained are being processed, but indications are that the film coefficient varied from about 105 Btu/hr-ft²- $^{\circ}$ F at 1500 F to 1350 Btu/hr-ft²- $^{\circ}$ F at a 450 F surface temperature. The temperature decrease of the surface was fairly constant with time from 1500 F to about 900 F (~ 20 sec) at which point there occurred a rapid temperature decrease (~ 6 sec) to about 250 F. The flow rate change









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was negligible when the tube was open to the atmosphere while the pressure variation was a rapid 10 psi \pm 10 psig. The temperature at which the switch from film boiling took place was about as predicted, but the resistance to flow through the test section from steam generation was less than expected.

Critical Flow of Steam-Water Mixture. Analysis of preliminary experimental data for the critical flow of high pressure steam-water mixtures was performed. The purpose of this analysis was to test the validity of the various critical flow models proposed in literature by determining the correspondence between theory and experiments performed at high pressures. In these experiments critical flow data were obtained with a short piece of pipe (L/D = 20) at upstream pressures approaching 2000 psia. Inlet enthalpies of 420, 460, 480 and 525 Btu/1b were considered. The experimental data demonstrated unexpected results. As long as a two-phase mixture entered the test pipe, correspondence between the data and the more advanced theories existed. For example, the Fauske theory(1) predicted the critical mass velocity with 5% accuracy at a critical pressure of 600 psi and stagnation enthalpy of 525 But/lb. The Levy model⁽²⁾ was observed to predict mass velocities within 10% at these conditions. When compressed water entered the tube and was required to flash to a steamwater mixture within the length of the pipe, the location of the primary choking was observed to occur at the entrance of the pipe rather than at the exit where it is normally assumed to exist. During the existence of upstream choking, the pressure at the entrance of the pipe was observed to remain at the saturated value regardless of flow rate. This behavior is similar to that observed in short tubes at low pressures and temperatures (HW-77594) and is suggestive of the metastable flow system observed in that investigation. Normally, a pipe with an L/D ratio of 20 is not considered a short tube and such behavior was not expected. A report summarizing these experiments and results is in preparation.

(2) S. Levy, "Prediction of Two-Phase Critical Flow Rate," GEAP-4395, USAEC Research and Development Report, October 1963.



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⁽¹⁾ H. Fauske, "Contribution to the Theory of Two-Phase, One Component Critical Flow," ANL-6633, USAEC Research and Development Report, TID-4500, 18th Ed., October 1962.

Present Reactor Studies. During laboratory studies with an electrically-heated, full-scale model of a charge of K-V selfsupported fuel elements in a smooth-bore Zircalcy tube, it was observed that the pressure drop across the tube would increase without reason. Pressure drop measurements were taken for the first 23 feet of test section and compared with measurements for the last five feet of test section. From this comparison it was learned that the downstream pressure drop was larger than the upstream drop. Since the best explanation for this difference in pressure drop would be a partial plug in the downstream annulus, the downstream hydraulic head was removed for examination of the test section. It was found that the annulus was partly plugged with broken ends of the ceramic pieces used to support the electrically-heated fuel model in process tube. This condition also existed at the thermocouple flange located five feet upstream.

The broken ceramic pieces were removed and the test section reassembled. The annulus plugged again as soon as flow was established. Since it was then apparent that the ends of the support pieces would continue to break off and plug the annulus, the test section was removed and disassembled and all ends of the supports were broken off. The test section was reassembled and installed in the low pressure heat transfer facility for further testing.

6. Shielding Studies

N-Reactor Shield Evaluation. During January installation of the N-Reactor shield evaluation instrumentation was completed, and an initial experiment was performed with the small core loading. For the first test setup, two ion chambers and two foil holders were inserted in the bottom of the thimble. Gamma ray fluxes were measured and the gold and cadmium-covered gold foils received sufficient activation to allow counting. These foils were exposed for approximately 25 kilowatt minutes. Test setup No. 2 consisted of three ion chambers and six foil holders which filled the entire thimble from bottom to top. Gamma ray measurements were taken for this setup. However, foil activation was not obtained because of fuel irradiation restrictions during this part of the small core physics test.

Test setup No. 3 was a test run especially to allow irradiation of the shield plug thimble and was conducted directly after core neutron spectrum measurements. The evacuated belium³ detector was placed in the thimble bottom, irradiated at 5 KW (small core power) for four minutes, and background was counted. The detector was then removed from the thimble and filled with belium³ at proper



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pressure. While the helium³ detector was being filled, the lithium⁶ detector was operated for 10 minutes at a reactor power level of 5 Kw at the bottom of the shield plug thimble. Then, test setup No. 3 was inserted. This consisted of the filled helium³ detector (at the bottom of the thimble), three ion chambers and five foil holders. The entire test setup was irradiated at 5 Kw for 30 minutes. Preliminary analysis of the data indicates that significant activation of the gold and cadmium-covered gold occurred for a distance of approximately 25 inches up from the bottom of the thimble. Sufficient flux was available at the bottom of the thimble to satisfactorily operate the helium³ and lithium⁶ detectors. At the end of each of these tests the foils were removed and counted. A total of 18 gamma measurements and 22 foil activation measurements have been made to date.

Preliminary analysis of these data indicates a high thermal spectrum in the shield plug thimble for the small core configuration. Extrapolating these preliminary data to the full core configuration, it is indicated that for comparable power densities, the full core will produce measurable flux levels in the shield plug thimble and through the first few inches of the inner shield regions.

The data obtained from small core tests are currently being analyzed in detail. Full core zero power physics tests are expected approximately the second week in February. A test report to cover shielding irradiation measurements during power ascension tests is being prepared.

MAC Code Development. A formal report has been prepared and submitted for publication giving an expanded and corrected 31-group cross section library for MAC code and also giving a 15-group library which will enable users to make calculations in about half the computer time with little sacrifice in accuracy.

7. Graphite Studies

Oxygen Bomb Calorimeter. Calibration and modification of the calorimetry equipment is continuing. A multiple-deflection device was installed on the galvanometer, increasing its sensitivity by a factor of 4. The new stirrer assembly has been installed and is working satisfactorily. Measurement of heat transfer coefficients, between the calorimeter and water jacket, and heat generation rates due to stirring of the calorimeter water are in progress.



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B. WEAPONS - 03 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.

C. REACTOR DEVELOPMENT - 04 PROGRAM

1. Plutonium Recycle Program

Fuels Development

Fuel Element Refurbishing. Design criteria for underwater fuel element inspection and refurbishing equipment were prepared. This equipment is to be installed in the proposed addition to the PRTR basin.

Fuel Element Rejuvenation. The rejuvenation fuel element successfully completed its third cycle of irradiation, after operating with a maximum surface heat flux of $375,000 \text{ Btu/(hr)(ft}^2)$.

Fuel Element Development. Assembly of an eddy current detector fuel element for monitoring vibration in EDEL-1 was completed. The twelve outside fuel rods each contain an eddy current coil surrounded by ZrO₂ in a matrix of vibrationally compacted UO₂. X-ray examination of the completed rods shows minor migration of UO₂ into the insulating ZrO₂ medium. This may affect optimum irradiation exposure if the element is later charged into the PRTR. Following assembly, the element will be charged into EDEL-1 for a period of definitive tests to correlate rod motion with observed fretting.

PRTR Fuel Fabrication. A new vibrational compaction facility was put into operation to fabricate fuel elements for the PRTR. The new facility is in the south line in Room 138, 308 Building; the north line was previously set up to fabricate EBWR elements. The line is capable of compacting two fuel rods at a time on a 30 to 45-minute cycle; actual vibration times are 15 to 20 minutes. The rods are compacted to 87% TD. Five vibrationally compacted clusters were fabricated for the PRTR, two of the clusters utilized a new end closure which eliminated the crevice between the cap and can. (Postirradiation examination of vibrationally compacted fuel elements has shown corrosion attack in the crevice region on some fuel rods.)



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Three swage compacted fuel elements were fabricated for the PRTR. Two of the elements were made with new, as-received tubing (0.030" wall instead of 0.035") which permitted 2% more fuel to be loaded into each rod. In addition, the starting tubing length was increased; this permits more variation in the distribution of particle size in the fuel which, in turn, permits more economic utilization of the fuel material.

Elevated temperature tests in the fuel outgassing facility permitted an increase in the minimum temperature to 500 C, with no significant increase in the total outgassing cycle time. The higher temperature will be more effective in removing volatile impurities in the fuel.

A review of the present, over-al PRTR Vipac process is nearly complete. The essential materials receiving and final assembly operations are under study. The study is complete enough at this time to indicate some initial action plans for instituting an improved, formal quality control plan.

During the last month and a half, 1200 pounds of uranium dioxide were prepared and canned for densification by high energy rate impaction.

Weld closures were made on 20 Zircaloy-clad, Physics elements for experiments in N-Reactor.

The core material for the 36-inch PCTR rods has been densified by impaction.

Swageable type end closures (no crevice between end caps and cladding) were made on 20 PRTR $\frac{1}{2}$ -inch diameter vibrationally compacted fuel rods.

Examination of PRTR Fuel Elements. During approximately two-year storage in the PRTR Basin an irradiated (600 MWD/T) UO₂ fuel element developed a longitudinal cladding split. No defect was discernible during examination immediately after the element was discharged as a leaker in October 1961. The $\frac{1}{4}$ -inch wide, 12-inch long split is located approximately 18 inches from the bottom of an outer fuel rod. Unexpected oxidation and subsequent swelling of the fuel had occurred.

The possibility of catastrophic defect propagation in irradiated UO_2 fuel elements must be considered in establishing time limits for underwater storage of elements awaiting reprocessing or re-irradiation.

Anticipated cladding failures by massive zirconium hydriding occurred in four UO₂-PuO₂, PRTR fuel rods. In each case the failure was predicted from suspicion of oil contamination of the fuel during

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fabrication. Three of the clad failures occurred in UO_2 -1 wt% PuO₂ fuel elements that were fabricated just prior to discovery of an oil leak in a piece of fuel processing equipment. A special irradiation test of these elements led to failures of three fuel rods which supplied strong evidence that oil contamination was the cause of the recent series of PRTR fuel element failures. Further evidence was gained from the fourth predicted rod failure when a UO_2 -0.48 wt% PuO₂ rod containing fuel which also passed through the defective process equipment failed after 400 MWD/T exposure.

Al-Pu Element Post-Failure Irradiation Performance. The satisfactory behavior of a failed, Zircaloy-clad, nonbonded Al-Pu spike fuel element operating under power reactor conditions was demonstrated. An Al-Pu spike enrichment fuel element (#5088) suspected of failure during earlier operation in the PRTR was irradiated 406 hours in the water cooled (250 C at 537 gm/cm²) Fuel Element Rupture Test Facility (FERTF). The exposure accumulated by the element during irradiation in the rupture loop was 0.047×10^{20} fissions/cm³, to complete a total of 0.32×10^{20} fissions/cm³. Preliminary post-irradiation examination revealed severely cracked and hydrided Zircaloy cladding in the failure region. However, only minor swelling occurred. Fuel alloy corrosion products were eroded and some evidence of localized intergranular attack of the fuel was noted. There were no indications of waterlogging in spite of the presence of water throughout the nonbonded, core-clad gap. Monitored activity releases indicate that approximately 4 mg of Pu or 0.1 cm³ of fuel alloy was released to the coolant during the loop test. Fission product ratios in the coolant showed the release mechanism to be predominantly fission recoil. A steady state activity release rate was attained during the early stages of loop irradiation and did not interfere with reactor operation.

Cladding Evaluation. Ultrasonic inspection of 0.505-inch ID Zr-2 tubes was initiated. These tubes are used to fabricate fuel rods for the PRTR by vibrational compaction. Approximately 20-25% of the 200 inspected tubes were rejected because of signals originating on the inner surfaces. Some of these reject indications appear to be caused by dirt or other foreign material on the inner surface. However, some rejects are valid. After cleaning, questionable tubes that still show defects will be sectioned for metallurgical examination.

Corrosion and Water Quality Studies

Heat Exchanger Tube Examination. A tube in the PRTR rupture monitor heat exchanger failed in November 1963. The heat exchanger tubes are 0.035-inch wall, 3/8-inch diameter, seamless 304 stainless steel. An eddy current probe was used internally on the failed tube to locate the area of failure. Four positive indications of ID defects were seen in a $l\frac{1}{2}$ -foot length of the tubing. This section of the tubing was removed from the heat exchanger.

Examination of the removed section of tubing showed the failure resulted from wear against a support plate. Approximately 22 mils of the wall on one side of the tube had been removed, and a split about $\frac{1}{h}$ -inch long was centered in this area.

The tubing was sectioned in the areas where the ID defects were indicated. The eddy current indications were caused by clusters of pits on the ID of the tube. The pits were of two types: (1) round pits about 15 mils in diameter and from 5 to 10 mils deep; and (2) elongated pits about 35 mils long, 2 to 5 mils wide, and 10 to 20 mils deep. No other nonuniform attack was seen on the tube wall.

Under the exposure conditions (< 150 F - 66 C - pH 10 deionized heavy water), pitting attack of the stainless steel heat exchanger would not be expected. The pitting may be related to inclusions in the tubing which were dissolved out. There is no relationship between the external erosion and the internal pitting.

Zirconium concentrations in the PRTR primary coolant were significantly above normal three days. Some fretting is possible; severe fretting is unlikely.

The laboratory for plutonium decontamination studies was completed, and tests of plutonium oxide dissolution were started.

Reactor Components Development

Shim Rod Development. Assembly of one second generation shim rod is complete and the assembly has been operated in the environmental mockup at 170 F for about 250 hours. The only difficulty encountered has been with sticking and binding of the lead screws and shim rod elements. This is attributed to (1) out-of-tolerance shim rod elements which cause binding of the lead screw under certain conditions, and (2) close fitting ball bearing lead screws which do not have a "tolerance" for foreign matter. Currently, the assembly is operating satisfactorily.



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caused fretting $\frac{1}{2}$ -mil deep. The fretting marks previously formed during the previous operating period by the fuel element without wedges were also $\frac{1}{2}$ -mil deep. New springs of stainless steel spring Fretting Corrosion Investigation. The EDEL-I Loop was operated 521 hours with a wedged-type fuel element "locked" in the pressure tube with carbon steel wedges. While removing the fuel element wedges were also <u>1</u>-mil deep. New springs of stainless steel sprir wire have been wound and tested under tension in an even prior to from the tube, it was found that the carbon steel springs used to Borescope examination revealed that the fuel pads opposite the wedges had hold the wedges tight had relaxed due to temperature. repeating the wedged fuel element experiment.

the fuel element when wedged was rechecked. No difference in dif-ferential pressure across the pressure tube could be found between The previously observed reduction of differential pressure across wedged and unwedged elements.

including scrams, power level changes, and changeovers of pump combinations. A strip chart recorder has been added to the instru-ment package to show vibration amplitude on a continuous basis. tions greater than the "normal" level of 1.2 mils amplitude (at the detector) occurred with any interruptions of reactor operation, С which time the signal from the vibration pickup failed. Vibra-tions greater than the "normal" level of 1.2 mils amplitude (at The "excessive vibration detector" which was installed on PRTR operated satisfactorily until Jan. 6, 1964, at Dec. 20, 1963,

The instrumented fuel element to be used for study of the relative motion of fuel element and pressure tube is being assembled. Calibration of the eddy current coils is in progress.

magnetic pump, an electromagnetic flowmeter and an in-line circula-Fuel Re-Use Study. Detail design has started on the liquid sodium loop for use in making engineering evaluations of the fuel re-use concept. Specifications were prepared and inquiries for purchase sent out for an oxygen control and indicating system, an electrotion heater.

Design Analysis

Additional analyses have power, energy release, and time constant in a prompt critical ex-<u>Nuclear Safety of Mixed FuO2 UO2 Fuels</u>. Additional analyses have been carried out to develop a reliable relationship between peak cursion, where the delayed feedback may be nonlinear. A satis-factory expression has been derived for a "long delay" model in which the feedback time constant is long relative to the prompt period. Results obtained are being compared with independent UNCLASSIFIED

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transient analyses of identical excursions carried out by analog and digital computer methods.

PRTR Increased Power Density Study. A preliminary assessment is being made of several possible means of increasing the PRTR power density at its present power level by reducing the fuel loading size. This would have the benefit of increasing the rate of accumulation of exposure and recycle of plutonium through the reactor and at the same time would permit fuel testing at more demanding conditions. For initial purposes, five cases of decreasing core size were considered. These involved reducing fuel loading from 85 tubes to 55 or 43 tubes, and shortening fuel from 88 inches to 60 or 48 inches. In the smallest core considered, peak heat flux would be ~ 800,000 Btu/hr-ft². Enrichment would probably be increased to about 1.5%. Changing of the fuel geometry has not yet been investigated.

Fuel Re-use. Further studies on the fuel re-use concept have been initiated. Previously the only thermal reactor type considered was a heavy water moderated type, so this new work will expand the previous work to include three additional reactor types. These include a boiling light water moderated reactor, a pressurized water reactor, and a simulates spectral shift reactor.

Other expansion in scope will include two cladding types, zirconium and stainless steel, and a specific power rating of 20 Mw/ton for all the reactor types. Previously, 10 Mw/ton was used.

The economic parameters to be used will be largely unchanged except where appropriate to allow for expansion to other reactor types.

Thermal Hydraulic Studies

Heat Transfer Conditions for Higher Power Densities in the PRTR. Five different cases of power density increase were studied for the PRTR as a means of achieving fuel exposure goals more rapidly and increasing the diversity of fuels testing programs. The cases involve decreasing the number of active fuel channels to as low as 43 and/or decreasing the active fuel length to as short as 48 inches while holding the total power level at 70 Mw.

The results of a thermal hydraulic analysis show that the power density in the core could reasonably be increased up to 3.6 times the present level without encountering flow instability and without relaxation of the boiling burnout safety limits. This would represent a peak heat flux of 801,000 Btu/hr-ft² at normal power level conditions.



In each of the cases studied, the following general bases were applied:

- 1. Total reactor power equals 70 MW.
- 2. Flux distribution will not change appreciatly:

Axial = 1.31 peak/avg Radial = 1.30 peak/avg.

- 3. Percent of total power appearing as heat flux from fuel surfaces equals 95.
- 4. Uniform distribution of fuel enrichment.
- 5. Present fuel element configuration 19-rod bundle.
- 6. Outlet temperature from maximum tube equals 540 F.

The analysis indicates the desirability of decreasing the normal outlet temperature to 522 F.

All cases studied which include a change of the number of active fuel channels would require a change of the flow monitor sensing elements and would require changing the flow restricting orifices which are presently upstream of the fuel elements. The cases involving shorter fuel lengths would require the fuel rods to be cut shorter and the fuel hanger rods to be adjusted to achieve optimum nuclear physics parameters.

Pressure Transients in the PRTR. A document is being written which presents a simplified thermodynamic analysis to be used to evaluate pressure transients for the PRTR primary system. The problem of particular interest is the failure of the reactor to scram when pumping power for the primary system is lost. In this case, as vapor is generated and pressure rises, a pressure trip is reached which scrams the reactor. The problem then is to determine the maximum pressure rise and determine the required safety relief capacity of the system.

Calculations have been completed for one phase of the problem which has not been previously considered during this transient. This is the problem of condensing vapor bubbles. As vapor is formed, the two-phase mixture is at saturation conditions but as the pressure rises, the saturation temperature rises and the exit fluid from the reactor then is subcooled. The vapor bubbles are initially superheated during this process but will cool and begin to condense. To account for this problem, an effective mean bubble lifetime of the generated vapor is considered. This is used to obtain an effective vapor generation rate.

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The solution to the equations describing this problem has been programmed for the computer and the effects of reactor inlet temperature and initial pressurizer gas volume were considered. The results of these calculations for initial conditions of 120 Mw, 9600 gpm, 1040 psia pressurizer pressure, 1165 psia scram pressure and 427 F inlet temperature were determined. Nonuniform power generation with a maximum-to-average power of 1.3 was used in all cases. The results indicate that the size of the relief valves would not be a problem since they would not even open at their set point of 1265 psia.

2. Plutonium Ceramic Fuels Research

Plutonium Ceramics Irradiation Program. A program is under way to study the irradiation effects upon plutonium ceramics and cermets. The materials under study are in the form of small, thin wafers to allow a high burnup rate to be attained at relatively low temperatures. The materials presently under irradiation are: PuN, PuC, PuO₂, β -Pu₂O₃, and PuN-W and PuN-Pu cermets. These materials were prepared by pneumatic impaction, arc-melting, and/or sintering.

One sample of PuC has already been examined out-of-reactor and samples of PuN and PuO₂ are scheduled into Radiometallurgy Operation during February 1964.

Irradiation of UN-20 wt% PuN Capsules. Three capsules of UN-20 wt% PuN solid solution pellets were fabricated. Powders of -325 mesh UN and PuN were blended, pelletized, and heated for five hours at 1800 C in 2/3 atmosphere of nitrogen. The product of this reaction was a solid solution of PuN in UN, as identified by x-ray diffraction. These pellets were then sintered, centerless ground, and loaded into stainless steel capsules with MgO guard pellets at each end. The stainless steel capsules with welded end caps were then shrunk-fit into heavy wall aluminum sleeves and a second set of end caps were welded on.

These capsules will be irradiated in the MTR at a capsule surface heat flux of 218 w/cm² (~ 700,000 Btu/(hr)(ft²)) which will give a central fuel temperature of about 1300 C.

Thorium Nitride. ThN-PuN combinations may be quite attractive as breeder fuels. Single-phase ThN was synthesized for use in preparation of (Th-Pu)N compounds for phase and irradiation studies. ThN itself is a refractory with almost unknown properties and will also be studied in the pure form. Thorium metal was nitrided by soaking -100 mesh powder at 900 C under 2/3 atmosphere of pre-purified

nitrogen. This treatment resulted in Th₂N₃ which was subsequently decomposed under vacuum at 1750 C to form ThN. The lattice parameter of the single-phase, cubic ThN is 5.159 ± 0.001 A.

Thorium Cermets. Four thorium metal-plutonium oxide cermets were fabricated by the Nupac process. The cermets consisted of Th-25 wt% PuO₂, Th-50 wt% PuO₂, Th-25 wt% β -Pu₂O₃. The samples will be used for compatibility and volatility experiments.

Metallography. The primary goal in activating the metallography facility is capability of producing quality photomicrographs of alpha active samples. The equipment has been designed so that samples may be examined within the glove box through a cover glass, or outside of the glove box as specified by a supplemental Radiation Work Procedure.

Photomicrographs were made of UO2-cermet samples with the recently installed Bausch and Lomb research metallograph. Tests were made of different contamination barriers with several combinations of illumination and magnification. Samples were compared and photographed bare, through a cover glass, and through a thin plastic coating as described by Bierlein and Miller in HW-79845.

Brew Furnace. All equipment necessary for operating the Brew furnace in flowing hydrogen has been installed and trial runs will be made in the very near future. Flow rates of 250 cubic feet per hour at temperatures of 2800 C are anticipated.

Plutonium Research Facilities. The new plutonium research facilities in the 308 Building are approximately 15% complete. The new facilities include five nitrogen-atmosphere plutonium glove boxes and additional x-ray diffraction equipment. Equipment being placed in the glove boxes will permit the measurement of electrical conductivity, thermoelectric power, thermionic emission, thermal expansion, vapor pressure, and thermal diffusivity of plutonium-bearing materials. Capabilities for thermogravimetric and differential thermal analyses are also included.

3. Ceramic (Uranium) Fuels Research

Fuel Element Rejuvenation. Successful operation of a twicerejuvenated, Zircaloy-clad UO2 fuel element at a surface heat

flux of 375,000 $Btu/(hr)(ft^2)$ further demonstrated the feasibility of extending fuel element life (and thus reducing over-all cladding costs) without utilizing burnable poisons or without resorting to special Phoenix fuel combinations. The element was rejuvenated by inserting new, enriched UO₂ into the central cavity and remotely rewelding. The irradiated element will be returned to Hanford (from MTR) for destructive examination.

Transmission Microscopy of Single Crystal UO₂. Single crystal UO₂ (0.002 cm thick) containing metallic uranium has been examined by optical transmission microscopy using infrared film at temperatures to 700 C. The uranium inclusions exhibited no discernible change in structure after being heated in vacuum to 1000 C. A decrease in the optical transmission (between 0.65 and 0.9 μ) with increasing temperature is attributed to a shift to longer wavelengths of the absorption band.

<u>ThO2</u> Irradiation Studies. A deliberate defected thoria element was irradiated in the MTR GEH-4 Facility until loop activity exceeded operating safety limits. A 0.0025 cm (0.010 inch) hole was drilled in the side of an aluminum clad, vibrationally compacted thoria element. The element was irradiated in the MTR GEH-4 Loop at full power for a period of 2.5 hours. At the end of this period the loop coolant activity was 400,000 $\frac{dis.-cc}{min.}$ of loop coolant. Reactor

power was reduced and a decrease in activity noted. Upon return to full power for a 20-minute period, the activity level returned to that previously recorded. At the end of the additional 20 minutes of operation at full power, the reactor was shut down and the test element discharged. A nondefected thoria element was charged for irradiation during the remainder of the cycle.

Two other thoria elements were returned to Hanford for destructive examination.

Irradiation of Uranium Monosulfide. A small uranium monosulfide sample has been discharged from ETR after 154.5 effective full power days with an estimated exposure of 3.5×10^{20} fission/cm³ (12,000 MND/ton_{II}, 1.44 at% burnup).

Fission Product Migration. Preliminary results indicate Pu and some fission products relocate in UO_2 under an applied potential gradient. A potential gradient of 1 v/cm was applied to an indirectly heated, irradiated UO_2 sample. The sample was held at 1800 C for 20 hours. Initial experiments were run in reducing atmosphere. The isotopic distribution was normalized to the uranium present in each section.



Experiments to eliminate the possibility of uranium diffusion in a substoichiometric sample are in progress.

Thermionic Emission. A number of components for the thermionic emission experiments have arrived and construction of the apparatus has started. The necessary instrumentation is near completion.

Fabrication of Cermets. Hardware was fabricated for pneumatic impaction of 80 wt% UO₂-Mo into molybdenum tubing. Loading of a stainless steel clad, 80 vol% UO₂-stainless steel, four-rod cluster, complete with impacted stainless steel end caps and spacing brackets was begun.

4. Basic Swelling Studies

Irradiation Program

Two irradiation testing capsules which were previously brought to operating conditions at 625 C (1157 F) and 425 C (797 F), respectively, are operating satisfactorily.

According to present plans two more irradiation tests will be conducted during this fiscal year. One capsule will be charged in April or May 1964, and will contain high purity uranium specimens of varying geometries. It will be for the purpose of observing what effect varying the specimen shape and cross section has on the nature and amount of intercrystalline distortion and/or fission gas accumulation. The other irradiation test which is planned for charging some time in June will involve a 70 Kg/cm² high pressure capsule for which the design was just completed.

A study is currently being conducted by instrument development personnel on a solid state temperature controller which is under consideration as a replacement for the controllers currently in use at the reactor. Solid state devices offer a high level reliability unobtainable in components of other types. Reliability has been the major problem with the units now in service.

Second Phase Distribution in Dilute Alloys of Uranium

Of the 30 dilute alloyed dingot specimens included in the study which features three levels of alloy content at each of 10 different heat treatments, nine have been replicated and photographed in the electron microscope. Particle size data obtained from the Zeiss Particle Size Analyzer have been processed in accordance with

quantitative metallographic techniques on five of the samples in order to obtain information on the density of particles of the dispersed phase and their size distribution. The statistical calculations show that the population density and average particle size of the second phase are significantly affected by both composition and heat treatment.

Preliminary evaluation of the quantitative metallographic results reveals some interesting phenomena. A specimen in the alpha-rolled condition, containing 150 ppm Fe and 100 ppm Si, had, initially, a high concentration of particles in the 0.1 to 0.4 micron range. A solution anneal in the high alpha range at 643 C (1190 F) for seven hours followed by a water quench reduced the small particle concentration by a factor of three and slightly increased the number of particles in the 1.0 micron range. An additional treatment of one hour in the medium alpha range at 590 C (1094 F) resulted in a slight increase of 0.1 micron particles. A rough draft of an interim report on this portion of the program has been written and is undergoing revision to be issued as HW-80349.

5. Irradiation Damage to Reactor Metals

Alloy Selection

Two capsules containing refractory metal tensile specimens irradiated in the ETR G-6 cold water facility were opened for examination. The TZM, Mo, and W-25 Re specimens were corroded to the extent that identification of the specimens was possible only through weight measurements. Similar specimens of Cb and Ta alloys showed no signs of corrosion.

A temperature effect was observed in the tensile properties of Inconel 625 and Hastelloy N. These specimens were irradiated at 50 C (122 F) and 280 C (536 F) to an exposure of about 1 x 10^{20} nvt. Room temperature tensile data for Inconel 625 indicated that the 280 C (536 F) irradiation caused an increase in yield strength of about 35% with no charge in total elongation. For the 50 C (122 F) irradiation, a 58% increase in yield strength was observed with a marked decrease in total elongation.

Hastelloy N specimens irradiated at 280 C (536 F) showed a 25% increase in yield strength compared to a 57% increase for the 50 C (122 F) irradiation. The 50 C (122 F) irradiation also caused a large decrease in total elongation as compared to the 280 C (536 F) irradiation.



In-Reactor Measurement of Mechanical Properties

The final in-reactor creep test on 20% cold worked Zircaloy-2 is now in progress. This test is being conducted at 20,000 psi stress and 377 C (711 F). After 2300 hours of operation, the creep rate appears to be 1.9 x $10^{-6}/hr$.

A heating element capable of producing specimen temperatures of 2090 C (3800 F) for short periods of time and 1648 C (3000 F) for sustained periods of time has been developed for possible use in high temperature in-reactor testing. The heating element is 0.080-inch diameter molybdenum wire, noninductively wound. Power is supplied to the element through coaxial water cooled copper leads from a D.C. source. The element draws approximately 130 amps at 17 volts for a specimen temperature of 1648 C (3000 F). In tests, the element was fixed in a three-inch diameter, 12-inch long stainless steel cylinder which was immersed in a water bath.

At the present time there is one capsule with a 20% cold worked Zircaloy-2 specimen and three capsules with annealed 304 stainless steel specimens ready to be charged into the reactor.

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 is being given to the determination of mechanical property changes produced in metals by irradiation at elevated temperatures.

Tensile tests were performed in AIST 348 and 304 type stainless steel specimens which were irradiated at temperatures between 282 and 300 C (540 and 572 F) in the ETR G-7 hot water loop. Specimens which had received integrated neutron exposures of 6.0 x 10^{19} nvt and 1.5 x 10^{20} nvt (fast) were tested at room temperature and at 300 C (572 F). In previous tests performed at 300 C (572 F), crosshead motion alone was used as a measure of elongation. Recently, an extensometer was developed for use at temperatures up to 300 C (572 F). Tests were performed at 300 C (572 F), utilizing the extensometer on identical specimens from the same quadrants which were previously tested at 300 C (572 F) without an extensometer. As was the case in the room temperature tests, the extensometer was removed shortly after reaching 0.2% offset and the remainder of the elongation was measured by crosshead motion. Tests conducted at room temperature, both with and without an extensometer, indicate that

the greater portion of the error incurred is introduced during the initial loading up to the 0.2% offset. Hence, the use of crosshead motion as a measure of elongation in the plastic range is acceptable for tests conducted at room temperature.

The ductility of these materials as indicated by total elongation was found to be 50% less at 300 C (572 F) than at room temperature when an extensometer was used for measuring elongation. However, without an extensometer, tests at 300 C (572 F) showed the same apparent elongation as was observed at room temperature. This relative error was large because of the rather low values (6-8%)being measured. These values, however, are typical of irradiated materials. This clearly points out the virtue of using high temperature extensometry even on a relatively hard-beam machine such as the Instron.

Both the irradiated and control tensile and 50-hour stress-torupture tests will be performed on Instron tensile machines suitably adapted for testing at 1292 F (700 C). The necessary preliminary equipment for this adaptation is either being designed or assembled, and includes a hinged three-element furnace and temperature control equipment for use in both types of tests.

Damage Mechanisms

The objective of this program is to determine the mechanism by which defects produced by neutron bombardment interact with dislocations to modify the plastic deformation characteristics of metals. The investigation is currently concerned with the role of interstitial impurities in alpha-ircn.

Tests were run on Ferrovac "E" to determine the temperature dependence of the lower yield point, activation energies, and the effect of strain rate at 240 K (-28 F). One test was also run on a zone-refined iron sample at 294 K (70 F).

Initial analysis of these results indicates the primary effect of impurities is to modify the temperature dependence of \checkmark *, the effective stress on the dislocations. The profile of the barrier controlling plastic flow is not changed, but the barrier height for a given temperature of deformation is slightly lower for the purer material. An interpretation of the mechanism responsible for this effect is being developed.

Eighteen iron specimens have been received after irradiation to an exposure of about 1 x 10^{18} $\rm nvt_{\odot}$



Environmental Effects

Corrosion specimens of 348 stainless steel exposed 19 days in the ETR-G7 loop, cycle 52, have recently been descaled. Samples were exposed to a flux level of 9.4 x 10^{13} nv fast, at water temperatures of 280 C (536 F) and pH 9 (LiOH). Weight losses on the five descaled irradiated specimens ranged from 21.5 mg/dm² to 29.1 mg/dm² with an average of 25.0 mg/dm². Metal losses for five descaled ex-reactor specimens ranged from 32.6 mg/dm² to 37.1 mg/dm² with an average of 35.3 mg/dm². It is not known whether this difference is due to neutron irradiation or a difference in water chemistry between the ETR loop and the ex-reactor loop. The ETR loop water contains 0.8 ppm of oxygen compared to <50 ppb of oxygen in the ex-reactor loop. Oxide losses to the testing environment are similar for both the irradiated and nonirradiated samples.

ATR Gas Loop Studies

High Temperature Pyrometer. The Milletron recording pyrometer has been received and is in the process of being checked out. This is a two-color pyrometer which is usable over the range of 800 to 3000 C (1472 to 5432 F). The sensing head can be mounted on a tripod, or a fixed stand. The indicator and recorder are mounted in a separate portable cabinet.

Primary Piping. The welded two-inch schedule 40 Hastelloy X and Haynes 25 piping has passed inspection and is being sent to a subcontractor for bending to final dimensions. The pipe is expected to be ready for final inspection the first week in February 1964, and delivery will be about March 1, 1964.

The backup order of seamless Haynes 25 piping was ready for final inspection on January 20, 1964. Delivery of this pipe should be early in February 1964.

Design Test 1172. Thermal and bending tests of Grayloc connections for the ATR loop were terminated after 188 cycles. No helium leakage was detected during the testing. The report on these tests should be complete February 1, 1964.

Corrosion of High Temperature Alleys. A test was conducted to determine the corrosion resistance of various superalloys in wet helium at 1700 F (927 C). Coupons were abraded on 3/0 paper, degreased, and exposed to wet helium at 1700 F for three test periods of 100 hours each. The partial pressure of water vapor in the

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helium atmosphere was maintained at 15 ± 2 torr. After each test period the samples were weighed on a microbalance, and some samples removed for metallography. In order of decreasing oxidation resistance at 300 hours, the alloys tested were Hastelloy N (0.3 mg/cm²), Hastelloy X-280, Hastelloy C, Inconel 600, Haynes 25, Inconel 625, Incoloy 800, Inconel 718, Inconel 702, and R-235 (3.6 mg/cm²).

Samples of superalloy weldments Hastelloy X-Hastelloy X (Hastelloy X filler wire), Haynes 25-Haynes 25 (Haynes 25 filler wire), and Hastelloy X-Haynes 25 (Hastelloy X filler wire) were exposed to flowing He + $0.5\% O_2 + 0.54\% N_2$ at one atmosphere and 1200 C (2192 F) for seven days. Welds were made on one-fourth-inch plate, which was machined to a thickness of one-tenth-inch and abraded on 240-X emery prior to corrosion testing. (A description of the welding procedure may be found in a document entitled 'Dissimilar Metal Welds of Certain Superalloys and Stainless Steels," R. L. Knecht, HW-78574, August 1963.)

The Hastelloy X material retained its metallic integrity and was covered with a tenacious oxide film. The weld zone appears as oxidation resistant as the parent metal. The Haynes 25 material was in various stages of degradation, ranging from severe penetration to complete consumption, whether welded to Hastelloy X or to itself. When welded to Hastelloy X, the weld zone appears to have corrosion tendencies roughly intermediate between Haynes 25 and Hastelloy X.

The test proved so severe that only qualitative observations can be made. The oxidation resistance of Haynes 25 will determine the oxidation resistance of a Haynes 25 Hastelloy X couple.

<u>Gas Analysis</u>. The gas chromatograph under development by Chemical Metallurgy Operation has been moved from the 326 Building and installed in its permanent location in the 314 Building. The automatic attenuator integrator has been connected and the analysis system is performing very well for H₂, O₂, N₂, CH₄, and CO contamination in He over the range of 1 ppm to 1% of any, or all, components. Reproducibility of standard samples is \pm 1% or better. Detailed calibrations are due to start immediately with the exception of the low ppm range which must await the availability of ultra pure helium. Parts are on order to add a silica gel column for CO₂ analysis.

Dynamic Materials Testing Apparatus. Babcock and Wilcox ran a test of the loop on January 3, 1964, at a temperature of 2500 F and the loop operated satisfactorily. When the loop was restarted on January 7, 1964, it was noted that one thermocouple had failed. B & W requested permission to move this thermocouple to a more



suitable location and the re-test was scheduled for January 13, 1964. No further word has been received as to whether the final performance test has been made.

Neutron Dosimetry

Instruments are being tested to measure fast-neutron spectra. Two types of systems are being studied, one using the $He^3(n,p)$ reaction and another the $Li(n,\alpha)$ reaction. In each case the charged particles produced by the reaction cause ionization in solid-state detectors which is proportional to the energy of the incident neutron plus the energy released by the reaction. When developed, these systems will provide a means to monitor the energy spectra of neutrons with energy exceeding about 0.5 mev. Such a capability is valuable in madiation-damage studies where irradiations are performed in facilities for which it is not possible or practical to calculate the spectra. It also will provide a means to verify calculated spectra.

The detectors have recently been calibrated at several energies using monoenergetic beams generated by the positive-ion accelerator. The systems are currently being used to monitor neutron spectra in the N-Reactor core during the physics startup tests.

Neutron-Flux Monitoring

The degree of consistency to be expected in flux monitoring with threshold monitors was estimated from results obtained in the ETR G-6 SE facility. Integrated fluxes at various vertical positions were calculated using nickel and iron monitors for two separate irradiation periods, each comprising three cycles. For all vertical positions values of 80.2 mb for Ni and 61.9 mb for Fe were used for the effective cross sections. Corrections for the thermal burnout of Co-58 and Co-58m were made.

Intercomparison of the results from the long term monitors and flux values provided by ETR operations showed a maximum deviation of about 30% with most of the sets of data agreeing to within 15%.

6. Gas-Cooled Reactor Studies

<u>Gas-Graphite Reactions</u>. Systems involving carbon, carbon monoxide, and carbon dioxide were irradiated in sealed quartz capsules in a Hanford reactor at about 550 C. Changes in gas composition and graphite weight were measured, and the nature of deposited materials was examined.

The radiation-induced disproportionation of CO at pressures of 1.5 to 18 atm was measured. The loss of CO was accompanied by the formation of CO₂ and black solids which were deposited uniformly on the walls of the container. The yield of CO₂ was iependent upon the CO pressure. Electron micrographs of the deposits showed two distinct structures; one similar to carbon black and one believed to be polymerized carbon suboxide.

The radiation-induced reaction between graphite and CO₂ was measured at about 550 C in the pressure range 1.5 to 45 atm. Both graphite and CO₂ were consumed, resulting in the formation of CO. The ratio of the number of moles of CO₂ reacted to moles of CO formed was 0.86, in contrast to the value of 0.5 which would have been observed if the familiar reaction, $C + CO_2 = 2$ CO, were the only one occurring.

In the pressure range 1.5 to 61 atm and the temperature range 360 to 675 C there was negligible thermal reaction between graphite and carbon dioxide. In the temperature range from 1200 to 1400 C and the pressure range 17 to 19 atm the rate of reaction with CO_2 was rapid, and the reaction product was CO. The stoichiometry corresponded to the familiar equation, $C + CO_2 = 2$ CO. A report on this work, HW-67241, has been issued.

Oxidation of Large Graphite Samples by Water Vapor. Work has been continued to determine the effect of diffusion when large graphite bars oxidize in flowing helium containing small quantities of water vapor. It was previously reported (HW-79726 A) that the rate expression

Rate
$$(hr^{-1}) = 91.8e^{-33.2} \times 10^{3}/RT$$

applied to the reaction of a TSX graphite sample exposed to 7000 ppm water vapor in helium at 725 to 813 C. The graphite sample was 12 inches long, 0.5 inch in diameter, and weighed about 65 g. It was also pointed out that the activation energy of 33.2 kcal/mole was lower than expected for a sample of this size.

The rate measurements have now been extended to 857 C on this same sample and a revised rate expression,

Rate
$$(hr^{-1}) = 4.51 \times 10^6 e^{-54.9} \times 10^{3} / R_{\perp}^{2}$$

has been calculated. This in much better accord with results obtained on samples 1.5 and 2 inches in diameter, as the revised activation energy is, as would be expected, greater than that found for the latter samples, 48.5 kcal/mole. The low activation energy obtained

earlier for the 0.5-inch sample is due to an anomalously low value for the rate measured at 813 C. The newly derived rate expression is in reasonable agreement with an expression calculated from data reported for an even smaller TSX graphite sample (August 1963), viz.,

Rate $(hr^{-1}) = 3.24 \times 10^7 e^{-59.2} \times 10^3 / RT$

Here, as expected, the activation energy is even higher, although oxidation rates in the same temperature range, 725 to 860 C, are in good agreement with those for the 0.5-inch diameter sample.

Rate measurements will be extended to TSX graphite samples, 0.75, 1.0 and 3.5 inches in diameter.

7. Boronated-Graphite Studies

Boronated Graphite Irradiations. The first long-term irradiation capsule continues to operate satisfactorily. The temperatures have increased somewhat during the past month but are well within the acceptable tolerance limit of \pm 150 F. The two 1000 F (nominal) sections are operating at 915 and 900 F, respectively, and the maximum variation in the monitored samples is 3 F. The 650 F (nominal) section is operating at 725 F and the maximum variation of sample temperature is 27 F.

The pressure of the stagnant helium in the capsule continues to decline slowly, indicating a small leak. The leak rate has been estimated at 5-10 cm³/day (STP). A leak of this magnitude is inconsequential with respect to the irradiation part of the experiment but may cause difficulty in monitoring the rate of helium formation from the (n, α) reaction.

Photomicrographic studies of the boronated graphite have provided useful information concerning the structural differences of the nominally 5 and 7 wt% "grey" and "black" materials. The photomicrographs reveal the following information on structure:

- 5 wt% Black similar to artificial polycrystalline graphite.
- 5 wt% Grey modified, nonhomogeneous structure. Some areas are unaffected; other areas partially recrystallized.
- 7 wt% Black modified, nonhomogeneous structure. Some areas are unaffected; other areas partially recrystallized.
7 wt% Grey - almost totally recrystallized structure. No evidence of random orientation within the recrystallized particles although the particles are randomly orientated with respect to each other. The number and size of B₄C particles are reduced. The boron is believed to remain in the recrystallized structure to a great extent.

Machined parts for the second long-term boronated graphite capsule were ordered during the month and have been partially completed.

A sample loading for the 2-C-2 capsule very similar to that of Capsule 2-C-1 is being considered. Preparation of the samples is to begin soon and pre-irradiation measurements will be taken as soon as the samples are completed.

During the past month a series of tests was started in order to determine the effects on the heat transfer characteristics of a nonconcentric annulus between a hot and cold body in line contact.

8. Aluminum Corrosion and Alloy Development

<u>C-1 Loop</u>. The first corrosion test of aluminum-clad fuel elements was started in the C-1 Loop. The test charge consists of a train of eight fuel elements with Al-Pu cores clad in X-8001, A-288, and KYZ aluminum (all Fe-Ni alloys). The temperature of the upstream element is being measured by a thermocouple inserted in its core. The test is being conducted at a bulk outlet temperature of 290 C and at neutral pH.

9. Metallic Fuel Development

Irradiation of Thorium-Uranium Fuel Elements. The irradiation of three tubular Zircaloy-2 clad thorium - 2.5 wt% uranium fuel elements continued in the ETR P-7 loop. At the end of the fifth cycle of irradiation the elements had achieved an integrated exposure of 1.5×10^{20} fissions/cm³ (4200 MWD/T). During the last irradiation cycle the fuel elements operated at a maximum core temperature of 540 C, surface heat flux of 65 cal/sec-cm² (8.7 x 10⁵ Btu/hr-ft²), and a specific power of 60 watts/gm (183 kw/ft).

Thorium Fuel Defect Testing. In an attempt to index the effect of heat treatment upon bond trength a peal test is being developed. Tooling which will permit the specimen to rotate during pealing of the Zircaloy-2 cladding was designed, built, and tested during the



month. The tooling is used in conjunction with the 60,000-pound Baldwin Universal Testing Machine. Specimens are being prepared by machining circumferential grooves through the cladding and splitting the resulting bands longitudinally. One end of the band is bent upward to provide a tab. The specimen holder permits rotation as the tab is being pulled. Preliminary data show a force of 275-300 pounds per inch of tab width is required to peal the cladding as extruded.

Thorium-Uranium Alloys. The fabrication, irradiation and defect corrosion behavior of the Th - 2.5 wt% U - 1.0 wt% Zr alloy has been described. The effects of higher zirconium and uranium compositions on the structure, fabrication and defect corrosion behavior of thorium base alloy fuels have not been thoroughly studied. A program has been initiated to study these effects.

Nine additional alloys have been double vacuum arc melted into seven 8-inch long x 2.9-inch diameter ingots weighing approximately 25 lbs each. Five Brinell hardness readings were taken along the length of each double melted ingot and compared with those taken on the single melted ingots.

Very little difference in hardness was noted between first and second melting. Side-wall quality of the double melted ingots was far superior to the single melted ingots. These ingots have been machined for copper canning and will be extruded to billet stock from which Zr-2 clad rods will be made for defect corrosion testing.

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

Thermal Hydraulic Studies

Analysis was continued of the two-phase pressure drop experiments performed to determine the pressure loss behavior of various piping components at typical reactor operating conditions. Those components which were examined include 2 x 3-inch and $1\frac{1}{2}$ x 2-inch pipe expansions, contractions of 3 x 2-inch, 2 x $1\frac{1}{2}$ -inch, and 2 x 1-inch pipe, 2-inch valves of gate and globe design and a 1.551-inch diameter orifice in a 2-inch pipe. These experiments were run at 1200 psia and covered qualities up to 24%. Other experiments, performed with a straight pipe section and a 3-inch radius bend, provided data on the pressure parameter effect at pressures of 800, 1200, and 1600 psia.



The ratio of the two-phase pressure loss to the single phase liquid pressure loss was calculated for the various components and plotted as a function of quality with flow as a parameter. The expansion and contraction sections were formed by abrupt diameter changes, so they may be considered a sudden expansion and contraction. The single phase pressure losses for these sections were compared with theoretical values, and it was found that the expansion losses were an average of 82% of the calculated values, while the contraction losses ranged from 37 to 100% greater than calculated. The two-phase to single-phase pressure loss ratios for the 2 x $1\frac{1}{2}$ -inch contraction and the $1\frac{1}{2}$ x 2-inch and 2 x 3-inch expansions all coincide closely with the ratio for straight pipe throughout the quality range and were insensitive to flow in this range. The ratio for the 2 x 1-inch contraction ran below the straight pipe ratio at the lower qualities and above at qualities greater than 13%. The 3 x 2-inch contraction had ratios which were quite scattered, due primarily to the lack of fully developed flow at the inlet to the contraction.

The experimental two-phase to single-phase ratios for straight pipe at 800, 1200 and 1600 psia were compared to ratios from the Martinelli-Nelson correlation, the Bankroff variable density model and the Levy momentum exchange model. The best agreement throughout the pressure range was with the momentum exchange model. At 1600 psia, Levy's model coincided with a smooth curve through the data; at 1200 psia, the model agreed within \pm 12%, while at 800 psia, the model was 7% low at 21% quality and a maximum of 19% low throughout the quality range.

Previously reported were correlations of the two-phase to singlephase ratio of various bends and a tee as functions of the straight pipe two-phase to single-phase ratio. These correlations were all based on data obtained at 1200 psia. Subsequently, the ratio for a three-inch radius bend was calculated from the data at 800 and 1600 psia and plotted against the corresponding straight pipe ratio at those pressures. For a straight pipe ratio greater than 1.5, all the data grouped in a band which included the earlier 1200 psia data. The band spread was small enough to allow a single correlation of the bend ratio to be satisfactory for the 800 to 1600 psia range. For example, at a straight pipe ratio of 2.5, the threeinch radius bend ratio at 1600 psia was 5.9; at 1200 psia, it was 6.1 and at 800 psia, it was 6.9. It is reasonable to assume that the correlations of the other bends would apply satisfactorily through the 800 to 1600 psia range.

A complete report on the two-phase pressure drop studies is being prepared.









11. Advanced Reactor Concept Studies

Fast Supercritical Pressure Power Reactor. Comments have been received on the rough draft of the report on this study and several revisions are being made in the report. Approximate cost estimates for power from 500 and 1000 Mwe reactor plants are being added. Revisions should be completed and the report submitted for publication at month end.

A-42

Military Compact Reactor Plutonium Studies. Additional studies on the effects of substituting plutonium for uranium in the Military Compact Reactor have been completed. Descriptions of the studies and the results obtained are being issued in an informal document as a supplement to HW-79449. The study included a review of previous calculations for plutonium-fueled MCR cores, recalculation of fuel temperatures and coolant pressure drops by more precise methods using FUGUE and STHTP computer programs, and analysis of a tungsten-reflected core to determine potential weight-saving advantages through reduction of shield weights. Calculations of reactivity changes due to burnup of the plutonium fuel for all cores considered were also repeated with allowance made for isotopic changes in the fuel during burnup.

Recalculation of the conditions for previously studied core cases indicated essential agreement with those previously calculated, except for the compact cores using plutonium fuels of advanced ("gridplate" and "honeycomb") design. For all cores less than 9 inches in diameter, a diameter increase of 0.2 inch was required to increase the fuel inventory sufficiently to compensate for the increased burnup reactivity requirements indicated by the revised calculations.

For the ceramic fueled compact "honeycomb" core (7.2 inches diameter), the use of physical properties of UN rather than UO_2 to represent the PuN fuel resulted in significant improvement in the indicated capabilities of this core. By assuming the higher thermal conductivity of UN to apply, an increase in coolant channel diameter was permitted, resulting in a lowering of the pressure drop across the core from 65 to 25 psi.

For the 7.7-inch cermet "gridplate" fueled core, early manual calculations had been based on a rough approximation to the fuel geometry, since time did not permit more detailed calculations. For the present analysis, a more precise model of fuel geometry was used for computer calculations. In order to maintain fuel temperatures sufficiently low, the use of smaller, more closely



spaced fuel channels was required, boosting the pressure drop from 35 to about 70 psi. For comparison, calculations were made assuming tungsten or molybdenum rather than niobium as a cermet base. Considerable improvement in thermal performance was indicated. No calculations were made of the core nuclear requirements for such fuels.

In studies of a tungsten-reflected reactor, a cermet-pin fueled concept with a core 10 inches in diameter and 10 inches in length was studied. This reactor had a 5-inch thick tungsten shield, as compared with the composite BeO-W shield assumed for the other cases studied. Total reactor weight, including shield for this concept, was slightly less than the smallest (7-inch) reactor previously analyzed. Weight saving was 34%, compared to the MCR reference design used. In order to provide sufficient control strength with the tungsten reflector, boron strips on the control drum were assumed to be backed with a 2-cm thick layer of metal hydride (e.g., YH) to augment the effectiveness of the poison.

To obtain a rough estimate of comparative costs for fueling the plutonium fueled cores and the reference uranium fueled MCR design, basic fuel material (U + Pu) costs were estimated for all cores, using standard AEC uranium price schedules and assuming a plutonium cost of \$10 per gram Pu-239. Fuel material costs were markedly less for all plutonium fueled cores; for the smaller cores, calculations indicated plutonium would be competitive with uranium, even if plutonium costs were taken at \$25 to \$40 per gram Pu-239.

Large Sodium Cooled Fast Reactor Study. The cross section set reported last month is still not satisfactory. Analyses made with this set show a strange behavior of k_{eff} when sodium is voided and/or transverse leakage corrections are applied. The problem arises from an attempt to use "corrected" magnesium scattering cross sections as a substitute for sodium in a fast neutron energy spectrum. The "corrected" magnesium cross section gives a poor approximation in Mev and Kev neutron energy range. Unfortunately, the sodium cross section cannot be corrected by hand because the magnesium causes a different energy spectrum, thereby affecting the effective cross sections of the other materials in the core. A special "physics chain" cross section data tape will be required with real sodium cross sections included. While this is in preparation, existing cross section sets will be used. Calculations have been started, using the 16-group Hansen-Roach cross section set. However, the number of materials available on the present set is somewhat limited.

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Hydride Control Studies. In order to provide supporting data for feasibility studies on the utilization of a metal hydride of variable composition for control of reactors, studies of the evolution and absorption of hydrogen by zirconium hydride were performed by Chemical Metallurgy Operation. Hydrides with compositions from ZrH_{0.27} to ZrH_{1.3} were studied in the preliminary investigation recently completed. Absorption-desorption tests in an isothermal constant-volume closed system at 750 C showed that 90% completion, based on saturation values, was obtained for hydrogen absorption in from 3 to 24 minutes, and for desorption in from 2 to 8 minutes. The preliminary data indicate that (1) significant changes in hydrogen content can be achieved in relatively short times, and (2) the changes in hydrogen content are not so rapid as to preclude the overriding of resulting reactivity changes by other means. On the basis of the preliminary data, the use of a variable-composition hydride for control purposes appears feasible from a standpoint of thermal stability.

Sonar Power Systems. A review of energy storage and conversion methods is being made to permit a better preliminary evaluation of the most promising novel approaches to a complete system. Further study of the applicability of radioisotope heat sources for this service indicates that supply limitations would probably restrict the use of such systems to moderate power low duty cycle applications. Consideration is being given to possible experiments which could be performed in the thermal hydraulies laboratory on means of generating sound in water.

<u>Computer Codes for Parametric Studies</u>. The current parametric studies on the Military Compact Reactor utilized two computer codes for the evaluation of thermal hydraulic performance and fuel temperatures. Although this was useful in obtaining consistently accurate results, the time consumed in calculating and keypunching the input information was excessive. As a result of this, work was begun on outlining the amount of work necessary to simplify the input procedure, particularly for the parametric or scoping type of studies. The codes proposed for use in these studies are:

STHTP - Heat conduction code.

- FUGUE Thermal hydraulics code for liquid coolants (boiling and nonboiling).
- MAGNON Thermal hydraulics code for compressible coolants.



The STHTP and FUGUE codes were used in the MCR study. The MAGNON code has not yet been completely written or debugged. The following is a brief outline of code modifications which are being initiated:

- (1) FUGUE (and most likely MAGNON) must have the input changed to readily accept multiple cases with changes in flow channel dimensions. STHTP has this feature now and this technique will be used for the other two.
- (2) Mathematical models of various fuel configurations must be selected for analysis and input calculation procedures, necessary for each code, must be set up.
 As a result of the MCR study, input calculation procedures for three models have been set up. These are;
 - (a) Square channel gridplate fuel
 - (b) Round channel honeycomb fuel
 - (c) Pin-type fuel.

In addition, the following three configurations may be added later:

- (a) Hexagonal channel gridplate or honeycomb fuel
- (b) Plate-type fuel
- (c) Concentric ring fuel such as ANP used.

These modes are adequate for any parametric studies to be run in the near future.

(3) Each of the above calculation procedures can then be programmed as subroutines to the appropriate codes. By programming these as input subroutines, it is easy to add the calculations for additional fuel configurations, which will most certainly arise.

Hydride Moderator. Rates of absorption and desorption in the zirconium hydrogen system at 750 C (1382 F) have been measured over the composition range $\text{ZrH}_{0.2}$ to $\text{ZrH}_{1.18}$ in a closed system where no hydrogen was admitted or vented during the experiment. In six absorption experiments the time for 90% saturation to take place varied from three minutes to thirty minutes (16, 10, 3, 8, 30, 20 minutes for $\text{ZrH}_{0.40}$, $\text{ZrH}_{0.48}$, $\text{ZrH}_{0.69}$, $\text{ZrH}_{0.85}$, $\text{ZrH}_{1.07}$, and $\text{ZrH}_{1.18}$, respectively). The amount of scatter in the time to achieve 90% of saturation is probably due to oxygen contamination. In the six desorption

D. DIVISION OF RESEARCH - 05 PROGRAM

1. Radiation Effects on Metals

This program is directed toward 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. Present studies involve single and polycrystalline specimens of molybdenum, nickel, and rhenium.

Quenching studies are being conducted on nickel specimens representing three levels of purity (99.997%, 99.98% and 99.4%) in order to characterize the nature of vacancy impucity atom interactions. The specimens are in the form of foils 0.75 mm thick and are quenched from 1675 K (2552 F), or 0.97 T_m , at a rate of > 10⁴ ^OK/second. Tensile tests have been completed on foils quenched and slowly cooled from 1675 K, and other quenched foils have been aged for 10⁵ seconds at temperatures of 500 K and 700 K (437 F and 797 F) prior to testing. Nickel specimens of 99.4% purity did not exhibit any difference in yield stress between quenched and slowly cooled specimens, indicating that the quenchedin vacancies are immobilized by the high concentration of impurity atoms. This lack of hardening is also taken as good evidence that hardening due to quenching strains is minimal. Quenching of the higher purity specimens produced slight increases in yield stress. the magnitude of the increases being 3.1 and 11.4% for 99.98 and 99.997% pure materials, respectively. The 99.997% nickel specimens also exhibited a distinct increase in the rate of strain hardening. One quenched 99.997% Ni specimen has been shed for 10^5 seconds at 500 K (437 F) and tested. It had an increase in yield stress of 42.9% over the yield stress in the slowly cooled condition. Further testing is in progress. A guanched 99,997% nickel foil has been successfully electro-thinned and examined by the transmission technique in the electron microscope. No loops, tetrahedra, or clustered defects were observed; the dislocation density was found to be low, similar to that observed in foils annealed two hours at 700 C (1292 F),

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Molybdenum foils deformed in tension to strains of 1, 2, 5, and 20.5% have been precharacterized by x-ray techniques and encapsulated. These specimens will be irradiated to two levels of exposure, 5×10^{18} and 5×10^{19} nvt (E > 1 Mev). The goal of this experiment is an understanding of the effect of dislocation density on the distribution of point defects produced by irradiation.

Studies of x-ray line broadening in irradiated molybdenum foils are continuing. Two orders of the (100) reflection, i.e., (200) and (400), have been analyzed in the case of a high purity (< 10 ppm carbon) foil irradiated to 10^{19} nvt (E > 1 Mev). The results show that there is no particle size broadening after irradiation, all the broadening being the result of lattice distortions. Preliminary results obtained with a similar foil irradiated to 7×10^{18} nvt indicate a lower value of strain due to lattice displacements; the magnitude of the increase in strain at the higher exposure is roughly proportional to the increase in exposure.

Strain rate cycling experiments with polycrystalline molybdenum are continuing. Tests conducted at room temperature with basic strain rates of 1.67×10^{-5} and 8.33×10^{-5} sec⁻¹ and a strain rate change factor of 10 have revealed that the effective activation volume for thermally activated deformation is of the order 10^{-19} cm³. This is an exceptionally high value when compared with results obtained for iron, niobium, and tantalum. However, since the stacking fault energy of molybdenum is believed to be appreciably greater than those of Fe, Nb, and Ta, the observed value may be compatible with the mechanism of cross-slip of screw dislocations. Such a conclusion is, of course, highly tentative, and much more experimental work is required. Attempts to conduct tensile tests on polycrystalline molybdenum at 195 and 177 K (-108 and -321 F) were unsuccessful even at a strain rate of $1.67 \times 10^{-5} \text{ sec}^{-1}$, because of extreme brittleness of the specimens; tests conducted in iced brine at 255 K (O F) have proved successful in preliminary experiments. In order to extend these tests to lower temperatures, it will be necessary to test single crystal specimens.

A molybdenum single crystal tensile specimen containing approximately 450 ppm carbon was deformed in tension after an exposure of 10^{19} nvt (E > 1 Mev), and then oriented for sectioning parallel to (111) planes. Sections cut from the grip ends disclosed defect structures identical to those observed in undeformed thin foils; only a limited number of dislocations were seen. Channels were not observed in this region of the crystal, showing that very little deformation had

taken place therein. Distinct channeling was observed, however in the gage section of the crystal specimen.

Application of the wafering technique has been extended to sectioning high purity polycrystalline iron specimens. Wafers 3 mm in diameter and 0.6 mm thick were sectioned from a Ferrovac-E specimen with a silica wheel and thinned chemically in a bath composed of two parts HaPOL and one part HoOo. Final polishing was performed in a perchloric-acetic acid electrolyte at a potential of 18 volts. Specimens prepared in this manner are ring-shaped, with an edge thickness of 0.4 mm. When the specimen is placed in the gap of the split objective lens in the Philips EM-200 electron microscope, it is situated in a magnetic field with an intensity of the order 1000 Oersteds. Since that portion of the sample under observation is not directly supported, some deformation takes place as the sample is inserted in the microscope and is translated through the magnetic field of the lens. The extent of deformation introduced in the microscope is being investigated.

An article entitled "Dislocation Channeling in Irradiated Molybdenum" was published in the December 1963 issue of Journal of Applied Physics; one of the illustrations was used on the cover of the journal.

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.

An experimental study of the creep characteristics of the stable phases of plutonium and deformation during phase transformation under constant compressive stress is in progress. The steady state creep rates of the delta, gamma, and beta phases have been found to be related to the applied stress and temperature by the proportionality

$$\frac{1}{\epsilon} \alpha \ (e^{-Q/RT})(\sigma^k)$$

Q is the activation energy, T is the absolute temperature, σ is the stress, R is the universal gas constant, and k is a constant having

the value of 5 for pure metals. Values of 30 Kcal/mole for Q and 5.5 for k were experimentally determined for steady state creep of the beta and delta phases of plutonium. This proportionality is not entirely valid for steady state creep of the gamma phase because the value of k varies from 2.7 to 7 at strain rates of 0.01%/hr to 10%/hr, respectively; Q was determined to be 24 Kcal/mole.

High purity electro-refined plutonium has been received from the Los Alamos Scientific Laboratories for basic research studies and for comparison with selected high quality as reduced and vacuum cast metal. Metallographic observation revealed that both types of metal have very few inclusions and approximately the same amount and distribution of microcracks. The kinetics of the beta to alpha transformation of the two metals are comparable below 75 C (167 F) with the difference in reaction rates being within experimental error. However, the beta to alpha reaction rate of the electro-refined metal is greater than that of the as-reduced metal above 75 C. For example, the incubation times for initiation of transformation for the electro-refined metal and the as-reduced metal are 800 and 4000 seconds, respectively, at 89 C (192.2 F).

The high purity LASL plutonium has been evaluated from the standpoint of its use in rolling and deformation studies. In the asreceived condition it is available as cast bars essentially 1.3 cm in diameter. Such stock is not adaptable to rolling and must be recast into thin sheets. Since it is highly desirable to maintain the initial purity of this material, preparations have been made to cast in place using high density magnesium oxide molds. Such procedure will not only minimize the amount of material required but will also eliminate the contamination potential which arises in the crucible during the normal casting process.

Metallographic examination and density measurements of the asreceived material indicate, on the assumption of high purity, that microcracking is a significant problem. Since it is reasonable to assume that these internal flaws will influence the rolling behavior, microcracking must be minimized.

A bright field electrolytic etchant for unalloyed plutonium consisting of water, ethyl alcohol, and nitric acid reported by Battelle has been under investigation for use in optical and electron microscope studies. Various concentrations and electropolishing conditions have been utilized. Grain boundaries have been revealed, but only under conditions for which an oxide layer also forms on the

specimen. This etchant has revealed much twinning in plutonium which had been beth plustically colormed prior to a slow beth to alpha transformation rate. This twinning has not been revealed by any other etchant. Some grain boundary relief has been observed. Microcracks are appreciably attacked by this etchant.

A report, HW-80464, entitled "The Rolling of Alpha Plutonium," by F. E. Bowman and V. C. Asmund is currently in preparation.

E. CUSTOMER WORK

1. Radiometallurgy Laboratory

Examinations and Measurements

Routine examinations and measurements are or will be reported as part of the sponsoring research and development programs.

Metallography

Samples	Processed	111
Photomosaics		18
Autoradiographs		11

Chemistry

Burnup dissolutions	42
Decladding dissolutions	 l
Rare gas collections	6
Vacuum induction furnace runs	27
Defilming for film analysis	6

Physical and Mechanical Properties Testing

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Tensile Tests (rm. temp.)	98
Rockwell hardness tests	20
X-ray analyses	17

General

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Negatives processed 388

Physical and Mechanical Property Testing Cell Project CGH-857

Dilatometer and Annealing Furnace. Repair of the annealing furnace has been delayed by parts shortages. Shipment was rescheduled for this month. All parts for repair of the dilatometer were received and installation is in progress.

Equipment

Microhardness Blister for I Cell. The quotation received from a commercial vendor on a basic microhardness tester with a rectilinear stage was much higher than expected. Additional information is being sought to guide future action.

Metallograph Camera. Preliminary design was completed on a system which will automate a series of five repetitive, sequential steps that are followed during the preparation of each micrograph used in construction of a photomosaic. The purpose of this system is to reduce operator fatigue and associated human errors.

High Temperature Tensile Testing Machine. Most of the major components have been received and are being mocked up for final testing and training of operators.

Stereo Zoom Hot Cell Macroscope. Final adjustment of the stereo zoom macroscope is still in progress at the vendor's plant. New objective end prisms were installed and adjustments made which improved the quality of the transmitted image.

Remote Induction Furnace. Modifications were made on the remote induction furnace and associated equipment was installed in "B" Cell so that an irradiated N-Reactor inner fuel element could be heated to 980 C. One irradiated element was successfully heated to the required temperature at three places along the length of the element. Equipment modifications are being made to improve the temperature control and permit colored moving pictures to be taken during the heating operation.

2. Metallography Laboratories

During the report month 409 samples were processed, a total of 552 macrographs and micrographs taken, 1452 negatives printed, and 6024 prints processed.

Routine Metallography Laboratories activities will be reported as part of the sponsoring research and development component's work;

however, items of unusual interest or representing departures from routine operations will be reported here.

Examination of a Zircaloy-2 - titanium weld zone revealed needlelike particles resembling hydride in the Zircaloy adjacent to the weld. More extensive examination, however, revealed that this needle-like phase, which was oriented nearly perpendicular to the weld interface, was not zirconium hydride but was apparently subgrains of Zircaloy which had picked up, by diffusion, enough titanium to alter their etching characteristics.

Replicas from two metallographically polished 304 stainless steel specimens were examined in the electron microscope. An electrolytic polish-etch technique was used to prepare the surface of one specimen. The second specimen surface was cathodically vacuum etched. It was found that particles having diameters less than 0.5 micron were easily hidden by shadows from the chemically roughened regions in grains of the electrolytically etched specimen. Quantitative analysis for size and distribution of particles, to include those less than 0.5 micron in diameter, cannot, therefore, be accomplished using this technique. Particles having diameters less than 0.5 micron were located in grains of the cathodically vacuum etched specimen, but with some difficulty. The matrix of the specimen is preferentially attacked during cathodic etching and, generally speaking, the particles rise above the surface of the sample. The one-step, or negative replica, technique transforms the protruding particles on the specimen into depressions in the plastic replica. Vaporized heavy metal. deposited onto the plastic from an acute angle, characterizes the particles by subtle shadows in the depressions.

3. High Temperature Lattice Test Reactor

HTLTR Vault Thickness Calculations. Revised core and shield region criteria were established to allow shield thickness calculations for the HTLTR vault. These data have been forwarded to Vitro Engineering for their analysis and a MAC calculation utilizing these input data is presently being programmed here.

High Temperature Lattice Test Reactor Prototypes. Detail design of all phases of the HTLTR prototype are 99% complete. Fabrication work being performed by J. A. Jones was halted on January 10 due to lack of funds. The status of some of the main items of the construction effort are:

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- (1) Fabrication of the steel containment shell is 65% complete. The shell is now ready for the thermal insulation to be installed.
- (2) Concrete footing for the shell is 100% complete.
- (3) Graphite fabrication which includes the core and keys, core supporting timbers, recuperator and the bus bars is 94% complete.
- (4) Electrical work is 30% complete.
- (5) Fabrication of the main heat exchanger is 100% complete.

Most of the materials required for completion of the prototype are now on site.

The heater testing mockup was heated to temperatures up to 1020 C. During the heating up period the mockup was evacuated to a pressure of about 2.5 in Hg absolute to remove moisture from the various components. Approximately three gallons of water have been removed from the mockup to date.

Operation of the mockup was terminated on January 17, 1964, due to the failure of the heating element. This failure was attributed to air leaking through the sight tube during the vacuum condition, thereby oxidizing the heating element. The heating element has been replaced and the mockup is now ready to be put back in operation.

All fabrication work has ceased on both safety and control rods. The stepping motor and controls for use in both assemblies has been ordered. The drawings of the driving head prototypes are complete.

4. Assembly of Elements for N-Reactor Physics Tests

Special N-fuel assemblies constructed to contain flux monitoring pins and foils were assembled for use in N-Reactor physics tests. Two different assemblies were made, one for a cold water test and the other for use at the normal N-Reactor operating coolant pressure and temperature. The cold test elements were clad in unbonded 1100 aluminum with welded end closures. The elements for the hot test were prepared from regular N elements which were altered by drilling in the ends to a depth of four inches to accommodate uranium rods containing monitoring pins. The end closures consisted of unbrazed Zircaloy-2 rings welded in place.



5. EBWR Fuel Elements

EBWR Plutonium Fuel Element Fabrication. More than 400 EBWR-size fuel rods, containing Nupac UO2-1.5 wt% PuO2, have been fabricated for the PRCF, 300 of them by means of the resonant beam vibratory compaction technique. Effort is continuing on an enlarged scale to determine and isolate the variables encountered in all steps of the process. Future tests will be supplemented with quality control measures to increase the acceptance rate of vibrationally compacted fuel rods, as well as decrease the amount of scatter in data obtained during initial fabrication studies.

Cladding Procurement. An additional 554 EBWR cladding tubes were received from the manufacturer. A total of 2593 are on site. Of these, 1243 remain to be tested. Testing is currently delayed by examination of tubing used for fabrication of PRTR fuel rods by vibrational compaction.

Irradiation Testing Program for EBWR Prototypic Fuel Rods. Ten capsules (GEH-14-517 through 526) containing vibrationally compacted UO₂-1.5 wt% PuO₂ were inserted into the MTR on January 13, 1964. Three additional capsules (GEH-14-527 through 529) were shipped to the MTR on January 17, 1964, for insertion into the MTR in February. The capsules employ near-EBWR-size Zircaloy tubing and contain impacted oxide fuel which was vacuum outgassed at 600-700 C prior to vibrational compaction. Average bulk density of capsule fuel was 82.3% TD (range 79.1-84.3% TD). Carbon analysis of the capsule fuel indicated 103 and 106 ppm.

Two capsules containing vibrationally compacted UO₂-2.5 wt% PuO₂ are to be discharged from the ETR on March 2, 1964. Estimated exposure at discharge may be slightly greater than the goal of 1.43 x 10^{20} fissions/cm³ (5000 MWD/ton of fuel). One capsule (GEH-14-421) contains impacted fuel and one capsule (GEH-14-424) contains a physical mixture of UO₂ and PuO₂.

A mockup of a 42-rod cluster for irradiating EBWR fuel rods in PRTR (16 replaceable rods) was assembled and provided to the Thermal Hydraulics Operation for hydraulic flow testing. The tests, conducted with cold (12 C) water, indicated a pressure drop of 2.88 g/cm² (5.9 psi) at a flow rate of 6.3 liter/sec (100 gpm), which corresponds closely to the value obtained with a standard PRTR cluster under near-similar conditions. The flow test durmy will be modified to provide an articulated element comprising 17-rod assemblies. In the modified design 24 rods (12 in each assembly) would be replaceable

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and the position of the assemblies (one above the other) will be reversible to achieve more uniform burnup.

assembly will be prepared with EBWR prototypic fuel rods and irradiated in the PRTR. 34-rod ы A 42-rod

EBWR Tube Volumes. To determine if difficulties encountered during preliminary vibratory compaction of EBWR fuel rods were related to tube was immersed in water. Contrary to previous indications, in-ternal diameter was found to be closely related to tube volume were within an acceptable range and it is not likely, at least for the lot of tubes studied, that volume variations are sufficiently when an average tube diameter was employed. The volumes obtained excessive variations in tube volumes, the volume of each of 40 tubes was determined by means of the weight change when a closed large to cause fabrication difficulties. immersed in water.

start-- HOO that some lots of material produced acceptable elements in shorter time periods. Because of this variation, six 45-kilogram lots of Characterization of "Nupac" EBWR UO2-PuO2. During vibrational con paction studies of EBWR fuel elements containing high energy rate pneumatically impacted ("Nupac") UO2-1 wt% PuO2, it was observed Fuel element fabrication histories were better for the three lots that showed better particle bonding, highest densities, and 0/U ratios closest to the value of the material were examined. ing material.

By using controlled times, it Seven lots of prepared materials were tested for uniformity of pre-Sieve Analysis of EBWR UO2-PuO2. Vibrational compaction by a hori-zontal resonant beam is being used to produce EBWR fuel alguents. **Createst** variation was found in the coarse (..6+10) mesh. By using amounts of material on the screens and sufficient sieving pared composition of mesh fractions by sieve analysis. was possible to prepare reproducible mesh compositions.

after surprisingly little particle breakup, i.e., less than 1% of the -6+10 mesh fraction and no change in the -325 mesh fraction. This suggests relatively little particle movement in a horizontal system, as opposed to a vertical vibrational compaction system. showed formulated to coarse, medium, and fine particul size distribution. This composition was compacted in a relatively short time. A One unsuccessfully compacted composition was screened and closely similar composition of the high density material, not thoroughly blended after formulation, was not successfully computed, even 40 minutes of vibration. A sieve analysis of thus material show

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6. NASA Fuel Development

Tungsten Honeycomb Fuel. The fourth of a series of tungstenmolybdenum extrusion experiments designed to establish conditions for tungsten-tungsten bonding has been extruded. The billet consisted of seven 0.045" thick parallel tungsten plates equally spaced in 0.060" deep notches inside a 0.125" wall, 1.875" OD tungsten sleeve. The spaces between the plates were filled with machined molybdenum sheet and the entire assembly vacuum canned in a 0.200" wall pressed and sintered molybdenum can. The billet was preheated to 2000 C (a 200 C differential was noted between front and rear of the billet, with the rear at 1800 C) and extruded at 60 in./min ram speed and 9:1 ratio. No lubricant other than water dag in the container was used. The die was ZrO coated with a 90° cone and had been used for one previous extrusion. Visual examination shows an excellent surface on the extrusion, good surface on the tungsten tube with no indication of lack of bonding between the tube and plates. The plates are somewhat wrinkled in the transverse direction but otherwise uniform in thickness, spacing and straightness. The severe wrinkling is probably due to the coarse grained structure of the molybdenum filler pieces.

Tungsten-Uranium Coprecipitation. A program has been initiated to demonstrate the feasibility of fabricating a cermet fuel material containing submicron size UO₂ particles uniformly dispersed in a tungsten matrix. The work is being done under the sponsorship of Lewis Research Center, NASA. The approach under investigation is the coprecipitation of tungsten and uranium from solution to produce a matrix containing 10, 20, and 30 vol% UO₂. Initial development work using uranyl nitrate, UNH, and ammonium paratungstate, APT, as starting materials has revealed the following problems:

- 1. The ammonium paratungstate has a relatively low solubility requiring large volumes of solution in the initial stages of the process. A more soluble tungstate, ammonium metatungstate, has been procured from Sylvania, Chemical and Metallurgical Division.
- 2. The insoluble uranyl tungstate, UO₂WO₄, is less dense than the precipitated tungsten resulting in partial separation during centrifuging. Vacuum filtration and thickening of the slurry prior to separation are under investigation.
- 3. Optimum pH value has not been established. Basically, tungsten is soluble in basic and acidic solutions while uranium is the reverse. Solubility of pH data is being obtained.



Examination of Irradiated Cermet Fuel Plates. Examination of a NASA-supplied UO2-tungsten cermet plate after irradiation in the ETR was completed. The plate remained chemically and dimensionally stable. Upon opening the NASA-constructed test capsule, the outer molybdenum box and the structural and supporting members were intact. Disassembly of the nested boxes revealed that the two inner tungsten boxes and their support wires had become broken during or subsequent to the irradiation. The fuel plate was intact, but the temperature monitoring wires were missing. Thus, the plate temperatures actually attained during irradiation could only be inferred from other data. Appearance of the plate indicates that it operated at a temperature greater than 3000 C during the irradiation. No significant quantities of fuel were lost from the plate during irradiation, although a few particles of UO2 had been vaporized or otherwise lost from near the surface. There was no evidence of gross swelling or other distortion, nor was there any evidence of chemical reaction between the UO2 and tungsten.

Cermet Joining and Fuel Element Fabrication Development. Pneumatic impaction bonding of tungsten clad, tungsten-UO₂ cermet plates is a new technique that was used for fabricating a small square honeycomb grid section. As presently developed, the joints between the tungsten-UO₂ plates are filled with packed tungsten powder and formed into a solid bond by pneumatic impaction. Deformation of the plates is prevented by mild steel mandrels which are removed by chemical techniques after impaction. The tungsten clad, tungsten-UO₂ cermet plates, 0.029-inch thick, were supplied by NASA. The use of pure tungsten powder in the joints would eliminate possible "hot spots". However, tungsten-UO₂ cermet powder could be used for the joint if desired.

Cermet Fuel Development. Two tungsten foil clad, 80 vol% W-UO2 cermet plates were fabricated by pneumatic impaction. The UO2 and tungsten particle sizes were less than 1 micron. The thin, approximately 0.025-inch plates were used in studies concerning UO2 loss during thermal cycling at high temperatures.

A vibrational compacting technique was employed for loading 80 vol% W-UO2 powder grid assemblies prior to densification by pneumatic impaction. Initial packed density of greater than 60% TD was obtained. Two 1/8-inch hexagonal honeycomb grids, 2 inches in diameter and 2 inches in length, were fabricated by this process. The 1/8-inch hexagonal mild steel bars are held at both ends with hexagonal broached plates in order to maintain alignment during pneumatic impaction. After impaction, the mild steel mandrels were removed by chemical techniques. Good grid dimensions were obtained.





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<u>Magnetic ForceWelding</u>. A bond was made on the longitudinal joint of a l-inch wide section of a 2-inch OD x 0.020 wall tungsten cylinder (supplied by NASA). To obtain more test pieces for this experiment, one of two 2-inch wide sections supplied by NASA was cut in half. The first section cracked excessively during the weld operation; bonding was obtained with the second section. The second 2-inch long piece was to be cut in half and used for weld parameter studies, but it broke into small pieces during the cutting operation and could not be used. This tungsten seemed to be considerably more brittle than tungsten-UO₂ cermet used in other welding studies.

Vibration Testing of Cermets. Work has been initiated on vibration testing of $W-UO_2$ cermets. An electrodynamic vibrator capable of thrusts up to 50 lbs and frequencies up to 10,000 cps is being employed for these tests. To date, two cermet grids have been vibrated at room temperatures to failure for the purpose of testing the feasibility of the concept. These tests indicate the possibility of microcrack formation in the tungsten matrix during vibration.

A system is now being fabricated which will enable vibration testing to be carried out at elevated temperatures. In addition, tests are now being planned to simultaneously subject specimens to thermal cycling and vibration.

Spalling Studies on W-UO2 Cermets. Spalling of W-UO2 cermets in a plasma is being investigated. The samples were thermal cycled in an H₂-Ar induction plasma. Samples containing small UO₂ particles showed greater mechanical stability and lower fuel loss than samples with larger UO₂ particles.

Cermet Fuel Loss Studies. An intensive investigation of the accelerated fuel loss from cermet fuels, resulting from thermal cycling has shown:

- (a) a decrease in UO₂ particle size significantly reduces fuel loss;
- (b) accelerated UO₂ loss from thermal cycling results from the mechanical and chemical instability of the metal matrix and cermet cladding, i.e., the thermal expansion differences; and
- (c) fuel loss is a function of atmosphere and atmosphere purity, being affected by hydrogen, water vapor or oxygen, and matrix metal surface purity.

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The results of this study have led to a very significant improvement in the fuel loss characteristics of cermet materials.

Manager Reactor and Fuels Laboratory

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PHYSICS AND INSTRUMENTS LABORATORY

MONTHLY REPORT

JANUARY 1964

FISSIONABLE MATERIALS - 02 PROGRAM

REACTOR

N-Reactor Lattice Parameter and Spectral Measurement Tests at Startup

N-lattice parameters to be determined include p, C_0 , ϵ , the neutron temperature, and the r-value of the epithermal flux. The spatial and energy dependence of the flux in the concentric tube fuel is to be determined using Pu-Al, U-235-Al, fuel enrichment uranium, depleted uranium, Lu, Eu, Au, and Cu pins, both bare and cadmium covered.

The irradiation of the specially instrumented fuel elements in the cold small N pile, and subsequent disassembly and pin counting has been completed. A simultaneous irradiation in the PCTR provided monitor pins of each type irradiated in a highly thermal spectrum and these were counted along with the above pins to monitor decay and any possible counter drift. All data have been processed using computer program APDAC and initial reduction is started.

The fuel elements required for the hot test have been welded and are presently being autoclaved. Other arrangements for the hot test are nearly complete.

NPR Utilization Studies

Utilization studies of the NPR have continued during the past report period. To test the analytical methods employed, temperature coefficient calculations for a reference NPR lattice have been carried out and are being analyzed.

Other

See Reactor Theory and Code Development sections under Plutonium Recycle Program and also EXPERIMENTAL REACTOR PHYSICS FACILITIES section.

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Instrumentation

Satisfactory performance of N Reactor physics test instrumentation has been achieved. All instrumentation and special mechanical devices were installed and made operational prior to the initial fuel loading. A portion of the system was designed, procured, and installed in cooperation with Instrument and Electrical Development, IPD. Engineering assistance was provided on a shift basis. No significant instrumentation failures occurred to impede the progress of the tests.

Testing was continued on the fission counters for use in the N Reactor low level flux monitoring channels. The flux monitoring system is being reviewed, tested, and modified as necessary.

Evaluation of the N Reactor fuel rupture monitoring system has continued with testing at KE Reactor of one production model gamma spectrometer. Using natural uranium in the loop water, tests were made to determine the general system capabilities and necessary operational adjustments. Complete test data will be presented in a report prepared by the cooperating group--Control, Instruments, and Systems Analysis, NRD.

Tests at the PRTR fuels test loop in which a Pu-Al alloy element having a defective cladding was purposely installed in the loop revealed a lOO-fold increase in delayed neutron activity over the clean loop condition. The gamma spectrum analysis instrumentation indicated a general factor of 10 increase in signal up to about 4 MeV. In general, the instrumentation is functioning satisfactorily although difficulties with commercial solid state amplifiers have been noted. Intermittent troubles with the paper tape system seem to have been satisfactorily resolved. The only major instrument failure to date has been a high voltage power supply.

System Studies

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N Reactor steam generator simulation studies are being extended to include the analysis of major systems tied together. First efforts will be to combine the secondary loop flow simulation with the steam generator simulation for study on the new computer. Process control equations and simulation diagrams for this purpose have been completed. A list of nomenclature, constants, units, and descriptions is approximately 80% complete.

A revised N Reactor primary system simulation was started which includes a three-delayed group reactor kinetics model with fuel, water, and graphite heat transfer and reactivity effects, the primary size of the steam generator, a two-group primary pump and hydraulic system simulation, and the pressurizer. Also, the primary coolant volume contraction resulting from

scrams and power level changes was included.

A new nomenclature list, which was compiled for the over-all N Reactor simulation, removes ambiguous subscripts and makes the symbols used in equations and simulation diagrams easier to identify. The new list was compiled for punching on IBM cards to facilitate updating the information and printing of new nomenclature lists.

The N Reactor pressurizer simulation was simplified for inclusion in the over-all system simulation and test runs were made to determine the performance. The check runs compared favorably with the more complex model.

A revised N Reactor primary flow system simulation was tested for correct functioning of the pump drop-off circuit and was found to be satisfactory. Also, several repeat runs were made to determine the effect of an error found in the computation of the primary loop inertial values. The effect was noticeable but not sufficient to invalidate information gained in previous runs. Estimates of good controller settings for the primary flow systems remained the same.

The N Reactor control system simulator is in the process of fabrication. All significant material is on site or on promise of early delivery. Completion of the number of controllers required for the secondary system simulation run is expected early in February.

MIDAS programs being written for simulation study of N plant reactor kinetics, primary coolant system, steam generators, and secondary coolant loop are about 70 percent complete.

Two noise tests were made on the N Reactor. The first test consisted of a 15-minute recording at low power levels, the second a two-hour recording at approximately ten times the power level of the first test. The power density spectrum of the output of a neutron detector is being analyzed to determine pertinent characteristics of the process.

A simulation is being prepared to study the N Reactor system characteristics when cold water is injected into the reactor. The simulation diagram is complete except for determination of the appropriate constants.

SEPARATIONS

Critical Experiments with PuO2-Plastic Mixtures

Further critical mass experiments were conducted with PuO₂-polystyrene compacts and the Remote Split-Table Machine. Seven critical core configurations consisting of rectangular prisms were assembled during the month. The plutonium concentration of the core material was $1.12 \text{ g/cc} (2.2\% \text{ Fu}^{240})$, with a H/Pu atomic ratio of ~15. Various reflector combinations involving Lucite were used on the assemblies, one of which consisted of a rectangular prism wrapped with cadmium sheet and reflected with Lucite, to determine the effect of the cadmium on reflector savings. Comparative data were obtained for the effect on criticality of complex reflectors consisting of steel plate and Lucite. The various critical assemblies and experimental results are discussed in more detail below.

In order to reduce the data from the critical experiments to an equivalent "clean" system, it is necessary to apply corrections for the effects of the control and safety rod channels on criticality. To obtain additional data for evaluating these effects, experiments were performed with a rectangular parallelopiped of 6-in. x 7-in. cross sectional dimensions for which previous criticality information was available. The basic difference in the current experiments was that the control and safety rods were of the reflector removal type rather than of the fuel removal type previously used. The critical length of the parallelopiped when reflected with 6-in. thick Lucite on all surfaces was 22.06-in.; the critical mass was 17.1 Kg Pu, which is 1.44 Kg less than the value obtained previously with the fuel removal type of rods. With the two ends of the assembly unreflected, the critical length was 26.0-in. and the critical mass was 20.3 Kg Pu, or about 0.9 Kg less than previously measured. These differences, which show the cumulative effect of the control and safety rod channels, are comprised of several different effects: Displacement of fuel by the channel; neutron leakage along the channel; neutron absorption in the steel channel and its drive mechanism; and decreased reflection due to the void in the reflector through which the drive mechanism operates.

Critical data were obtained for a rectangular prism of 12-in. x 12-in. base dimensions which was fully wrapped with 0.03-in. cadmium sheet and reflected on all surfaces with 6-in. thick Lucite. The critical core height was about 9-in., and the critical mass under these conditions 23.7 Kg Pu. In the absence of the cadmium sheet, the critical mass for an assembly with the same base dimensions would be about half of this value or ~13 Kg Pu, which indicates the effectiveness of the cadmium. The purpose of this experiment was to determine the effect of cadmium in reducing the reflector savings of the Lucite, or other hydrogenous (water) reflectors.

Additional data were obtained with complex reflectors for evaluating the effect on criticality when neutron poison sheets or absorbing materials are interposed between the reactor core and its reflector (Lucite in this case). Various reflector combinations were used on one end of the assembly (a rectangular prism of cross sectional dimensions 12-in. x 12-in.) which was otherwise unreflected. Critical experiments were conducted with the

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following material at the interface between the core and its 8-in. thick end reflector: 1) a 0.19-in. thick steel plate containing 0.3 w/o gadolinium, 2) 0.061-in. steel plate, 3) 0.25-in. thick steel plate, and 4) 8-in. thick Lucite reflector only.

The results of the experiments show that a 0.19-in. steel plate would reduce the reflector savings of the 8-in. Lucite by ~ 0.6 cm, whereas the 0.19-in. steel plate containing 0.3 w/o gadolinium reduced the reflector savings by ~ 0.8 cm; thus the Gd in the steel accounted for only 0.2 cm of the total decrease. These values are to be compared with 2.7 cm, the reduction in reflector savings on placing a 0.03-in. thick cadmium sheet at the interface. The ineffectiveness of the Gd is not explained; an analysis will be made to check the stated Gd content in the steel.

GAMTEC - A Neutron Slowing Down and Thermalization Code

Several major revisions and improvements have been made in the GAMTEC Code. These changes reduce the amount of input data and improve the theoretical accuracy of the code. The revisions are as follows:

- a. The input instructions were modified such that only correction cards will be required for problems following the initial problem of a run.
- b. The recent revisions in GAM-I, which include resonance theory calculations for isotopes other than U^{238} and Th^{232} , have been adapted.
- c. Corrections have been made to GAM-I which result in more accurate treatment of the resonance capture contributions to the multigroup constants in GAMTEC.
- d. GAMTEC presently punches the multigroup constants in a format for the HFN multigroup diffusion code. In the future, GAMTEC will also, by option, punch the multigroup constants in the SNG and S code formats.

SMC - A Monte Carlo Code for Spherical Geometry

A Monte Carlo code, SMC, has been written for studying neutron slowing down and diffusion in multi-region finite assemblies. SMC will compute criticality in spherical geometries containing one-to-three material regions. The SMC code will utilize group averaged cross sections with up to 1600 energy groups. The major components for SMC have been compiled and debugging is presently under way.



Theory of Measurement of Neutron Spectra

One of the most difficult and long standing problems in reactor physics involves the measurement of the neutron spectrum, particularly in the neutron energy range between thermal and fast systems. Yet any suitable theory for predicting criticality, in other than predominantly thermal systems, must also yield values for the neutron spectrum.

One of the methods of measurement which has been used in fast systems, and which in theory now appears promising for spectrum measurements in Pu systems at the Critical Mass Laboratory, is described in the following. The method requires that a number of foil materials, whose activation cross sections are known, be irradiated in an experimental assembly. A set of theoretical reaction rates are then generated from the theoretical flux to be compared. From a comparison of these reaction rates, a set of correction functions can be generated which indicate the region or regions where the assumed theoretical flux departs from the true experimental value.

A study of the method has been made by generating a set of reaction rates in the Wigner-Wilkins spectrum calculated for a 500 g/s plutonium nitrate solution, i.e., this spectrum was taken to be the true experimental one. The theoretical flux was then taken as another Wigner-Wilkins spectrum-that for a 100 g/s plutonium nitrate solution. The analysis was performed for neutron energies in the range O-1.13 eV utilizing six typical foil materials; Pu^{239} , U^{235} , Lu^{175} , Lu^{176} , Eu^{151} , and In^{115} . The convergence of the theoretical flux (100 g/s Pu) to the experimental flux (500 g/s Pu) was found to be satisfactory for the six chosen materials.

Critical Mass Laboratory Instrumentation

Work has proceeded on the design of an on-line noise analysis system for the Critical Mass Laboratory. The proposed system will have 40 frequency channels and provide a continuous display of the amplitude spectrum, with readout being accomplished with a digital printer or X-Y recorder. Provision will be made to incorporate a reactivity meter should the theory prove workable.

Consulting Services on Nuclear Safety - Criticality Hazards

1. Nuclear Safety in HL

Two new specifications were issued in HL:

K-8 - Storage and Transporting of EBWR Fuel Elements in 309 Building (PRTR Operation)

M-3 - Storage and Handling of EBWR Fuel Rods in 309 Building (PRCF Operation)

The void date on temporary specification No.3 (Storage of C and J) Fuel Elements Prior to Testing in 100-K) was extended from January 1, 1964, to July 1, 1964.

A nuclear safety inspection was made of the new Vibra-Pak machine area in the 308 Building for Ceramic Research and Development. Six glove boxes are set up in this area for processing dry PuO₂ and UO₂. It was confirmed that each of these glove boxes could be covered by N.S. Specification J-1. This specification permits up to 3.5 Kg of Pu in a glove box providing there are controls to prevent overbatching and preclude the introduction of liquids.

Favorable comments were received from the AEC, Division of Operational Safety, concerning the proposed GE Class I shipping container for fissile materials. A work order has been issued to have three containers fabricated for test purposes. Testing is planned to begin during March.

2. Nuclear Safety in CPD

Participation on the Redox Criticality Review Committee, in connection with the fire in the 233-S Building on November 6, 1963, has terminated with the completion of the review. Standard operations have now been resumed at Redox.

A nuclear safety evaluation of the Z-9 crib, based on neutron pulse measurements and soil sample analyses, is about 90% complete. The results will be submitted to Research and Engineering during February.

Maximum material bucklings for UO₂ rods in light water with enrichments of 2.7, 3.9, and 5.0 w/o U²³⁵ were provided to Advance Technical Planning. The values obtained for the three enrichments were 102 m⁻², 123 m⁻², and 136 m⁻², respectively. These maximum bucklings are about 30% lower than the bucklings for uranium metal rods in light water. This information is from a general study now in progress on enriched UO₂ systems.

3. Nuclear Safety in NRD

The nuclear safety study of N-fuels processing in the 333 Building for NFD is about 90% complete. Two rough draft documents have been issued to NRD and HL management for comment: HW-80312 RD summarizes the

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results of the study, and HW-80313 RD gives revised nuclear safety specifications for N-fuel processing.

4. Nuclear Safety Training and Education

The Course B, Group II lecture series in nuclear safety for CPD was completed on January 23, 1964. The course was comprised of eight sessions. The next session, Group III, will begin in February. This series is oriented toward the non-technical supervisors and specialists who have need for further understanding of the nuclear safety problem and of criticality control practices.

Separations Instrumentation and System Studies

Initial calculations were made to evaluate methods of monitoring large, inhomogeneous waste cartons to determine the amounts of included plutonium. The calculations indicate that a detector reading taken at a single point on each face of the carton would be inadequate and analyses are being continued to determine the effectiveness of complete surface scanning over a time interval of several minutes.

General systems requirements were studied regarding the development of a plutonium X-ray (17 keV) sample counting instrument which will be used to measure the plutonium content in liquid sample vials.

Design and partial fabrication was completed on a solid state portable gamma spectrometer instrument to provide measurement of plutonium in hoods and glove boxes. The instrument, with a scintillation detector, will monitor the 384 keV photons from plutonium.

NEUTRON CROSS SECTION PROGRAM

Scattering-Law Measurements for H₂O at 95°C

The triple-axis spectrometer at 105-KE was inoperative most of the month while modifications were being made to allow the use of the adjacent beam facility, 4-B, for slow-neutron scattering measurements by time-of-flight. The planned modifications to the triple-axis spectrometer have been essentially completed. The results previously obtained on the neutron scattering from 95°C H₂O have now been reduced to the scattering-law representation. These results show significant internal inconsistencies in some regions which will require further measurements. A series of measurements designed to determine the apparent systematic errors has been planned. An analysis of the energy sensitivity of the analyzing spectrometer has been completed.

Time-of-Flight Spectroscopy for Slow-Neutrons

Development of components for the measurement of slow-neutron inelastic scattering by time-of-flight has continued. Modification of the shielding of the triple-axis spectrometer at 105-KE to provide access to the beam hole 4-B has been substantially completed. The study of time jitter in slow neutron detectors has progressed to the analysis of electron drift velocities in several counter gases. These results should be of value in determining discrepant reports of drift velocities in BF3. Time-jitter measurements were obtained for additional gas mixtures using heliummethane and helium-carbon dioxide.

Fast-Neutron Cross Sections

The computer program CONTOUR which emphasizes certain features of fastneutron cross section variations has been almost completely debugged. Listings of the master tape of Hanford total cross section results were transmitted on request to Princeton University, Atomics International, and Los Alamos Scientific Laboratory. Samples of D, N, and O have been prepared for future measurement. All of these elements are to be measured in compounds. The blank samples to correct for other elements in the compounds are being prepared.

REACTOR DEVELOPMENT - 04 PROGRAM

PLUTONIUM RECYCLE PROGRAM

Phoenix Fuel Program

1. Utilization of the ZODIAC Burnup Code

The choice of the burnup time step increment, Δt has an important effect on the resulting reactivity-time curve. In past Phoenix reactor calculations, fairly coarse Δt 's were selected to save time and reduce machine costs. The development of the automatic ZODIAC burnup routine makes it now possible to utilize a finer time mesh and to obtain more accurate results.

The effect of Δt choices was investigated for a 100 kg Mark I Phoenix core. Nominal time increments of 500, 1000, 2000, 4000, and 8000 FPH were selected and reactivity time curves produced. A practical time interval (from a machine cost viewpoint) might be about 2000 Full Power Hours (FPH). Such a choice seems to introduce an error of about one percent in k_{eff} for a 10,000 FPH Phoenix-type core.



2. Beryllium-Plutonium Systems

A series of Be-Pu systems containing a variety of Pu composites are presently being analyzed. Such systems are of interest in connection with the Phoenix program, because the relatively hard spectrum obtainable in a Be medium might result in a favorable reactivity-time characteristic. The burnup-to-buildup ratio of Pu-240 represents an important figure of merit for these cores. For example, for a core with 20 a/o Pu-240 plutonium, the optimum 240 burnup-to-buildup ratio is $1.92\left(\sim \frac{.20}{.80} \ge 7.7\right)$. This is more than twice as high as the comparable value for a water-moderated core. On this basis, it would appear that Be/Pu systems would warrant further investigation.

3. Calculations for Proposed PPA-Phoenix Experiments

Critical experiments in the PPA (possibly the PCTR) have been proposed to obtain integral data characteristic of Phoenix-type cores. The early experiments will probably utilize 20 w/o Pu-Al discs with polyethylene or Be moderators. Calculations for Pu-Al-CH₂ media have started. Thus far the analysis has been restricted to homogenized systems ("thin" plates). Future studies will take heterogeneities into account.

Critical and Critical Approach Experiments

Low Exposure Pu02-U02 Lattice Studies in the PCTR

The Ceramics Research and Development Operation is working on the fuel for these experiments. High energy impaction is being used to densify the core material. Fifteen cans of 99% T.D. material have been produced. An additional 14 cans were impacted, but will have to be redone in order to achieve the required density. The 35 cans should provide enough material to complete the job. The 99% T.D. material must still be ground, particle sized, and then vibrationally compacted into the fuel rods.

Approach-to-Critical Experiments Using EBWR Fuel

The 0.71 inch lattice templates are in the approach-to-critical tank in the TTR reactor room. At the present time, all available fuel rods, approximately 400, have been loaded in this lattice. The experiment will be completed when sufficient fuel is obtained from Ceramics Research and Development Operation.

HoO Moderated Experiments in the PRCF

In the PRCF, light water-moderated core, a control rod consists of a cluster of four separate cadmium cylinders which have the same diameters as the fuel rods. The four cylinders are installed in lattice positions in the core. The lattice is hexagonal with a lattice spacing of 0.71 inches.

In order to estimate the reactivity worth of the control rod, an effective radius of a single control rod that has equivalent neutron absorption rate as the four separate rods, is being calculated. The Monte Carlo method is being used for the calculation and the main part of a flow chart is almost completed.

A report, "PRCF Experiments with H_2O Moderator," has been approved and issued as an informal document, HW-80092. This report describes critical experiments which are to be conducted in the light water-moderated PRCF. The experiments are divided into:

- 1. Load-to-Critical Experiments
- 2. Rod and Sheet Worth and Calibration Experiments
- 3. Moderator Level Worth and Sensitivity Measurements
- 4. Void Measurements
- 5. Kinetics Studies
- 6. Substitution Measurements
- 7. Neutron Spectrum Measurements
- 8. Temperature Coefficient Measurements
- 9. Flux Traverse Measurements

In addition to describing the experiments, the document provides the formal approval for conducting the experiments in the PRCF.

Process specifications for the H2O-moderated PRCF have been reviewed and comments forwarded to the author.

Preparation and planning for the EBWR experiments are proceeding. Efforts so far have been mainly on the void measurements and determining the requirements for a poison injection mechanism for the $\beta/2$ experiments.

D₂O Moderated Experiments in PRCF

The noise which was recorded during the noise experiment in the D₂O moderated PRCF has been frequency analyzed to obtain the transfer function of the loading. Preliminary analysis of the transfer function results in a value of ~ 5.5 for the ratio of the delayed neutron yield to the neutron lifetime.

The moderator level coefficients have been calculated from data obtained at moderator levels between 69 inches and 105 inches. Best fitting straight lines have been fit to the coefficients as a function of $(H)^{-3}$ where H is the moderator height at which the coefficient was measured. The slopes of the straight lines are proportional to the migration areas. One straight line was obtained for the data in the top reflector and another for the data in the core. The two straight lines intersect at the level where the ends of the safety rods remain in the reflector in their out position.

Reactor Theory

The Modified Heavy Gas Equation and Light Water-Moderated Systems

The neutron flux distributions produced by the modified heavy gas equation for infinite, homogeneous, light water-moderated systems were compared to that produced by program SPECTRE S. The modified gas equation produces accurate flux distributions for uranium systems up to a $\Delta = 1.0$, where $\Delta = 2$ (2200 meter absorption cross section)/(free atom scattering cross section). Accurate flux distributions were produced for plutonium systems for $\Delta < 0.3$. Since many thermal reactors have a Δ less than 0.3, the neutron flux distribution is adequately described by the modified heavy gas equation for water-uranium and water-plutonium systems. A report of this work has been prepared for inclusion in the October, November, December, 1963, Physics Research Quarterly Report.

High Energy Inelastic Scattering

The statistical model code which will calculate the excitation probabilities of discrete energy levels of a target nuclide is being debugged. References on the subject indicate that the probabilities are functions of the initial and final states of the target nuclide only, provided that the statistical model is valid. This model is valid only for high energy bombarding neutrons. For lower neutron bombarding energies, the more detailed properties of the compound nucleus must be considered. This latter approach is also under study.

Code Development

External Distribution of Machine Analysis

The latest active version of Hanford's multi-energy transport-theory analysis (GE-HL Program S-XII) was forwarded to Argonne National Laboratory for cooperative external distribution. Major improvements in nuclear analysis capabilities contained in the deck, and not previously available

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off-site at a similar level of generality include: (1) Angular sensor activation, (2) critical metric tensor, (3) kinetic normal modes, (4) full use of numeric rounding.

RBU

Review of Vol. III of RBU documentation is complete, suggested actions by reviewers have been inserted, and the manuscript has been submitted for publication as a formal document, HW-80003. In addition, a general cleanup of the RBU system decks and listings is under way in anticipation of external distribution. Successful execution of a test problem followed the first phase of cleanup. Aside from minor alterations during its checkout period, the RBU output program appears to operate in a satisfactory manner.

Adaptation of the PRTR 19-rod cluster analog to RBU provided additional nonserious difficulties during the month. Considering the long running time anticipated for the analysis (10-20 hours, estimated) a thorough checkout of the analog to achieve near optimum (RBU) operating conditions seems warranted. In total, the difficulties now being encountered are of a problem description nature and not system execution problems.

RBU Basic Library

To provide a more flexible description of the cross section data in the RBU Basic Library, an additional delta range is being added. This will allow $\ln \sigma$ to vary linearly with $\ln E$. A BARNS subroutine and a plotting code have also been written to enable plotting of the RBU Basic Library cross sections by the Benson-Lehner plotter. These programs will aid in identifying those energy ranges in which the additional analytic form is more appropriate than the present descriptions.

BARNS-II

BARNS-II is being revised to handle the additional delta range being added to the RBU Basic Library. Availability of this additional flexibility in the Basic Library and in BARNS before release of the RBU-BARNS package is considered essential.

HRG

Users of HRG frequently want to transcribe the calculated group cross sections into a form for subsequent input to Program S. To facilitate this process, a subroutine has been inserted in HRG which optionally punches these cross sections in Program S format. A single thermal group is assumed, for which ingroup cross sections can be read in or, alternatively,

the subroutine provides a dummy value. Suitable identification is provided in columns 73-78 of each output card. A listing of the cards follows the normal HRG output for the case. The subroutine has been debugged and is ready for trial use.

Physics Chain

A Physics Chain now exists which employs the use of COMBO as a means of input to TEMPEST and GAM. COMBO, as discussed in the monthly report of October, 1963, has been modified to allow multiple passes through TEMPEST and GAM, thus making it possible for the user to stack output from these codes on a TAM Library Tape - B7. This chain differs slightly from the previous Physics Chain in that each code other than COMBO clears its own storage before reading in and analyzing new data; also, after having done its analysis, each code has the facility of calling a new link on the chain by means of a variable input number. Due to these changes, the transportation code VIA is no longer needed for normal runs.

ZODIAC Chain

Further testing confirms that the iterative feature of ZODIAC is operating satisfactorily. Its use has cut the amount of time required to obtain results of a burnup calculation by a factor of ten or more.

The capacity of ZODIAC has been expanded from 10 to 18 groups. Work directed toward easing other restrictions is in progress.

An informal document, HW-80502, was issued which describes the capacities and input requirements of ZODIAC in its present state of development and gives a sample case.

Theory-Experiment Correlation

Analysis of Pu-Al Light Water Experiments

An analysis of computational methods is being conducted for the three sets of Pu-Al, light water critical experiments that have been conducted at Hanford: The L_x , the H_x , and the 5 w/o. A basic calculation of k_{eff} for each experiment has been calculated using HFN with four-energy groups. Cross sections for the two fast groups (ll.7 keV to 10 MeV, and 2.38 eV to ll.7 keV) were obtained from CAM-I. For the thermal (0 to 0.683 eV) and epithermal (0.683 eV to 2.38 eV) groups, average cross sections were obtained from cell calculations using THERMOS. The thermal group energy spectrum was generated both with the Brown-St. John and Nelkin scattering models. The Pu-240 cross section for the epithermal group was calculated, in addition

to the THERMOS method, using the NRIA approximation for calculating resonance integrals. The influence of the Pu-240 resonance peak at 1.054 eV on the thermal flux shape was investigated and, finally, a test case was run using the tightest lattice H_X case to determine what errors might result from using the cylindrical cell model for the lattice cell.

In brief summary, the basic calculation (using Nelkin scattering model for water) predicts values of k_{eff} for the various critical loadings that range from 1-3% high at the tight lattice spacings to 1-4% low at large lattice spacings. The results using the Brown-St. John model run $\sim 1\%$ higher. It should be noted that no attempt has been made to "fit" the experimental values. Consequently, these errors represent what might be expected were one to predict critical loadings with no experimental data to serve as a normalization.

The resonance integrals (for the Pu-240, 1.054 eV resonance only) run consistently higher when calculated with THERMOS than by the NRIA approximation. The difference ranges from \sim 5% for the 1.8 w/o L_x Pu-Al fuel to \sim 20% for the 5 w/o L_x Pu-Al fuel.

Burnup Experiments

Pu-Al Fuel Elements

The experimental data obtained from the destructive sampling of element 5108 have been received and the evaluation of the data has been initiated. The element is a plutonium-aluminum alloy fuel element which initially contained 2.57 w/o Pu, of which 16.5 a/o was Pu-240. The element was iradiated in the PRTR during the period April 2, 1962, to August 31, 1962, and received an exposure of 33.1 Mwd. Ten rods were subsequently selected from the element and sampled.

The processing of two other H_X Pu-Al elements is progressing. The majority of samples on element 5111 have been dissolved, and the preliminary burnup results appear to be of the same quality as the data on element 5108. Thus the scrutiny of the experimental procedures of Chemical Research Operation appear to have produced experimental data which is of higher quality than that obtained on earlier elements. Requests have been made for the disassembly of elements 5103 (L_X Pu-Al ~ 100 Mwd), 5109 (H_X Pu-Al ~ 150 Mwd), and 5187 (PuO2-UO2 ~ 100 Mwd). Rods from these elements will be gamma scanned as soon as possible.

The PRTR Gamma Scan Facility has been redesigned to improve operation. The changes made should result in a lower background counting rate, and more positive positioning of the fuel rod with respect to the scanning head.
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Correlation calculations using the ALTHAEA code, one-dimensional option, have been done for the case of the L_X Pu-Al fuel elements. Input cross sections were obtained using THERMOS and HRG for the diffusion calculation in ALTHAEA. Burnup parameters were not altered from those used in zerodimensional ALTHAEA except that the thermal disadvantage factors are now determined from a diffusion calculation. In the first attempt the fuel rods were considered to be one burnup region containing seven mesh points. After this description proved successful, the fuel rods were described as seven burnup regions containing one mesh point each and matched the detail that is available experimentally from macrodrill samples (see September, 1963, monthly report).

The ALTHAEA calculation using the description of a fuel rod as one region agrees well with the variation of Pu isotopic composition as a function of burnup obtained experimentally. For the variation of Pu-241 atom percent as a function of burnup, the ALTHAEA one-dimensional results are in better agreement with experimental results than were the zero-dimensional calculations. As with the zero-dimensional ALTHAEA, two sets of cross sections were considered. Since diffusion theory underpredicts thermal disadvantage factors, one would expect that ALTHAEA would not predict the correct difference in burnup between the center rod, the middle ring rod, and the outer ring rod at a given time. Nor should one expect that ALTHAEA would predict the correct variation of Pu isotopic composition through a rod by describing isotopic composition through a rod by describing the fuel as seven regions unless a number of the input parameters are adjusted. Therefore the ALTHAEA one-dimensional calculations were done as stated above, but further changes in ALTHAEA are not contemplated. (Correlation calculations for the macrodrill data will be reserved until such a time when a calculational tool becomes available.)

Isotopic Analysis of PRTR Samples

Isotopic analyses were provided on 33 plutonium samples of PRTR-irradiated fuel elements in support of the Plutonium Recycle Program. Of these, 14 were macrodrill samples from UO₂ element No. 1041, 8 were macrodrill samples from UO₂ element No. 1101, 8 were burnup samples from Al-Ni-Pu element No. 5111, and 3 were rechecks from Al-Ni-Pu element No. 5108.

Instrumentation and System Studies

The PRTR secondary activity coolant monitor was reviewed and several suggestions were made for its improvement.

The prototype aural-signaling monitor, developed for use with the PRTR automatic controller, was completed and is in routine use. It will be

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used in present form until the final model can be fabricated.

A complete instruction manual for the PRTR liquid effluent gamma monitors is being prepared.

The analysis of the power density spectrum of the PRTR vibration was completed by utilizing filters to measure directly the power density spectrum in each of the filter ranges. The data were computed and turned over to the Mechanical Equipment Development Operation. The tests were run to determine the origin of vibrations in the PRTR system. Vibration transducers located on the process tubes provided the vibration signals.

Extensive efforts have been made to place the 400 channel analyzes into full operation with existing time-of-flight equipment at the Van de Graaff facility. Satisfactory performance has not yet been obtained.

EBWR PROGRAM

Analysis of Uranium Critical Experiments

A series of homogeneous uranium critical systems have been analyzed to aid in the checkout of the cross section data for uranium on the updated RBU Basic Library, as well as comparing various methods of spectral averaging these data.

The critical systems were "well thermalized" (i.e., large H/U ratios) bare spheres. The calculated effective multiplication factors were all within 1% of the experimental value. The GAM-TEMPEST-HFN calculational method was employed.

The Yankee critical experiments were analyzed using the Nelkin scattering model for water and a comparison of k_{eff} 's was made with the Brown-St. John scattering model. The Nelkin k_{eff} 's ranged from 0.5% higher to 1% lower than the B.S.J. model when going from tight to loose lattices.

HIGH TEMPERATURE REACTOR PHYSICS PROGRAM

Preparations have been completed for the fourth in a series of high temperature tests of materials in nitrogen. Samples of Ni, thoria dispersed Ni, Hastelloy-X coated with aluminum, Hastelloy-B, Mo, and Inconel 600 samples will be heated to 1200 C in the nitrogen atmosphere. The samples will be contained in a Hastelloy-X box of internal dimensions $5-1/2 \times 5-1/2 \times 18$ inches and 3/8 inch wall thickness, and held in racks of graphite, Hastelloy-X, or ceramic brick. Samples of UO₂, $B_{\rm H}$ C, and possibly BN, each in capsules of Ni and graphite, will also be included in the test which is





to run for 800 to 1000 hours. The latter materials are being considered for use in the control rods and neutron chopper collimator. The box will be surrounded by an argon atmosphere while in the furnace.

A graphite beater element, 2 inches in diameter and 4-1/2 feet in length, has been tested in a preliminary reactor mockup assembly. The heater passes through a glindrical hole traversing the 4 foot length of a 4 inch x 4 inch graphite bar, and is connected to a low voltage transformer by graphite and copper leads. A layer of insulating bricks surrounds the heater rod. The heater and bricks are contained in a steel box which can be evacuated and then filled with nitrogen. In an initial run, the heater was operated at 5 kW (1000 amps), and reached 960 C after about 24 hours. Later, in an attempt to remove further impurities from the atmosphere, the system was pumped down while still hot. A leak in a sight tube, used for pyrometric temperature measurements, directed a stream of air at the heater, causing an eventual break at about the midpoint. A new heating element, of somewhat smaller diameter, has been constructed and installed. The smaller diameter will permit higher temperatures to be developed at currents of about 1000 amps. The tests are to provide sufficient information for the heater element design of the HTLTR and for construction of full scale prototype heaters to be tested in the larger mockup assembly.

The construction of the outer shell of the larger mockup has been completed and most of the insulating material for it is on hand. At the moment, work on it has been stopped because of lack of funds.

A report, HW-80536, "Proposed Techniques for k_{∞} Measurements in the HTLTR," is being written. This report proposes several methods for determining the mass of poison required to adjust a test cell in a lattice to a multiplication of unity and several methods for confirming the spectral adjustment. The need for the development of materials with the proper neutron activation characteristics and the ability to withstand high temperatures is emphasized.

A report, HW-80540, "HTLTR Time-of-Flight Spectrometer (Neutron Chopper)," has been written. It gives the specifications for the neutron chopper in sufficient detail for the preparation of design drawings.

NEUTRON FLUX MONITORS

The four uranium integrated exposure measurement samples, irradiated at KE Reactor with the plutonium elemental detectors, were analyzed using a mass spectrometer. The following isotopic compositions were obtained:

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	Isotopic Abundance (reicentage)					
	Original	Sample 1	Sample 2	Sample 3	Sample 4	
U-238 U-236	5.380 0.201	6.43 2.84	6.80 4.82	7.94 8.61	8.92 11.77	
U-235 U-234	1.082	1.22	1.30	1.47	1.60	

Isotonia Abundance (Percentage)

Now that these data are available to determine the integrated neutron exposure, it will be possible to compare the experimental changes in isotopic composition of the plutonium regenerating elements to the calculated changes expected. If satisfactory agreement is obtained, this phase of the experiment will be concluded and a report prepared.

Work continued on the boron-ll beta-current generator neutron flux monitor concept and a third experimental monitor was designed and is being fabricated. Considerable laboratory testing was carried out following irradiation tests with the second monitor to determine the cause for the excessive noise current being generated. The results indicated that included water vapor in the detector chamber and in the rigid metal-sheath conductor could be one cause of the problem and that a second possible cause could be ionization of the air surrounding the leads to the detector element. The complete 20 feet long assembly for the third experimental monitor is being carefully cleaned and will be evacuated in an attempt to alleviate the problem. This assembly, now nearly complete, will contain a boron-ll detecting element plus one additional lead wire to be used to measure cable effects. Irradiations will commence in a KW-Reactor facility as soon as the monitor is completed and sealed.

Arrangements have been completed, with the acceptance of the Production Test Request, for the testing of the experimental microwave neutron flux monitor concept at KW-Reactor. The necessary test loops were assembled and plasma capsules have been fabricated. Two tests have been planned. The first will provide measurement of the resonant frequency of the nitrogen plasma filled capsule; the frequency change should be proportional to the neutron flux impinging the capsule. The planned second test will consist of two waveguides coupled by a gas cylinder. The signal level coupled to the return waveguide should be proportional to the neutron flux. The planned +ests will be of short duration with later long-term experiments to be conducted at a DR-Reactor facility.

NONDESTRUCTIVE TESTING RESEARCH

Electromagnetic Testing

A major advance has been made in the concept of the multiparameter eddy

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current nondestructive testing device which will simplify the calibrating and operating procedures. Initial application of the new concept to just one parameter readout channel of a multiparameter tester showed that a calibration which would have previously required several hours can now be accomplished in a few minutes. The new technique will expedite the application of the multiparameter test to new problems and should increase customer confidence and acceptance of the new testers. The new modification will permit the calibration of the tester to be accomplished by the positioning of multidimensional control levers, each of which simultaneously sets all the coefficient potentiometers in the transformation (summing) section associated with one parameter readout. The operator will adjust the control levers to obtain the desired degree of discrimination between the different parameter readout channels. Previously, the coefficient potentiometers were set after a rather laborious routine including measurements of the effects of individual parameter variations, setting up a matrix representing the relationships between parameters and eddy current signals, the computation of an inverse matrix, and the setting of each coefficient potentiometer in accordance with the values of the inverse matrix elements. Following this, the individual settings of the coefficient potentiometers would be trimmed to give optimum instrument performance. The new modification will eliminate all of the foregoing steps except the last one.

The newly required mechanical modifications are being incorporated in the multiparameter tube tester and being developed.

A prototype 360° phase shifter was built for use with the 1004 eddy current tube tester. Tests show that the phase shifter expedites the set-up of the tester and simplifies its operation. Amplitude variation is less than 0.5 percent over its full range. The prototype operates at any one of five frequencies; 100 KC, 200 KC, 400 KC, and 1600 KC.

Heat Transfer Testing

The new dual radiometer emissivity independent heat transfer testing concept was experimentally demonstrated on aluminum clad fuel elements containing mica bond defects. A larger range of infrared emissivity differences than normally found in newly fabricated fuel elements was used in the experiments. Some of the fuel element surfaces were partially covered with hammer dents to determine the effect of a bumpy surface. Bond defects 3/8inch in diameter could be detected in all cases. Based on these results, use of the method to study the effect of irradiation on the heat transfer quality of fuel element core to cladding bonds appears to be one possible application.

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Areas having low emissivity were produced on the cladding surfaces of fuel elements by polishing to a mirror finish. High emissivity areas were obtained by painting with black tempera paint, flame heating, and rubbing the areas with a cloth to remove any loosened paint. The latter step is necessary to reduce non-uniformities in heat transfer through the paint so that it will have less effect on the surface temperatures sensed by the radiometer.

Experimental results were almost entirely independent of surface emissivities of the samples, even though variations in the original signal due to emissivity differences were as great as signals from the 3/8 inch diameter mica defects. Improvement over the earlier dual radiometer method was obtained by inserting a constant signal from an oscillator into the output of one radiometer. This compensates for a constant infrared signal from an apprent unknown stray source which is "seen" by the second radiometer.

A bond fracture occurred in one of the dented fuel elements upon subsequent thermal cycling during testing. Once initiated, the fracture propagated during each test. The element had been tested several times prior to denting with no effect on the bond. It is felt that the surface denting was a strong factor in initiation of the fracture since no fractures have occurred during several hundred other fuel element tests (one of the bench standards has been tested one hundred times with no deterioration of the bond). The fracture initiated near the cap end of the fuel element during the cooling portion of the thermal cycle.

Investigation of a possible new concept in analysis of heat transfer test data is under way. This concept is based on basic heat flow theory and should be applicable to a wide variety of heat transfer testing problems.

Zircaloy-2 Hydride Detection

Process tube sections prepared by Chemical Metallurgy, HL, for burst tests by Materials Engineering, HL, are being used to evaluate the eddy current hydride mapping equipment.

A total of 18 tube sections have been tested to date and the hydride contents are being determined metallographically. Four of the sections were prepared by an electrolytic method, and no hydrogen content was detected by the eddy current equipment. These results were confirmed by metallographic examination. No further samples are to be produced by the electrolytic method. Five samples had been prepared from a length of tube containing longitudinal gouges about .020 inch deep and .5 inch wide. The signal variations caused by this surface condition were greater than the signals from the hydrided areas containing 500 ppm hydrogen and obscured



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the test results as reported last month. It has been determined that these samples are not typical of acceptable process tubes and, at present, it is felt that such gouges will not be a problem to in-reactor tests.

Scans during present hydride tests are made on a lathe. This limits the length of tube sample to about 21 inches. A rotating test head is being developed to handle longer samples and perform tests at frequencies of 44 KC and 455 KC simultaneously. A rotating transformer will be used in the initial model to couple the test coils to the other circuits.

A sample of process tube has been machined to produce a bench standard having eight steps in wall thickness from 0.205 to 0.272 inch. Results from tests with this standard show that the 44 KC detector is independent of wall thickness down to about 0.235 inch. The wall thickness variations in the standard do not affect the 455 KC detector.

Fundamental Ultrasonic Studies

In order to analytically study the ultrasonic wave behavior associated with liquid-solid and solid-solid interfaces, it is necessary to obtain phase velocities and attenuation coefficients for typical solids (such as metals). Longitudinal velocity measurements are easily obtained with equipment on hand and shear velocity measurements will be made possible with transducers now on order.

The measurement of attenuation coefficients is more difficult. The analytical studies currently being conducted deal with plane, constant amplitude waves which are spatially infinite. Such waves are difficult to generate experimentally because of diffraction effects which accompany the transmission of waves from transducers of finite size.

The diffraction results in interferences, in the near or Fresnel field, between plane and spherical waves leading to undesirable variations in wave amplitude and phase. Also, in the far or Fraunhofer field the waves become spherical and, because of beam spreading, a correction is necessary to obtain true, plane wave attenuation coefficients. As practical transducers are not ideal piston radiators, however, the accuracy of these corrections is questionable.

The usual solution is to confine the attenuation measurements to wave propagation distances which do not exceed the extent of the Fresnel field. The amplitude variations are then averaged over the transducer face. Actually, some experimenters have found that the useful distance over which the attenuation can be measured is about twice the extent of the Fresnel field.

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To demonstrate the accuracy of determining attenuation by this method a previously measured sample of aluminum was re-evaluated. The longitudinal wave attenuation was found to be uniform over a distance approximately equal to twice the Fresnel field length and the measured attenuation coefficient agreed well with values quoted in the literature. The method therefore appears to be valid.

The effects of surface curvature on critical angle tests of compressively stressed samples has been investigated. Unstressed samples were prepared which had surface curvature in excess of that measured on the stressed samples. When these curved samples were compared with an optically flat sample no differences in critical angle signals were measurable. On this basis the signal changes observed on the stressed samples are assumed to be due to the compressive load.

USAEC-AECL COOPERATIVE PROGRAM

Nondestructive Testing of Sheath Tubing

The mechanical system of the prototype sheath tubing tester was altered slightly to accept eight-foot lengths of 30 mil wall, 500 mil diameter Zircaloy tubing. Utilization of the prototype station to test various diameters and wall thickness of Hanford tubing has continued. The tester has shown its general utility in that tubing of various diameters with wall thickness ranging from 17 mils to 50 mils have been tested with this unit. For each test the 3/16 inch diameter spherically focused crystal is aligned so that shear waves at 45° are generated within the tube walls. In each case, signals from the standard notches appear at intervals of time which agree with the propagation time for zigzag shear waves. With the defect gate positioned so that both inner and outer diameter defect signals pass through, defects down to one mil depth in 17 to 50 mil wall Zircaloy tubing are reliably located.

Destructive analysis with adequate photographic records of defects located in the reject tubes received from AECL is nearing completion. All ten defects located by the tester have been located destructively. The amplitude of the ultrasonic signal received from the defects agreed within 20% of its measured depth on all but one defect. This defect was located by both the axial and circumferential tests with amplitudes less than the maximum defect depth observed by sectioning. At one section the defect appeared as a crack oriented 45° to the tube wall and extending through 50% of its tube wall. Examination of the samples also revealed areas which appeared to have extensive hydriding along the deeper defects. Test records of the remaining reject AECL tubes not sectioned have been



assembled. These records and tubes are being forwarded to AECL for evaluation and comparison to AECL test results.

Fabrication of standards using a punch fitted into the Rockwell hardness tester is continuing with good success. Notches 15 mils in length are punched, measured optically for depth and then polished off to within 0.1 mil of the desired depth. This entire operation requires one hour per notch on the outer surface. Inner surface notches will require more effort, however. An auxiliary lever arm modification was incorporated on the hardness tester so that notches can be punched on the inner surface up to one inch in from the tube end. Due to the added difficulties of measuring and polishing, these notches will require more time to fabricate and the notch depth accuracy will decrease.

Fabrication of a complete set of 1, 2, and 3 mil notches in 10, 17, 25, 45, and 65 mil wall tubing has been accomplished with the Rockwell hardness punch. These standards will be used to gather data for the AECL cooperative program final report. Preliminary tests indicate the notches are of a good quality, and that by utilizing the 3/16 inch diameter crystal, one mil notches can be detected in 65 mil wall Zircaloy tubing.

Modifications were accomplished on an earlier draft of topical report, HW-77464, "Ultrasonic Response of Notches in 35 Mil Wall Zircaloy Tubing." This report will be published in the near future. A second report describing the instrument and techniques developed for measuring ultrasonic transducer characteristics is under preparation with the rough draft 50% complete.

BIOLOGY AND MEDICINE - 06 PROGRAM

Atmospheric Physics

The mathematical model for the lateral growth of a diffusing plume of material dispersed to the atmosphere derived from Hanford ground source data was shown to apply also for some elevated source releases. Independent verification of the model was made using data from 16 releases at 200 feet height made during 1955-1956. The wind variability factor was determined from wind velocity measurements at the release height. Agreement between the new model and the data was far superior to the power function used earlier.

The experimental data of the scavenging action of rain with zinc sulfide particles have been analyzed. These data were taken with 0.36 to 1.5millimeter diameter raindrops (which include most numerous drops in natural rains) and 3.4 to 13.8-micrometer diameter particles. After defining the

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scavenging efficiency to be the ratio of the number of particles collected by a raindrop to the number in the drop's path, the following characteristics may be noted:

- 1. The collection efficiency observed with a drop diameter of 0.4 millimeter is greater than those observed with larger drops,
- 2. All raindrops show efficiencies greater than 1.0 for ZnS particles larger than 13 microns diameter. Particles must, therefore, be moving transversely into the cylinder swept by the falling raindrops,
- 3. The collection efficiency increases quite rapidly with particle size in the general region of 10 microns diameter, and
- 4. There is a minimum collection efficiency for all particle sizes with intermediate drop diameters (in the region of 0.8 millimeter drop diameter).

These characteristics do not conform with those currently used for prediction. The theoretical calculations of Langmuir show, instead:

- 1. A rather flat peak in efficiency with drop sizes of 1.2 millimeters, and a low efficiency at 0.4 millimeter diameter, and
- 2. A rapid increase in efficiency with particle size for very small particles, asymptotically approaching 1.0 as a maximum.

The tentative explanation of the Hanford results is as follows. Those particles that barely miss the drop as it falls gain another opportunity for contact when the fluid convergence in the wake and electrostatic attraction carry the particles into the raindrop wake. The vortices present in the wake provide a circulation which can impact the particles on the trailing or upper side of the raindrop. In the case of the smaller raindrop, these vortices remain attached to the drop as it falls and provide a large time interval for particle collection. In the case of larger drops, the vortices may be shed before the particles can impact. While the vortices are attached, particles therein fall with respect to the drop and are attracted toward the drop by electrostatic forces. The collection of the heavier and larger particles is thereby favored. The wake explanation has support in that data taken by Kinzer & Cobb with freely falling water drops in the laboratory show a peaking in efficiency for drops of 0.4 millimeter diameter. Collection in the wakes of falling drops has also been observed by several other researchers, and even mathematically demonstrated by Pearcey and Hill.

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In instrumentation work, the wind component meter circuitry was modified to include a filter system to remove the heating current noise frequencies from the turbulence signal. Field operation to date has shown this modification to greatly improve the signal to noise ratio. Temperature sensors on the portable mast were compared in an extensive series of operational tests. Five channels were found to be well within required accuracy limits, but the sixth channel consistently behaved erratically, and will require correction.

Radiological Physics

A field party took a shadow shield whole body counter to Anaktuvuk Pass, Alaska, and measured the radioactivity in the Eskimos there again. Preliminary reports say that the amounts of Cs^{137} in these Eskimos have decreased from the high values of last summer.

Studies with the plutonium counter were aimed at reducing its background. A partially completed graded shield (lead-cadmium-copper) reduced the background under the plutonium X-ray photopeak by about 20% and under the combined X-ray and Am^{241} photopeaks by about 40%; these were backgrounds of the empty counter. Placing a single unit of the counter inside the large well-crystal scintillation counter of Radiological Chemistry and operating the two in anticoincidence produced a substantial reduction in background.

The positive ion Van de Graaff operated satisfactorily during the month. Last year the accelerator was available for experimental work 77% of the time; this is 20% more than the preceding year.

The neutron efficiency of a LiI scintillator moderated by either 2 or 3-inch diameter boron-loaded polyethylene spheres proved to change too rapidly with energy to be the adjunct to pulse shape discrimination that we desired.

A calibration of the precision long counter using the S(n,p) reaction gave efficiencies of 1.012 at 4.8 MeV and 0.73 at 14.6 MeV. These compare favorably with 0.965 and 0.75 obtained from the literature.

The proportional counter for the neutron spectrometer began to work and testing is giving encouraging results. Pulse shape discrimination will be used to reject gamma rays. The counter gives pulses of the right shape for this purpose. The resolution still needs to be improved.

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Good-geometry transmission experiments are in progress to select materials to test as neutron shields.

Calorimetric measurements of Pm^{147} were resumed.

The new calorimeter is meant for use in alpha or beta ray measurements and in radiation dosimetry. These require very much less mass for absorbing the radiation than in the present gamma-ray calorimeter. Less mass means smaller heat capacity and shorter time constant. Development versions of the new calorimeter gave time constants of 2 to 20 minutes depending on the insulation used, compared with 13 hours for the present one. Sensitivities were about the same as in the present one.

Studies of an improved pulse forming circuit for an ionization chamber reader advanced to the point where fabrication of a reading device was started.

Instrumentation and System Studies

Progress was achieved on the biological function telemetry system with the development and testing of a new transducer and transmitter for animal temperature measurement. Laboratory tests were satisfactory. A transducer for respiration rate measurement was designed using a breath-cooled, self heated thermistor which controls an oscillator that is energized once per breath. The laboratory results were promising and a packaged model is being fabricated. Work was continued on the development of the miniature blood pressure transducer; however, the blood pressure measurements are considerably more difficult than temperature and breathing rate measurements.

Successful modification was made to the controls of the special sliding valve used in experimental animal inhalation studies at the Biology Laboratory. The modifications reduced the 60 cycle noise and vibration to acceptable levels. The problem of incorrect transducer switching, caused by animal movements, was rectified by a new mounting procedure. The experimental system is presently being used in biology field tests.

Design was completed on the detector assembly head to be used with an alpha energy analysis air monitor for the Biology Inhalation Laboratory.

Progress was achieved on the development of a special radiological spectrometer monitor to be used in field tests by Biology personnel. The solid state single channel analyzer and the high voltage supply portions have now been completed.

Development effort was initiated on a pulse shape discriminator for dosimetry experiments. The discriminator will distinguish multiplier phototube noise pulses from pulses generated by NaI(Tl) crystals at pulse heights equivalent to those excited by plutonium X-rays. An additional benefit may be the use of the method for pulse shape discrimination in neutron detection experiments.

Engineering development was provided and the basic circuit design established for a pulse rise time discriminator circuit for use between proportional counters and a multichannel analyzer. Prototype fabrication was initiated on the circuit which will be applied in neutron dosimetry investigations.

Experiments were conducted with special low-noise, large-area multiplier phototubes, using a combination scintillator, in an effort to develop a sensitive combined alpha and beta detection method. Promising results were achieved using selected phototubes and a mosaic pattern for the employed scintillators.

Progress was made on the fabrication of a second data station for the HAPO Radiotelemetry System. The station was essentially completed and testing was started. Work was initiated on a special test function chassis which will be used for general testing of all data stations when completed. A number of the data station circuit drawings were also prepared.

Additional filters were designed and fabricated for use in the wind component meter system, which will be employed in atmospheric physics studies. The installation of these filters will complete the originally planned development and modification effort and extensive laboratory tests will then be carried out.

Work was essentially completed on the real-time fluorescent particle detection and measurement instrumentation to be used in atmospheric physics particle transport studies. Field tests appeared to be satisfactory and a descriptive report is being prepared.

Operational troubles persisted in the commercial digital voltmeter portion of the portable mast data logging and recording system. It appears that the voltmeter manufacturer will be asked to resolve the problem.

Development effort was initiated on a special single electron counting system which is to be capable of detecting and counting every electron emitted from the cathode of a multiplier phototube. The system will be employed in dosimetry studies.



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WASHINGTON DESIGNATED PROGRAM

Isotopic Analysis Program

Isotopic analyses were provided on program samples during the month in accordance with current schedules.

Studies of the operational characteristics of the mass spectrometer equipped with the newly installed scintillation-type ion detector continued after parts were received for repair of the 50 kV ion-detector power supply. A baffle was installed between the exit slit of the spectrometer and the detector aperture to prevent scattered ions from reaching the detector. Following installation of the baffle the abundance sensitivity of the spectrometer improved to the value it had been with the original ion-detector. The effective dead time of the ion-counting system was measured to be $0.28 \ \mu sec$. The vendor has still been unable to deliver the basic parts of the vacuum-lock sample changer.

EXPERIMENTAL REACTOR PHYSICS FACILITIES

PCTR Operation

The PCTR operated routinely during the month. There were three unscheduled shutdowns caused by electronic failures.

A set of foils was irradiated in the thermal column for standardization. At the same time a set of similar foils was irradiated in the N-Reactor.

NPR process tubes, cross-header tubes, and process water were measured for nuclear purity.

TIR Operation

The TTR was operated on a two nights a week basis for the University of Washington Graduate Center. There was one unscheduled shutdown caused by an electronic failure.

Subcritical Facility

The first loading of the EBWR fuel was started, using an 0.71 inch lattice spacing in water. Approximately four hundred rods have been received to date and loaded into the tank. The predicted critical loading based on this incomplete loading is 500-520 fuel rods.



HW-80560

CUSTOMER WORK

Weather Forecasting and Meteorological Services

Meteorological and climatological consultation services included additional work on the N-Reactor accident calculations and review of proposed release rates of oxides of nitrogen from a chemical processing plant.

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

Weather Summary

Type of Forecast	Number Made	% Reliability	
8-Hour Production 24-Hour General	93 62	82.9 81.0	
Special	178	88.2	

January was considerably warmer, drier, and windier than usual.

Mass Spectrometry

Isotopic analyses were provided on 5 samples of uranium in support of HAPO U^{233} -production studies.

Instrumentation and System Studies

Engineering drawings and specifications were completed regarding control circuits for use in the 324 Building. The work was done for Waste Calcination Demonstrations, HL.

All of the commercial instrumentation has now been received for use in the U-235 fuel enrichment measurement system designed for Metal Fabrication Development, HL. Several of the instruments were modified to fit established system requirements. One detector assembly is being fabricated.

An automatic range selector circuit was designed for use in a commercial control circuit for Coolant Systems Development, HL.

Circuit board layout work was completed on the final model solid state count-rate integrator unit, which will drive an electromechanical register, for Radiological Development and Calibrations, HL. The integrator will be used with portable neutron dose rate instruments to provide a measure

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of accumulated dose.

Bids for a complete system of temperature comparators to be used as outof-limit detectors at the 100-KW test facility were reviewed. No bidder met specifications. Either a revision of the specifications or a negotiated contract will be undertaken.

Preliminary instructions for the creep capsule data logger were given to acquaint Reactor Metals Research personnel with the system before proceeding with installation at the 100-KW Reactor. The data logging system operated successfully throughout the month. A few minor design modifications were incorporated as further debugging progressed. Writing of the system operation and maintenance manual started and when the manual is completed, an instruction class for operation and maintenance personnel will be held.

A high pressure mercury arc lamp for use in development of a remote displacement measuring system was received and given preliminary checks. It was determined that safety procedures in handling such lamps will be established because of the extremely high pressure and strong ultra violet spectrum associated with the lamp.

Optics

A second series of pictures were taken of electron microscope plates to test the ability of a mocked up dimension rectifying camera to restore original dimensions to pictures of metal surfaces taken by reflection. The prints restored proper relative dimensions to surfaces viewed at 8°, 12°, 16° , 20° , 24° , and 27° . The magnification varied in the ratio of 2.5/1.05 for the 27° versus the 8° picture. Prints are now being made to obtain the same magnification at all angles to assist evaluation of the results.

The Los Alamos program for lens design on a 7090 computer has been run on their sample problem and on a Hanford design problem. The sample problem was successful; however, three attempts to obtain an improved wide angle lens design for borescopes have failed. The design instructions have been modified for a fourth try.

Fabrication of a new traverse mechanism was completed and tested. Test runs in an ex-reactor tube at B Area were also made. Data from these runs are being used to calibrate the device and evaluate its performance. Data analysis of the first run are encouraging.



A traverse mechanism is being designed for use in the N Reactor graphite traverse holes. Fabrication has already begun although the design is only about 50% complete. The design incorporates improvements suggested by experience with developmental models and the traverse mechanism used in the old reactors.

During the five-week period (December 15 to January 19) included in this report, 522 manhours shop work was performed. This work included:

- 1. Fabrication of 10 glass bearings for CPD waste pumps.
- 2. Fabrication of a traverse mechanism for the old reactors.
- 3. Repair of one crane periscope for Purex.
- 4. Repair of four camera shutters.
- 5. Fabrication of two clip spacing measurement probes for Physical Testing.
- 6. Repair of one microscope and a metallograph for Chemical Metallurgy, HL.
- 7. Repair of two pan-bore camera probes for Testing Methods.
- 8. Fabrication of a lens mount to adapt a pair of binoculars for focus to within five feet.
- 9. Repair of a theodolite for Atmospheric Physics.
- 10. Repair of an M-2 borescope for Physical Testing.
- 11. Repair of the optical system of a balance for Radiometallurgy.
- 12. Fabrication of miscellaneous windows, discs, insulators and microscope slides for twelve different customers.

Physical Testing

Inspection service was provided N Reactor Department on an emergency basis to determine the position and the extent of damage of leaking tubes in the Graphite Heat Exchangers. The breaks in the tubing were found to always be in exactly the same position in the unit. Other tubes were inspected by the eddy current method and the information turned over to N Reactor engineers for their information and action.

Ultrasonic flaw detection was applied on the North head of Steam Generator 4-A at N Reactor in preparation for the welding of lifting lugs to retube the generator.

A program is under way to determine the stress relieving and annealing kinetics of stainless steel. The purpose is to provide data for the evaluation of stainless as a replacement for Inconel in the K Reactor resistance temperature detectors. Tests will determine thermal distortion as well as the corrosion resistance of the various stress relieving conditions.

Laboratory tests have now shown that the wear of tube under the baffle plates can be detected in the PRTR stainless steel heat exchanger HX-5. Tests showing simulated baffles and machined wall reductions indicate this condition can be detected and identified for wall reductions of 0.020 inches or greater and that 0.015 inches seem to be the threshold of sensitivity. Eddy current techniques are applicable to this problem.

Work is proceeding on the scoping of a storage tank examination facility to be installed in the 324 Building in conjunction with the Waste Calcination Demonstration Program (WCDP). This facility will comprise an integrated, remotely operated station for measuring such parameters as wall thickness, surface temperature, internal pressure, content homogeneity and changes in properties for the prototypical storage tanks.

A factor of two increase in the sensitivity of the apparatus being developed to simultaneously measure the electrical resistivity and the density of plutonium during phase changes has been achieved with modifications to the spring support and contact assembly.

The high voltage power supply has been received, modified, and tested for the capacitor discharge program. Only a few items have yet to be received before the system can be completed to inject high quantities of energy in small specimens to achieve instantaneous heating.

Another bonus has been realized from the introduction of the ultrasonic translator for locating leaks in the plant telephone lines. Cold weather, which used to curtail the search for leaks with the soap solution, has no effect on the translator and allows the telephone crews to provide a higher level of service during this season of the year.

Pressure vessel surveys were continued through the fourth quarter of last year and into the first quarter of this year to ultrasonically measure thicknesses at possible corrosion points, assuring the safe operation of this equipment as well as maintaining a lower insurance rate.

COMPUTER FACILITIES

Analog-computer utilization was as follows:

176	Hours	Up Time
24	Hours	Scheduled Downtime
16	Hours	Unscheduled Downtime

216 Hours Total

The times given above include all off-shift operation (swing shift and week ends).

Problems considered during the month were:

- 1. PRTR Vibrational Analysis.
- 2. N Reactor Primary Flow Simulation.
- 3. N Reactor Pressurizer Simulation.

The new analog computer is nowbeing tested for compliance with specifications; testing is approximately 40% complete. As the tests proceed, a number of operational difficulties are becoming apparent. These will need to be corrected before the computer is placed in service for general purpose use.

A one-week class on the operation of the new computer was completed. The one-week maintenance class was scheduled for the week starting January 20.

INSTRUMENT EVALUATION

All seven of the offsite fabricated combined alpha-beta-gamma scintillation hand and shoe counters, as obtained on the initial order, have passed acceptance tests and are ready for plant services. Testing was continued on three more counters fabricated by a different manufacturer. In several instances, incorrect parts had been installed and the detection efficiency appeared marginal for several of the probes.

Effort was continued in updating the Model II Scintran prints and purchase specifications. The bid package, necessary for offsite fabrication, was completed.

Engineering evaluation was conducted and design modifications incorporated regarding the use of Lucite light pipes for a special set of efficient scintillation alpha-beta-gamma counter probes.

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Manager PHYSICS AND INSTRUMENTS LABORATORY

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CHEMICAL LABORATORY

RESEARCH AND ENGINEERING

FISSIONABLE MATERIALS - 02 PROGRAM

IRRADIATION PROCESSES

Overheated N-Reactor Fuel Study

An NPR inner fuel element irradiated to about 2300 MWD/T and cooled approximately six months was inductively heated to 980 C successively in three positions along its length. The object of the experiment was to characterize the swelling or other changes in an element when water is lost from the coolant channel and the element overheats. When the goal temperature was reached smoothly in five minutes, slight swelling occurred during the ensuing five minutes at temperature. When the element was held for 30 minutes at temperature, swelling was more pronounced. The fuel cladding did not rupture.

Experiments are planned for full length heating of an irradiated element followed by steam and water quenching.

Disposal to Ground

A study is underway to define more accurately the minimum ground-water travel time from the 1301-N Crib to the Columbia River. The primary purpose of this study is to determine the effects on travel time of surface springs which will probably develop along the riverbank, 900 feet from the crib. (The Advisory Committee on Reactor Safeguards postulated that the predicted ll-day minimum travel time may be appreciably reduced due to the underground channelling of wastes from the crib to the riverbank springs.) Field phases of the study involve investigation of a site at 100-H Area, which has similar hydraulic characteristics to 100-N Area, where springs have developed over a period of time. Flow results determined from conductance-paper analogs of the two sites will be compared, and actual travel time from the 107-H Basin to the riverbank springs will be determined from measurements of radioiodine concentrations in samples taken at these locations. Preliminary evaluations indicate that a safety factor of from 10 to 20 is incorporated into the 11-day minimum travel time estimated for 100-N Area. This safety margin is due primarily to the fact that the actual soil permeability is appreciably less than the value assumed in the earlier 100-N Area travel time estimate.







SEPARATIONS PROCESSES

Purex Solvent Studies

A 250 ml sample of Soltrol-TBP solution (G-5), contaminated with fission products, was received from the Purex plant for examination to determine, if possible, the reason for the unusually high ruthenium retention

Two lines of investigation were employed. Gas chromatographic analysis, aimed at discovering whether or not a volatile, ruthenium-containing organic complex was formed, separated the mixture into three fractions water and low-boiling decomposition products, Soltrol and TBP. Only iodine was detected upon analysis of the fractions, the other fission products presumably remaining in the injection system. A liquid chromatography experiment was run to determine whether or not a silica gel column prepared with Soltrol 170 would be as effective in removing fission product complexing agents from a TBP-Soltrol solution as it is with Soltrol alone. The results of the experiment showed that TBP is more tightly held by the silica gel than are the usual Soltrol impurities. Further, feed-level fission product activity broke through the column after the passage of only two column volumes of the Purex solvent, indicating that this treatment will be of little value in cleaning up used Purex solvent, although it may be significant that appreciable quantities of ruthenium were held up on the gel at the top end of the column .

Trilauryl Amine Extraction of Neptunium and Plutonium from Purex 1WW

A flowsheet for recovery of neptunium and plutonium from Purex 1WW and subsequent further treatment in the Purex 3A-3C columns (J-Cell) was tested using actual Purex 1WW as feed. Feed for the F-Cell portion of the flowsheet was produced by stripping the trilauryl amine-Soltrol organic with a hydroxylamine sulfate solution. Behavior of ruthenium, cerium and zirconium-niobium during TBP-Soltrol extraction of this feed solution was comparable to that reported earlier for traced simulated feeds. It was necessary to add ferrous sulfamate to the hydroxylamine sulfate strip solution to prevent extraction of plutonium into TBP-Soltrol under 3A column conditions.

Scrap Recovery Flowsheets

The current flowsheet under study for 234-5 Building processing of scrap containing plutonium, uranium and thorium involves co-extraction of uranium and thorium into TBP-Soltrol in the first column. Batch contact studies have established operating conditions for adequate recovery of uranium and thorium in the first column. It is expected that mini-mixersettler runs to demonstrate the flowsheet will be completed during February.







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Ion Exchange Contactor Development

Initial studies of the Redox prototype countercurrent anion exchange column (described in the December monthly report, HW-79999 C) have been completed. A three-day continuous run simulating actual use conditions, using thorium as a stand-in for plutonium, showed the unit to be stable with dependable resin movement of 7 to 9 inches occurring in every pumping period. The time required for resin movement ranged from 11 to 13 percent, well below a plant specified maximum of 20 percent. Waste losses at a steady state were typically less than 0.1 percent although fluctuations to 0.9 percent were observed. Resin loading averaged 7 grams thorium per liter of resin. The run was terminated after three days. A new run had been started when a bellows failure prevented further testing. A new resin pump unit is being constructed and testing of the pump's ability to "push" as well as "pull" the resin bed around the contactor will continue.

The existing Purex anion exchange mockup will be modified to verify the reliability of resin movement for a proposed Purex contactor modification. The dependence of the contactor upon pumping variables, acidity, flow rates, and temperature will be studied.

Polonium-210 Processes

Current proposals for bismuth irradiation and processing for Po-210 recovery include alternates for recovery and recycle of the irradiated bismuth. During the month, bismuth recycle has been studied from the standpoint of containment controls and processing steps that would be required.

Actual containment requirements for recycle bismuth will depend on the polonium decontamination, quantities, the chemical form, and the operating conditions. The ability of polonium to migrate or escape from the bismuth will also be a factor. Since there are little data on most of the items, only rough conclusions can be made on the basis of activity and MPC_a comparisons with Pu=239, natural uranium, and PRTR Pu=Al alloy. Preliminary conclusions are as follows:

- 1. For a Po=210 DF of 100 or less, gloved=box handling of recycle bis= muth in any chemical form will almost certainly be required.
- 2. Gloved-box handling is probably necessary for a Po-210 DF of 10² to 10⁴ for many chemical forms (especially particulate solids). However, the closer a DF of 10⁴ is approached the more questionable the need for gloved-box handling for solid metal pieces.









3. With a Po-210 DF in the range of 10^4 to 10^5 , it is likely that ventilated open-face hoods can be used. Such handling would then be similar to that for uranium and thorium.

The preceding conclusions do not take into account any additional shielding or remote-handling requirements that might be encountered in recycle bismuth due to activation of initial or added impurities. Both beta and gamma activity would be present as a result of activation of impurities in the bismuth. The long-lived isomer of Bi-210 (2.6 x 10^6 years) would also be present in small amounts. Residual amounts of Po-210 and shortlived Bi-210 would also be present.

U-233 Separations Chemistry Studies

B-Cell processing of the first of the thorium (metal) target elements was completed during the month. This included dissolution in the in-cell one-slug dissolver, scavenging of protactinium with MnO_2 , multi-stage batch extraction of the uranium with 10 percent TBP, and isolation in small volume by stripping with dilute caustic. Following similar processing of the remaining two slugs, the uranium fractions will be combined, further decontaminated if necessary and removed from the cell for final cleanup and product isolation in the laboratory. Isotopic analyses of the dissolved slug are not complete, but the one pair of values so far reported for U-232 and U-233 content is in good agreement with the weighted average of earlier analyses on small samples machined from various positions in the slug. The fact that the two sets of analyses were performed by different analysts and by different analytical methods is particularly gratifying and lends additional confidence to the curves being used as a planning basis for current production studies.

In supporting laboratory work, experiments were aimed at defining the processing scheme to be used for the final purification of the above material. Scouting work was also continued on protactinium scavenging or adsorption processes, on ways to improve the thorium processing capacity and/or decontamination potential of the Redox and Purex plants, and on close-coupled processing schemes. In support of the latter, a quantity of thoria spiked with Pa=233 and uranium was prepared by spray calcination. Leaching studies are in progress to determine whether the protactinium and uranium can be removed without dissolving the thoria.

Production of Thoria by Spray Calcination

A brief study of the potential of radiant-heat spray calcination for the conversion of thorium nitrate solutions to target-grade thoria was completed, using the 8-in \times 10-ft. "cold" laboratory unit. It was found







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that thoria of high purity and low nitrate and moisture content was readily produced. (The fact that the spray droplets do not contact the walls of the calciner minimizes pick-up of corrosion products.) Tap density of the oxide was rather low (about 3 g/cc); however, it could be easily pressed to 65-70 percent theoretical density at modest pressures (40 tsi). The as-pressed pellets were quite strong and cohesive and could be readily loaded into cans. The pressed pellets sintered to as high as 87 percent theoretical density. High energy-rate compaction (Dynapak) gave a product of virtually theoretical density, which was suitable for crushing, grading and vibrational compaction. Surface area of the as-produced powder was about 3 square meters/gm and mean particle size about 1 micron. Efforts to form a stable gel from the powder were unsuccessful, probably because the particle size was larger and the surface area smaller than ORNL has found optimum for Sol-Gel feed.

Particularly noteworthy were the ease and high rate of dissolution of the pressed pellets in fluoride-catalyzed nitric acid. The pellets disintegrated after a few minutes exposure and were completely dissolved in less than 15 minutes, corresponding to apparent penetration rates of about 8400 mg/cm²/hr (based on the superficial area of the pellet). Pene-tration rates of sintered or Dynapaked material were much lower (by factors of about 20 to 25). Details are summarized in an informal report, HW-80531, "Tests of Thoria Pellets and Their Production from Radiant-Heat Spray Calcined Thoria by Pressing, Sintering, and High Energy Rate Impaction."

Thorium Processing

In addition to the foregoing work on spray calcination, the ORNL-developed Sol-Gel process is currently being considered for Hanford application as a means of preparing low cost, high density thoria for recycle in the production of "clean" U=233.

A modified Sol-Gel process being studied consists of four steps in which thorium nitrate is denitrated in the presence of steam; the thoria product is dispersed as a sol in water with nitric acid added in trace quantities; the sol is dried to form a cake; and finally the cake is fired to 1150 C to produce a near theoretical density product suitable for vibrational compaction. The tap density increases from 4 g/cc for the initial denitrated thoria to about 9.8 g/cc for the fired product. A program was initiated to study the four phases of the process with emphasis on applicability to existing Hanford equipment.

The preliminary work indicates that low viscosity stable thoria sols can be prepared in concentrations up to 7 <u>M</u> thoria. Thixotropic age= sensitive sols were prepared in the range of 8 <u>M</u> to 9 <u>M</u> thoria, the more









concentrated the sol, the more age-sensitive and gel-like the mixture. Sols prepared at 8 <u>M</u> thoria concentration, though initially fluid, aged without drying to thixotropic and near-solid gels. Studies also indicate that high viscosity or thixotropic sols prepared with high thoria concentration can be reversibly diluted to fluid sols. Additions of water and/or ammonium hydroxide can be used to control the viscosity of the sols.

Americium Extraction

Americium(III) was extracted from 2 <u>M</u> aluminum nitrate - 0.2 <u>M</u> nitric acid solutions with the quaternary amine, Aliquat 336, in a xylene diluent. The results show a third power dependence of the extraction coefficient upon amine concentration, E_a^o values ranging from 0.05 for 1.8 volume percent Aliquat 336 to 15 for 4.1 volume percent.

Preliminary results indicate that Am^{3+} is readily extracted from 1 <u>M</u> NH₁CNS by either 10 volume percent DBBP or 10 volume percent Aliquat 336 in xylene. E^o values of 34 and 14, respectively, were measured for these two solvents.

The distribution of Am^{3+} between TBP-DBBP-Xylene solutions and 4.75 MNaNO₃=0.25 M nitric acid solutions was measured. The results, listed below, do not suggest any synergistic effects:

% TBP	% DBBP	Ea
-	20	5.15
5	15	1:63
10	10	0.50
15	5	0.30
20	0	0.10

Neptunium-Plutonium Separation by Amine Extraction

Soltrol 170 has been substituted as the diluent for trilauryl amine in the batch amine extraction of plutonium(IV) from neptunium(V). Mixtures of n-octyl alcohol and dodecane were being used, but 10 percent octyl alcohol was needed to solubilize 2 g/l Pu in a 15 v/o trilauryl amine solution. Soltrol 170 apparently has enough aromatic content to increase the solubility of the plutonium-amine compound without the addition of octyl alcohol.

Solvent Extraction from Molten Salts

Solvent extraction of Eu(III) by triphenyl phosphate (TPP) in a polyphenyl mixture from $LiNO_3KNO_3$ eutectic at 150-160 C was found to be unformable. The distribution coefficient was less than 10^{-2} for a 0.1 mixture function of TPP. Further examination of Eu(III) extraction showed







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the magnitude of the distribution coefficients to have the following order: tributyl phosphate (TBP) > triphenylphosphine oxide (TPPO) > TPP. Other extractants will be examined to further establish the order of extraction. Thermal decomposition of TPPO is suspected and will be checked.

WASTE MANAGEMENT AND FISSION PRODUCT RECOVERY

CSREX Process Engineering

Planned pilot plant work on the CSREX process has been completed. Final pulse column runs were devoted to testing (a) the effects of flow rate and pulsing conditions on fission product extraction, (b) the effects of adding phosphotungstic acid (as a potential "head-end" cesium precipitant) to the feed, and (c) of using sodium carbonate washed solvent (all in the sodium form) on the performance of the column battery. The results of the runs can be summarized as follows:

- 1. Using a tartrate-complexed feed, strontium and cesium losses as low as 0.2 percent were obtained in the 1A column at volume velocities up to 520 gph/ft². The strontium loss increased to 3 percent at 710 gph/ft². Cerium losses increased from 2 to 5 percent over the same range of flow rates.
- Lowering the pulse frequency from ≥ 95 to 85 percent of the flooding frequency in the 1A column usually increased the fission product losses about two- to three-fold above the optimum values shown above.
- 3. Cesium losses in the 1B column were ≥ 1 percent at flow rates up to 600 gph/ft². Corresponding strontium losses were about 0.2 percent. Typical cerium decontamination factors (DF) were in the range of 20 to 50. There was no calcium DF. Varying the frequency from 85 to 95 percent of flooding had little effect on losses.
- 4. The lBP strontium-cesium product contained, as contamination, about 0.0007 g/l Fe, 0.02 g/l Ni, and $\geq 0.3 \text{ g/l}$ Al. The individual concentrations varied by a factor of 4 or more.
- 5. Typical cerium losses in the 1C column at 190, 390 and 590 gph/ft² were 0.2, 2.0 and 4.0% respectively. The losses were about doubled, however, in attempts to reproduce the runs with solvent from a different series of 1A-1B runs.
- 6. Decreasing the pulse frequency from 95 to 85 percent of flooding generally increased the cerium loss in the 1C column by about three-fold.









- 7. The addition of phosphotungstic acid to the feed did not affect cesium losses or column performance. Although a precipitate appeared to form on addition of the acid to the feed (at pH 5), it was quickly redissolved.
- 8. The use of freshly-carbonate-washed solvent (with no acidification) was satisfactorily demonstrated. The major effect of using a basic solvent was to increase the IAW pH from 4.5 to 5.0. As a consequence, the cerium loss in the IA column increased to 14 percent. The strontium and cesium losses were 3 and 0.1 percent, respectively. This flowsheet modification would eliminate the addition of concentrated acid to the washed solvent, a potential cause of solvent degradation.
- 9. Sodium and nickel decontamination factors of 10 to 20, and 3 to 8, respectively, were obtained in the 1S column. The typical product solvent from the 1S column contained 0.005 <u>M</u> sodium and 0.0002 <u>M</u> nickel.

Technetium Purification and Reduction to Metal

Laboratory studies were continued in the direction of defining the flow sheet to be used for final purification and reduction to metal of the onekilogram quantity of CPD-recovered technetium. Silica gel removal of $2r-Nb_s$ loading at low pH onto an IRA-401 resin column, washing with water and 0.1 M HNO₃, and elution with ammonium thiocyanate were piloted on a small scale with some of the actual feed. Decontamination from fission product contaminants was virtually complete, only an inconsequential trace of wethenium remaining in the product.

Elution with thiocyanate was found to be quantitative (contrary to the reports of other investigators, who, however, worked in chloride media). A concentration of about 1.5 <u>M</u> appears to be near optimum, and the ammonium thiocyanate in the product solution is readily destroyed by reacting with nitric acid. Greater decontamination from Zr-Nb is obtained with thio-cyanate than with HNO₃ elution, suggesting the formation of anionic zirconium and niobium thiocyanate complexes which strongly resist elution from the resin.

The final isolation of technetium in a form which can be reduced to the metal is usually done by recrystallizing $\mathrm{NH}_{1}\mathrm{TeO}_{1}$. Precipitation as TeO_{2} or TeO_{2} , $\mathrm{2H}_{2}\mathrm{O}$ is being studied as an alternate approach. Addition of formal-dehyde to a slightly acid technetium solution precipitated a black solid which was shown by analysis to be predominately TeO_{2} ; however, the yield was only 52 percent. Use of hydrazine precipitated TeO_{2} , $\mathrm{2H}_{2}\mathrm{O}$ quantitatively (> 95 percent). The dioxide was soluble in dilute HNO_{3} but not in ammoniacal $\mathrm{H}_{2}\mathrm{O}$, whereas the hydrate dissolved in both.

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Cesium Extractants

In support of the CSREX process, 4-methyl-2-alpha-methylbenzylphenetole was prepared and submitted for testing effectiveness in extraction of cesium. The measured E_a^O of 0.05 indicates that the hydroxy group of the BAMBP material is essential for extraction. Efforts to cleave the ether group with HI were not successful. Other more vigorous reactions will be attempted. Efforts to prepare 4-cyano-2-alpha-methylbenzylphenol and 4-nitro-2-alpha-methylbenzylphenol were not successful. Either a higher reflux temperature or a more effective catalyst, such as HF, is required for the substitution reaction.

Cerium-Yttrium Adsorption on Linde 13X Zeolite

Synthetic zeolite Linde 13X is selective for Ce^{+3} . On the basis of cation size, it could be predicted that Y⁺³ (1.84 Å diameter), smaller in size than Ce^{+3} (2.14 Å diameter), would be selectively less adsorbed on 13X. Experimental results confirmed that 13X is slightly less Y⁺³ selective, though the two isotherms have the same general configuration.

Generally speaking, zeolites with the higher silica-to-alumina ratios have shown a preference for large, univalent cations. Type A with the lowest silica-to-alumina ratio (2:1) has the highest selectivity for divalent cations. The most selective zeolite for trivalent cations was 13X (2.4:1), which also was divalent cation selective though less so than Type A.

If each type of cation is preferred by a specific group of anionic sites with a characteristic adsorption energy, as seems to be the case, then it should be possible to synthesize a zeolite with an optimum number or distribution of each of the three site groups. The zeolite 13X, possessing both divalent and trivalent-selective cation sites, may be a good starting point to develop a synthetic zeolite that prefers Cs^+ , Sr^{+2} or Ce^{+3} to Na⁺.

Thermodynamic Relationships in Multication Systems

The use of mass action relationships for predicting trace cation K_d 's in systems containing two monovalent cations was extended to trace strontium solutions. The equation used for computing strontium K_d 's from binary exchange data is as follows:











where: K^{Sr^*} and K^{Sr^*} = Mass action quotients for trace Sr^{+2} in A B monovalent A and B cation systems.

A and B = Concentration of A^+ and B^+ in equ:valents N N per liter.

Computed and experimental strontium K_d 's agree within experimental error for systems containing trace strontium, $NH_{l_1} - H^+$, $NH_{l_1} - Na^+$, and $Na^+ - H^+$ with IR-120 and for $NH_{l_1}^+ - Na^+$ systems with Decalso and clinoptilolite.

EQUIPMENT AND MATERIALS

Examination Cf Purex H-4 Titanium Bundle Heat Exchanger

The Purex H-4 heat exchanger was examined following cleaning and pickling by Chemical Processing Department, Waste Handling and Decontamination Operation personnel. The titanium tube bundle appeared well cleaned except for the portion above the shroud which could not be immersed in the cleaning solution. Some tightly adherent black scale, less than three mils thick, was present on the condensate chest and the tubes. Three of the stainless steel shroud support legs appeared in good condition. Metal surfaces were clean and bright; welds were in good condition with some evidence of intergranular attack. The other two shroud support legs were dull, greyish-brown in finish and exhibited non-uniform corrosion attack. Welds had suffered knife-line attack in the base metal and were corroded through completely in some places. Samples from both types of shroud support legs are being taken for further examination. The center parts of stainless steel tube spacers appeared significantly more corroded than outer parts.

Linear Polarization as an Evaluation Test

Linear polarization of sensitized 304 and 304L stainless steel in deaerated boiling 65 w/o HNO₃ was examined as a rapid means of determining susceptibility to intergranular attack. A significant different in potential vs. current density for the two materials occurred only after about 16 hours' exposure in the nitric acid. Exposure of materials in HNO₃-Cr(VI) solution prior to polarization was not effective in magnifying differences in polarization behavior. Reproducibility of results was difficult due to strong dependence of polarization (both anodic and cathodic) on prior specimen history (polishing, time since polishing, passivation and exposure time in the test solution).

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Non-Metallic Materials

Corning 7913 scintillating glass suffered no change in physical appearance or dimensions during prolonged exposure to one molar nitric acid at room temperature and at 50 C.

A carbon-filled, cross-linked, thermosetting polyethylene (CAB-XL) showed no increase in brittleness and less than one percent increase in thickness after 29 days' exposure in Purex HAX at 50 C.

An insulating varnish, Doryl (Westinghouse) had a volume resistivity of 10^{17} ohm-cm when applied to a metallic substrate and cured. After irradiation with Co- $^{\circ}0$ gamma to 10^{0} R, the varnish had a volume resistivity of 2 x 10^{14} ohm-cm. The 3.5 mil thick sample broke down under a potential of about 1000 volts; the manufacturer's literature indicated a breakdown voltage of 3000 volts/mil for non-irradiated varnish.

Two samples of shielding window lead glass (density 3.8) were exposed to mixed gamma radiation from casks of calcined waste in a 325-A Building cell. No damage occurred to either sample during exposure to about 10^6 R. Exposure of one sample is continuing at a rate of about 9800 R/hr. The sample is still undamaged at 3.5 x 10^6 R.

PROCESS CONTROL AND DEVELOPMENT

Automatic Control System for Plutonium Reclamation Facility

The control loops associated with the Gradient Control System and the pulse . control systems were given a final calibration check preparatory to plant delivery. The summing amplifiers used to correct the column density measurement for flow rate effects were calibrated to compensate at an amplitude frequency product of 60 inches per minute. Precise compensation can be achieved for any amplitude frequency product by adjusting the system to give zero response to variation in flow rates.

Initial plant cold runs will utilize the pulse control system. Installation of the prototype gradient control system will await completion and acceptance of major contractual work in Plutonium Recycle Facility."

Plutonium Detection Test Facility

The test loop was operated with plutonium solutions ranging from 0.1 to 755 mg per liter. Two in-line detectors were tested, a 17 kev X-ray crystal and an alpha-sensitive glass scintillator. Over the concentration range tested, the X-ray device provided count rates about a factor of two higher than those obtained with the scintillating glass. Modifications to







the latter detector are being made, however, to increase its sensitivity and signal-to-noise ratio. Both cells appeared to decontaminate acceptably well, but additional data is required concerning this effect. Garma tolerance tests are now being performed by the addition of cesium-134. At the low concentration so far used (2.7 uc per liter) plutonium count rates are unchanged.

C-Column Instrumentation

Analysis of response characteristics of the C-column to changes in uranium concentration of the feed is an important part of the overall C-column study. As an aid in investigating these characteristics, a blending system was devised to produce either a ramp or step change in the feed uranium concentration. The system consists of two storage tanks and two control valves with a single air operator. A uranium photometer measures the concentration of the blended solution and provides a signal to control the position of the dual blend valve. A ramping system enables changes in the concentration at any desired rate between any two concentrations within the range limited by the storage tank concentrations.

Installation of the blending system has been completed and successful operations were achieved.

Computer Control Studies

Work has begun on programming the GE-412 computer for optimal control of the experimental C-column in 321 Building. Several major functions which the computer and peripheral equipment must perform have been incorporated into the overall control scheme, i.e.:

- 1. Control the scanner-converter to bring current process information into the computer, check the data for being out-of-limits, compute time averages and variances, and store the information for use by other sections of the code.
- 2. Take computed values for the set points of the process controllers and output them through digital-to-analog converter.
- 3. Type out periodic logs of selected process information.
- 4. Sense emergency conditions and take appropriate action.
- 5. Using the mathematical model of the process, compute set-point values to optimize operation.
- 6. Using current and past process data, update the model to compensate for process changes, and alarm if changes are such that a significant upset is impending.





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7. Synchronize all the above functions in real time through "executive" control.

The capabilities of today's "second generation" process control computers, plus the availability of advanced software packages help considerably in reducing the complexity of the programming task.

ANALYTICAL AND INSTRUMENTAL CHEMISTRY

Burn-Up Analysis

An effort to improve the reliability of present methods for analyzing reactor fuel burn-up has increased the precision of analysis by a factor of about two. The major source of error most probably was in the stated sample concentration, caused by incomplete mixing of the sample prior to sampling in Radiometallurgy. The procedure has been slightly modified to eliminate this possibility.

The scatter in burn-up analysis is now small enough to reveal a definite bias between coulometric and alpha-count determinations of plutonium. It is believed that a major part of this bias lies in the half-life value used for Pu-240. A larger bias of 3.9 percent is evident between the coulometrically determined plutonium concentration and that specified as originally present by the fuels manufacturer. This makes a 6.7 percent bias between the alpha-count determination of plutonium and the original value. Efforts to locate this bias are underway.

Since the start of this investigation, several samples have been re-run. Since the burn-up values, as determined by the ratio method, agree within the experimental error of \pm 5 percent with the original burnup numbers, ratio burnups are and probably have been good values.

Detection and Measurement of Hg-203 in Duck Muscle

Forty-seven-day Hg-203 was found in muscle from four ducks taken in the vicinity of the 200-West swamp. Repeated gamma counting of the muscle over a period of time and plotting the counts in the 0.28 Mev region after correcting for interferences tentatively identified the nuclide. Separation of mercury confirmed the result. Chemistry included wet ashing with 1:1 concentrated H_2SO_4 and HNO_3 acids and precipitating the mercury as metal with ascorbic acid. Radiochemical yield exceeded 90 percent.





C-14



REACTOR DEVELOPMENT - 04 PROGRAM

PLUTONIUM RECYCLE PROGRAM

Salt Cycle Process

EMF Studies of Molten Salt Solutions - The studies of the Ni + Cl₂ = NiCl₂ reaction in various mixtures of KCl-LiCl, KCl-NaCl, and NaCl-LiCl have been completed. The data were considerably more reproducible than for the UO₀(VI) system, and the results invite more confidence. The plots of EMF vs. mole percent NaCl or KCl in the solvent produced nearly straight lines. In the NaCl-LiCl system, however, the behavior of UO₂(VI) and Ni(II) are considerably different. Although there may be some curvature in the EMF vs. mole percent NaCl (in LiCl) plots for Ni(II), in the solutions with low NaCl concentrations, the titration-type curve observed for UO₂(VI) does not exist. Another interesting feature is the greater slope for the UO₂(VI) solutions in the LiCl-KCl system than for Ni(II) solutions. Perhaps of more significance is the observation that the slopes of the EMF-composition curves for Ni(II) in the different solvent systems increased in the order: NaCl-LiCl, KCl-NaCl, and KCl-LiCl. For the UO₂(VI) solutions, the slopes increased as the melt chloride activity increased; i.e., increasing in the order NaCl-LiCl, KCl-LiCl, and KCl-NaCl.

Plutonium Oxychloride Equilibria and Reaction Rates - Stu :s have been made of the behavior of plutonium and uranium in equal-molar LiCl-KCl at concentrations (0.2 w/o Pu and 15 w/o Pu) and temperatures (500 to 600 C) representative of those employed in the C-cell demonstration runs. It was found that the constants for the equilibria

> $Pu0_2Cl_2 = Pu0_2Cl + 1/2 Cl_2$ $Pu0_2Cl = Pu0_2 + 1/2 Cl_2$

are such that only <u>ca</u> $0.1 \, \text{W/o}$ Pu can be maintained indefinitely in this solution as plutonyl species, even at Cl₂ partial pressures near unity. The rate of precipitation of PuO₂ from $0.2 \, \text{W/o}$ plutonyl solutions sparged with 0_2 -Cl₂ is fairly slow, however, so that some solutions containing more than the equilibrium concentration of plutonyl species can be maintained for some time. Rapid precipitation occurs when the chlorine is removed from the solution by sparging with helium.

Rates of oxidation of U(IV) and of Pu'(IV) by O_2-Cl_2 sparging were determined under various conditions. The rate of U(IV) oxidation (roughly 2 w/o, hr⁻¹ at 600 C) was about twice that of Pu(IV) and the rates at 600 C were roughly four times as great as those at 500 C. The rates depended more on the oxygen partial pressure in the sparge gas than on the oxygen flow rate. On O_2-Cl_2 sparging a melt containing both U(IV) and Pu(IV), oxidation of the Pu(IV) did not commence until the U(IV) had been oxidized.

DEFESSION









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Engineering Development - Sixty days' continuous operation of the Salt Cycle Process in C-cell of the High Level Radiochemistry Facility was concluded. During the operating period four UO_2 -Pu O_2 co-depositions were made at varying operating conditions using unirradiated mixed oxide elements as feed material. At present the cell is being readied for load-in of irradiated PRTR fuel elements.

Final analysis received on the second deposit as reported last month showed plutonium to uranium ratios from bottom to top of 0.0064, 0.0076, and 0.011 or plutonium enrichment factors of 0.71, 0.84, and 1.22, respectively. The chloride content of the deposit washed as 1/2 inch particles ranged between 50 to 140 ppm. The oxygen to uranium ratios on the average were less than 2.005.

A third deposit of 9.8 pounds was obtained by electrolyzing the salt bath which had an initial composition of 17.2 w/o uranium and 0.068 w/o plutonium at 1.40 volts with an initial current density of 0.19 amps/cm². The current efficiency of the electrolysis was 66 percent. The 550 C salt bath was sparged with 0.2 l/m Cl₂ and 1.8 l/m O₂. Preliminary analysis indicates that the plutonium content varied less than a factor of 2 across the cross section of the deposit with a maximum occurring in the middle.

The fourth electrolysis run at high voltage as a bath clean-up, produced a 1.25 pound highly dendritic deposit. Preliminary analysis indicates that the deposit contained approximately 20 percent uranium and 65 percent nickel metal resulting from a piece of metal inadvertently introduced into the bath before electrolysis. This type of deposition is a cermet which may have uses in its own right.

Polyvinylchloride Resistance to Salt Cycle Off-Gas - Unplasticized polyvinylchloride was exposed to 4 M NaOH or 4 M NaOH-0.5 M H₂O₂ solution sparged with chlorine and/or hydrogen chloride. Specimen temperatures varied from 40 to 65 C. Attack was negligible at 50 C or less. At 65 C a bleaching action occurred and the test specimen gained 0.2 percent in weight during 24 hours" exposure. Attack was primarily on that portion of the test specimen in the vapor phase.

Dissolution of PRTR U02=Pu02 Fuels

Laboratory studies have demonstrated the feasibility of a two-step procedure for dissolution of PRTR U0₂-Pu0₂ fuels. The uranium oxide is dissolved first to produce a low-acid high-uranium solution. The slower-dissolving plutonium oxide is then dissolved in a HNO₃-NH₄F solution at 70-80 C. Corrosion studies indicate aluminum may be omitted during the plutonium oxide dissolution if the temperature is maintained at or below 80 C; plutonium oxide dissolution rates were satisfactorily high. Separate dissolution of the plutonium oxide does however, present potential criticality problems in the Redox multi-purpose dissolver and may severely limit resible.charge size. These problems are being studied.





Plutonium Recycle Fuel Processing Economics Study

Evaluation of close-coupled fuel reprocessing feasibility for the advanced pressurized-water reactor (APWR) case has been essentially completed. The results are summarized in the following table in comparison with the previous case designated as a typical water moderated reactor (TWR):

MAXIMUM ALLOWED CAPITAL INVESTMENT IN CLOSE-COUPLED PLANT FACILITIES

1000 MW_e Reactor Complex with 1/6 of Fission Product Poisons Removed on Each Cycle

	Fuel Exposure At			
•	Theoretical Optimum	20,000 MWD/T	15,000 MWD/T	10,000 MWD/T
Typical Water Reactor (Theoretical Optimum Exposure 25,000 to 30,000 MWD/T)	\$4,400,000 to 6,900,000	\$5,100,000 to 8,000,000	\$6,400,000 to 10,300,000	\$9,700,000 to 14,000,000
Advanced Pressurized Water Reactor (Theoretical Optimum Exposure 30,000 to 40,000 MWD/T)	4,300,000 to 6,800,000	6,000,000 to 9,500,000	8,000,000 to 13,000,000	12,500,000 to 18,000,000

The comparison indicates the maximum allowable capital investment in the close-coupled plant facilities for a break-even situation relative to the alternative of central-plant reprocessing. The allowable capital investment would be reduced approximately \$1.7 million for each 0.1 mill/KWH savings advantage over the central-plant fuel cycle cost.

The range of investment values shown for each goal exposure represents the optimized high and low value calculated with the range of unit cost inputs and return on investment ratios used in this study. Results for a more specific set of conditions can be expected to lie somewhere in between. A 15-year plant life was assumed in all cases.

The results presented are also based on achieving an overall fission product poison decontamination factor of 6 for each fuel recycle from the close-coupled plant. This was chosen as being a reasonable target for a Salt Cycle type process. Complete poison decontamination would increase allowable capital investment approximately \$0.5 million for the APWR case and approximately \$0.9 million for the TWR case.







The allowed capital investment must cover the construction cost of the close-coupled process, a fuel fabrication facility for the portion of the fuel recycled as mixed oxide (50 to 25 percent of total fuel supplied to the reactor) and a fluoride volatility unit for processing any valuable by-product uranium for sale or return to the AEC. It is unlikely that the total complex could be built for anything less than \$5 million.

These results indicate economic feasibility for the close-coupled process serving a reactor complex on the order of 1000 MW_e capacity when fuel exposures are in the range of 10,000 to 20,000 MWD/T (90 to 45 tons per year). However, if optimum fuel exposures of 25,000 to 40,000 MWD/T are actually achieved routinely in operating power reactors, close-coupled reprocessing is not likely to be feasible in serving less than 2000 MW_e of reactor capacity.

RADIOACTIVE RESIDUE PROCESSING DEVELOPMENT

Calcination Studies

One run was completed during the report period in the A-Cell spray calciner; a run in which "cold" fission product stand-ins were added to Purex waste to simulate the chemical complexion of a 10,000 MWD/T waste, such as would result from high-burnup power reactor fuels. The feed, which was a fine slurry, was pumped and spray calcined without difficulty. Fission product behavior was somewhat better than normally observed with acidic waste in that only 6 percent of the radioactive ruthenium was volatilized. Glass will be made from the calcined powder to test glass behavior under high internal radiation conditions. (Because of the short cooling time of the Hanford Purex plant waste, gross beta-gamma level is about the same as for aged power reactor fuels.)

Cold Semiworks Spray Calciner

The experimental development program on the 18-inch diameter calciner continued employing a 16-inch diameter by 8.5-foot long draft tube and using a simulated Redox feed with borax and silica additives.

Operations with a 550 C wall temperature resulted in internal deposits in the calciner (probably because of the presence of low melting point $NaNO_3$). A later run with sugar added to the feed to destroy the $NaNO_3$ reduced the deposits even with a furnace temperature of 800 C.

Calcine Melter

Five very successful melt runs were made in the continuous melter using a modified powder feeding section and bellows discharge valve.




Thirty-five pounds of glass were collected in a 10-inch length of 8-inch diameter pipe during a melter operating time of ∂ -1/2 hours. The glass was discharged from the melter in batches forming 1 to 1-1/2 inch layers in the pot. The pot of glass was not annealed and the surface was covered with cracks. Radiographs of the pot showed relatively few cracks beneath the top layer of glass.

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Spectrographic and wet chemical analyses of two earlier samples of the lithium-phosphate ORNL-type "glass" were received. One sample consisted of melt which had been formed in the melter during early runs before corrosion of the pot wall was noted. The other sample was residual nonmelting solids which formed a crust on the melter wall. Significant composition differences were noted in sulfate, phosphate, and nickel content. Nickel content in the non-melting crust was qualitatively listed as strong while that in the regular melt was moderate to strong. The sulfate content of the crust was 3.2 weight percent while that of the regular melt was 16 percent. In the crust, the phosphate composition was 16 weight percent but was 42 percent in the regular melt. Particles of fused metal rich in nickel content could be seen in the crust sample. The crust sample had been subjected to temperatures near 900 C for possibly as long as 75 hours, while the regular melt sample had been molten for a period of only several minutes.

Materials of Construction for Waste Calciner Melt Pots

Crucibles fabricated from 310 and 304L stainless steels have been exposed to spray-calcined Purex-type waste containing phosphate and lithium additive at temperatures of 750 and 900 C for about 68 hours. Based on these single exposures, both materials appear more suitable for service in this environment than Inconel 600.

Continuous Glass Making

Laboratory studies of effects of composition (particularly fission product content) on glass proportions and preparations for the hot cell glass experiment continued.

The month's laboratory studies further emphasized the large chemical effect of fission products, at levels expected in power fuels, on the formation and properties of glasses proposed for waste fixation. In one series of experiments, varying the ratio of fission products to process chemical oxides in a phosphate glass system had a pronounced effect on drip temperature, appearance and chemical resistance. A sinter was obtained with > 80%FP oxides, glasses in the range of 40-70 percent FP oxides, and microcrystalline solids below 40 percent. The glass containing 60 percent fission product oxides was spectacularly resistant to attack by nitric acid, overnight exposure to 12 M acid failing even to dull the surface gloss.







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The 10 and 20 percent samples, on the other hand, were readily decomposed by acid treatment. In another set of experiments, changing the simulated burnup from 16,500 to 22,000 MWD/T altered the behavior from that of a sharp melting, compound-like material to a glass. These studies are continuing and will be extended to include fully radioactive systems. Further details are given in the "Waste Fixation Quarterly for October-December, 1963," HW-80526.

Shipment of the Brookhaven equipment for the hot cell glass experiment is currently expected prior to February 1. In the meantime, familiarization experiments with a "cold" laboratory apparatus have continued, including testing of several alternate components. A coiled tube evaporator was designed, installed and found to work well in initial tests. It was selfdraining and showed no tendency to plug. The new platinum inclined-pot melter was received and installed, but showed only limited success in its first trial. The falling-film evaporator was found to be unsuited for use with the new melter (due to seeing too much radiant heat from the furnace) but is probably not necessary with the coiled tube evaporator. It will be replaced with a water cooled nozzle as a melter feed device.

Intermediate-Level Waste Studies

A partial solution to solids problems involved in the treatment of an alkaline Purex waste condensate was obtained by diverting raw water from the condensate receiver tank. This raw water constituted about 25 percent of the total volume of the condensate stream and was temporarily introduced at the Purex plant as a result of changes made during construction of a new waste tank farm. Hardness in this volume of raw water caused premature ion exchange breakthrough, particularly of strontium.

Solids in the condensate still pose a problem. The solids are organic and may be largely bacteria. Bacteria counts vary from 10⁴ to 10⁶ bacteria per ml within the condensate receiver tank and process equipment. Centrifugation of 13 liters of condensate yielded about 40 mg of airdried solid. Inorganic impurities were minimal. The material exhibits no melting point and shows no immediate signs of dissolution or gas evolution when contacted with concentrated nitric, sulfuric, hydrochloric and hydrofluoric acid but dissolves readily in concentrated nitric and sulfuric acids at elevated temperatures. The material was insoluble in numerous standard organic solvents. Infrared spectra indicate a carbonyl grouping. The identification of the solid is not being pursued further.

Hypochlorite was added to the processing equipment and condensate transfer basin in an attempt to alleviate the bacterial problem. After one treatment promising effects were observed; however, additional treatments may be required to clean out the system completely.





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The electrodialysis unit was operated with acid condensate diluted 10:1 but with nitric acid added to the usual concentration of 0.02 N. Acid removal on a continuous basis was maintained at 75-80 percent at a flow rate of one liter per min_te through the diluting stream and 0.1 liter per minute through the concentrating stream. About 35 percent of the radioruthenium was removed under these conditions. On continuous recycling greater than 90 percent ruthenium removal was accomplished. Experiments are in progress treating undiluted acid condensate and following electrodialysis with ion exchange and charcoal adsorbents.

COLUMBIA RIVER SEDIMENT STUDIES

Columbia River Travel Times

The introduction of about two curies of I-131 into the Columbia River during a one-half hour period through a fuel element rupture provided a measurable tracer for this water so that travel times and perhaps dispersion rate in the river from the reactors to Vancouver, Washington, can be measured. U.S. Geological Survey personnel established sampling stations at bridges at Finley, Umatilla, Biggs Junction, The Dalles, Hood River and Vancouver. Samples taken $1 = \frac{1}{4}$ hours apart are collected so the arrival times and distribution curves can be determined. The I-131 is extracted in approximately 90 percent yield from the 3-gallon samples by three $CCl_{\frac{1}{4}}$ extractions after the iodine is oxidized with nitrite ion in sulfuric acid. The I-131 is back extracted, precipitated as silver iodide and counted 30 - 60 minutes in the Compton-Cancelling Spectrometer.

McNary Reservoir Sediments

A recent sediment core sample from the McNary reservoir was analyzed for sediment deposition rate by the isotope ratio technique and it was found that in the most recent six-month period the deposition rate was several times faster than the normal measured rate for older (deeper) sections. Since no immediately apparent reason seems to exist for such an increase, this observation may indicate that significant scouring at some later (high water) season removes part of the surface sediment layer, thus providing a lower average annual sediment deposition rate. Some further data which may provide information on this possibility are being analyzed.

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M-805

BIOLOGY AND MEDICINE - 06 PROGRAM

TERRESTRIAL ECOLOGY - EARTH SCIENCES

Hydrology and Geology

An 81-node, three-dimensional, resistance network was assembled and tested. The network had equal-grid spacing and was used to check errors associated with resistor tolerances and continuum simulation with discrete resistances. The combined potential error, in comparison with the analytical solution results, averaged about 0.1 percent; a maximum error of 0.4 percent was noted. One-percent precision resistors were used in network fabrication; thus, potential error appears to be about one-tenth resistor error. This information supports data from earlier studies in which two-dimensional networks were used.

The well-packer was used to attempt measurement of in-place permeability in well 699-47-35. Zones above and below the test section were packed-off to attempt sealing by pumping a bentonite slurry and a cement suspension into the formation. It appears that the packer did not operate properly during the grouting operation and ensuing test (either the top or bottom element failed to seat against the casing) due to the worn condition of the packer valve mandril. The procurement of a lightweight packer is being investigated (air operated) as a replacement for the present unit which is heavy, cumbersome and must be removed from the well and reset for each operation. A new packer should permit thorough investigation of this method for obtaining boundary permeability measurement.

Experience with the finite difference numerical method for solving transient flow problems involving imbibition shows that instabilities arise for short time intervals. These instabilities are possibly caused by a singularity at t = 0. The Boltzmann transformation overcomes this singularity. Philips' numerical procedure for one-dimensional problems is being programmed for the 7090 computer. This method, based on Boltzmann's transformation, will be used to evaluate the effect of fitting soils data with equations for the calculation of transient flow problems. The method may have value in obtaining an initial moisture distribution from which to start the finite difference calculations in two- and three-dimensions.

RADTOLOGICAL AND HEALTH CHEMISTRY

Background Reduction in X-Ray Scintillation Counting

Sensitivities obtainable by X-ray scintillation counting are limited by background radiation problems. Anti-coincidence shielding by means of a large sodium iodide well crystal was found to be effective in reducing the background by a factor of 3.5 in the 13-65 kev region and a factor of 2.5







in the 13-26 kev region. The 16-26 kev region corresponds to the Pu-239 X-ray counting region and this background reduction should result in a sensitivity improvement of a factor of about 2.5.

Fluorescence Analysis

Spectrofluorometer studies of uranium in aqueous solution showed that as little as 2×10^{-9} grams could be detected by this means. This is within a factor of 10 of the sensitivity of the standard calcium fluoride activator process. It seems likely that the aqueous method sensitivity could be increased, making this a simple, more useful, technique than the standard process. Quartz and nitrate ion are interfering materials which must be eliminated to obtain good sensitivity. The uranium emission is linear with concentration at least over the studied range of 1×10^{-6} to 2×10^{-9} grams.

Radiation-Induced Hemolysis and Volume Changes of Red Blood Cells

The Coulter Counter - 256 Channel Analyzer assembly was moved to the 325 Building and is now in operation. Calibration of the assembly with mulberry pollen (approximately 12 μ) and ragweed pollen (approximately $2 \downarrow \mu$) is currently being carried out, and it appears that volume changes of 50 μ^3 are detectable. This is about half the volume of the normal erythrocyte in its biconcave form, and about one-fifth the volume of a "sphered" erythrocyte. Considerable changes in the volumes of the cells were observed as a function of time after irradiation. Volumes of the modal species in the cell distributions differed by 200 μ^3 although this effect is just beginning to be explored. Hemolysis curves, using microspectrophotometry of the released hemoglobin as the detection method. showed that old (90-100 day) erythrocytes were more susceptible to radiation-induced hemolysis than a population of normal 0-120 day old) ones. Of course, in the present treatment procedure, the cells have not aged in their normal environment and so this observation must be amplified by future ones in order to make any useful statements.

ATMOSPHERIC RADIOACTIVITY AND FALLOUT

Aerosol Sampling Study

Subisokinetic sampling errors as a function of particle size were measured at an air flow rate corresponding to 9 miles per hour. For 5 micron particles and a sampling rate 4 percent of isokinetic, a sampling error in concentration of +50 percent is indicated. As the particle size increases to 20 microns, the greater inertia results in much greater collection than that which would be estimated from sampling flow rate alone. An air concentration 5.5 times higher than that judged by the isokinetic sample is obtained.







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The apparently anomalous higher errors at sampling flow rates 1/25 isokinetic than at 1/4 isokinetic are tentatively ascribed to effects due to a region at the very center of the test filters through which virtually no flow occurs. This no-flow region results from the inherent design of the filter support. This hypothesis will be tested using all data obtained to date. Obtaining a valid isokinetic sample at these higher flow rates is a serious problem in itself since a nozzle entry to the sample filter must be used. Possible entry losses and flow pattern distortion may be contributing to the unusual effect noted and will be further studied.

Iodine Studies

The removal of methyl iodide (a form in which radioiodine is found in irradiated fuels) from an air stream by several liquid scrubbing solutions was studied. Methyl iodide labeled with I-131 was prepared by the reaction of dimethyl sulfate and an aqueous solution of sodium iodide. The usual efficient scrubbers for elemental iodine such as caustic and sodium thiosulfate were confirmed to be very inefficient for the organic compound of iodine. A small quantity of silver nitrate in 95 percent ethyl alcohol proved to be highly efficient for removing methyl iodide. Pyridine showed an 85 percent efficiency.

ISOTOPES DEVELOPMENT - 08 PROGRAM

Acid-Side Promethium Purification Process

Six column runs were made during the month to explore the effect of various variables on the acid-side, HEDTA, promethium purification process, which continues to look very promising. HEDTA concentration, pH, flow rate, and elution distance were varied. Excellent separations were obtained in most cases, although further work will be required to optimize the flowsheet.

Source Forms and Encapsulation Studies

Preparative work continued on a variety of compounds and synthesis methods which are of interest as isotopic heat source materials. Included were the successful preparation of strontium fluoride, neodymium borate, samarium borate, cerium borate, strontium titanate, and neodymium metal (stand-in for promethium metal). Attempts were also made to prepare stoichiometric cesium borate, cesium sulfate and a quantity of cerium(III) oxide suitable for Dynapak compaction.

Seven hundred grams of strontium fluoride were prepared by treating SrCO₃ with aqueous HF followed by air drying and heating to 1200 C to remove residual moisture and HF. The product, which X-ray diffraction showed to









be pure SrF_2 (< 1 percent SrO), was loaded into a Dynapak can and welded shut without incident. (Great difficulty was experienced in welding an earlier batch of SrF_2 , which apparently still contained appreciable NP.) The can had not been compacted at month's end.

Neodymium, samarium and cerium borate were prepared by heating the respective oxides to 1000 C with H_3BO_3 . The products apparently have melting points ≥ 1500 C and could not be cast; however, pressing experiments suggest that they are probably amenable to compaction either by pressing and sintering or by Dynapak compaction. Initial attempts to prepare stoichiometric cesium borate, which would contain 87 weight percent cesium, were not successful (melts containing 50-60 percent cesium were prepared last month).

Several attempts were made to prepare cesium sulfate for trial Dynapak compaction. All attempts failed because of the formation of $CsHSO_{l_1}$, which decomposed to the pyrosulfate $(Cs_2S_2O_7)$ rather than to the sulfate when heated. Although the pyrosulfate decomposes to give the sulfate at temperatures of 800-900 C, it melts at about 400 C and remains liquid to the decomposition temperature. An attempt will be made, as soon as other work permits, to prepare $Cs_2SO_{l_1}$ from the nitrate or chloride by anion exchange. The power density from $Cs_2SO_{l_1}$ should be about equal to that from $CsCl_{l_2}$

Efforts to prepare a large quantity of cerous oxide by the method reported last month (reacting CeO_2 with graphite at 1000 C) gave a product analyzing $CeO_{1,75}$; as did hydrogen reduction at 1000 C. It is hypothesized that a higher temperature is required to carry out the reduction.

An apparently successful attempt was made to prepare di-strontium titanate (Sr_2TiO_4) for trial Dynapak compaction. Two moles of $SrCO_3$ were slurried with one mole of TiO_2 , filtered, dried, and ignited for 16 hours at 1000 C. The product was free of carbonate (no reaction with HNO_3), and X-ray diffraction showed only minor traces of $SrTiO_3$ and Sr_3TIO_5 . The material has been loaded into a can and will be compacted as soon as the Dynapak machine is available.

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Acting Manager Chemical Laboratory

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BIOLOGY LABORATORY

A. ORGANIZATION AND PERSONNEL

No significant changes occurred.

B. TECHNICAL ACTIVITIES

FISSIONABLE MATERIALS - 02 PROGRAM

Columnaris

The rainbow trout which have been held in water heated 4 F above river temperatures during the past summer were raised gradually from a temperature in the low fifties to a temperature of 64 F to determine whether columnaris organisms would increase within the trough at this time of year. Numbers of organisms did increase, as demonstrated by sampling over 20 columnaris organisms/ml of water flowing out of the troughs. There was no apparent increase in mortality within the troughs, presumably because the fish are resistant to the organism. Temperature will be maintained at 64 F in the trough and the course of columnaris followed through the winter months.

A large number of white fish and smaller numbers of scrap fish were obtained from the Priest Rapids area and sampled for presence of columnaris. Over 8% of the fish sampled contained columnaris on the gills. Although the river temperatures at this time were 42 F, one fish - a carp - had body lesions and these were heavily infected with columnaris organisms.

BIOLOGY AND MEDICINE - OG PROGRAM

METABOLISM, TOXICITY, AND TRANSFER OF RADIOACTIVE MATERIALS

Zìnc

Experiment to define radiation damage in rainbow trout from chronic ingestion of Zn^{65} was started on January 22, 1964. Four treatment groups with 50 fish each are being fed 0, 0.01, 0.1, and 1.0 μ c Zn^{65}/g fish Monday through Friday. On the average the fish weighed 98 g at start of test.

Trout fed single dose of 200 μ c Zn^{65} and killed at 211 days postadministration have on the average a body burden of 6.6 μ c. The GI tract continues to show a relatively high concentration and 3.6 μ c cr 54% of body burden is found in the GI tract.



Copper

The subcutaneous administration of an approximate LD_{50} level of 30 mg Cu/kg was found to decrease body weight of non-survivors more than an LD_{50} dose (850 r) of whole-body X-irradiation. Over a 30-may period the body weight of animals receiving a combined treatment of 1.2 LD_{50} of copper and 1/2 LD_{50} of X ray was depressed more than survivors of the LD₅₀ of X ray.

Hemoglobinuria was observed in all animals that died from a dose of 30 mg Cu/kg but occurred in only 2/5 survivors and 2/18 animals receiving the combined treatment. Urinary excration of hemoglobin accounted for only a minor portion of the nemoglobin loss.

Strontium

A group of over 50 crayfish each given $1 \ \text{Mc} \ \text{Sr}^{90} - \ \text{Y}^{90}/\text{week}$ for several weeks showed high accumulation in the rapidly growing gastrolith during the pre-molt period. It is generally believed little or no fixation of calcium in the exoskeleton occurs during the pre-molt period, but the deposition of $\ \text{Sr}^{90}$ in the exoskeleton observed before molt suggests significant exchange of calcium and strontium does occur between the exoskeleton and other tissues during this period.

The skeletal retention of $3r^{85}$ at ten days after oral administration of $3r^{85}$ TiO₃ was observed to be 0.15% of the administered dose for two miniature swine approximately $1\frac{1}{2}$ years old. A previous study with miniature swine of similar age has indicated the absorption and ten-day skeletal retention of $3r^{90}$ administered as $3rOl_2$ to be approximately 50 times greater than that observed in this study with $3rTiO_3$. Todane

Induce-131 vapor was generated in a chamber designed for exposing areas of pig skin. Approximately 385 cm² on the side of an anesthetized young miniature swine were excessed for 10 minutes to an air concentration of -2×10^{-4} uc/cc. Skin deposition totalled 18 µc, and the thyroid uptake of I¹³¹ reached a peak in six days at 0.9% of the original skin activity. Earlier studies with a solution of NaI applied to skin resulted in a 3-4% uptake in the thyroid gland. The difference in thyroid uptake may be due to the different forms of indime utilized. (Some moth-indications are being made in the apparatus to improve vapor generation, after which more pigs will be exposed.)

Radium

A review of data on seven female simisture swine that have developed a severe nephritis and died following intravenous injection of but Ac Ra²²⁶/kg reveals an earlier onset of symptoms and a shorter survival time for the older animals injected. Rather than being a strictly agerelated effect, it appears that the earlier appearance of the nephritis in the older animals may be related indirectly to the older sminals more



obese condition. If the injection dose were related to the animal's lean body mass (active body tissue), then the dose per kg of the lean body mass would have been greater in the older animals than in the young animals. If this is the case, then the age difference is not due to age per se but a dose effect relationship.

Neptunium

Neptunium-237 toxicity in sheep: two groups of three sheep, including rams, wethers and ewes, were given 3 mg/kg Np²³⁷NO₃ in Na citrate solution at pH 4-4.5 intravenously. Liver function tests using I¹³¹-labeled rose bengal were performed at two-day intervals. All animals died 2-5 days following Np²³⁷ administration. All sheep showed gross degenerative changes in the liver and kidneys, serofibrinous exudates in the pleural and peritoneal cavities, and mild to severe icterus. Less consistent findings included subcutaneous, subendocardial and intestinal hemorrhages, and pale skeletal muscles. Both groups were given freshly prepared Np²³⁷ solution, while an earlier group which had shown 100% survival was given the same amount of Np²³⁷ in a solution that was prepared 3-4 hours before injection. This may indicate a rapid change in the chemical form of the Np²³⁷ following preparation. (Serial spectrophotometric examinations of sample Np²³⁷ solutions are planned to determine valence changes of the Np²³⁷ with time.)

Plutonium

Intestinal perfusion studies indicate that Pu^{238} excreted in the bile is influenced by DTPA therapy in the same manner as Pu^{239} . Techniques employing the measurement of the biliary excretion of plutonium following perfusion of various segments of the intestine with DTPA and TTHA may be used to determine the site of absorption of these compounds.

Studies involving biliary cannulation in animals receiving Pu^{239} followed by DTPA treatment showed that normal liver function as evidenced by optimum bile flow is necessary to determine the partition of Pu between feces and bile. Approximately 69% of the Pu²³⁹ excreted into the gastrointestinal tract comes by way of the bile before DTPA treatment and 76% after DTPA. Under the same conditions urinary excretion of Pu increases 20%.

Inhalation Studies

One dog died four years after a single inhalation exposure to $Pu^{239}O_2$. The lung burden at death was 0.5 µc. Gross Lesions were confined to lungs and bronchial lymph nodes. The severe fibrosis of the pulmonary tissues was the cause of death. Neoplasia was not observed. Nine male beagle dogs were exposed to aerosols of $Ru^{106}O_2$ particles that were calcined at >410 C.

HW-80560

Two samples of Pu²³⁹O₂ dust were received from the British Atomic Energy Authority, Aldermaston, England, for use in animal inhalation studies. One sample was prepared by oxidation of the delta metal at 120 C, and the other at 450 C. The dusts are products of aerosol studies in which they were carefully sized and characterized. Inhalation studies comparing the friable dust (oxidized at 120 C) with more stable dusts, including PuO₂ calcined locally at 800 C will be done in dogs.

Nine male dogs were exposed to calcined RuO_2 aerosols. Six of the dogs were given 30 μ c Ru106 and the other three, stable Ru. Whole body-retention, excretion, tissue distribution, and biologic effects will be examined to compare with early mouse data.

The 132 dog colony was increased by 22 puppies. In order to have dogs for experiments beginning in the spring of 1965, a goal of 50 additional pups has been set for July 1, 1964. Because of the holdup in starting construction of an additional 60 dog runs, temporary measures to house the dogs will be taken. This includes dividing 24 existing runs and erecting 20 temporary runs with galvanized fence panels on the concrete area behind 144-F building. Failure to do this means no dogs for experiments for at least a year after construction of the new 60 run area is completed. Neither the research program nor our careers can tolerate that kind of delay.

Secondary Disease

Preliminary experiments to test toxicity and immune response suppression by anti-cancer agents such as phenylalanine nitrogen mustard (melphalan) and methotrexate, as well as epsilon-amino caproic acid (EACA), a drug used to diminish skin graft rejections, showed that only melphalan was effective in lowering HA titers. It also produced a body weight loss, a decrease in hematocrit values, and, at higher concentrations, a depression of WBC values and lethal toxicity within 7-14 days. At autopsy atrophied lymphoid tissue and intestinal damage similar to that produced by lethal amounts of radiation was evident. The effects of two other nitrogen mustard compounds, bisulfan and chlorambucile, were comparable but HA titers have not been completed.

Radioprotective Agents

The study of the radioprotective effect of diethyldithiocarbamate (DDC) was completed. Under the test conditions (750 r and 1000 r whole-body X-irradiation) levels of DDC up to 500 mg/kg showed no radioprotective effect when administered either 20 minutes before or 3 minutes after X-irradiation, in fact, a synergistic effect may be indicated.

Gastrointestinal Radiation Injury

Mating of rats surviving a neutron exposure was continued for three consecutive periods. In irradiated male x control female matings, 2/10 males exposed to 150 rads showed recovery of reproductive capacity. Litter sizes were normal. In control male x irradiated female matings, fecundity decreased from 9/30 to 6/26 to 2/27 compared to control x control values of 10/11, 13/15, and 13/16 in the three matings. Litter sizes were also decreased.

Plant Nutrition

As a consequence of rate studies of the uptake of iodide by plants, it is possible to describe uptake by the equation $C_T = K C_S + b$ where C_T is the tissue concentration intercept constant. The constant K (or slope) does appear to change slightly with time, but over short intervals the value approximates 0.8 for the ten hours over which the study was carried. The efficiency of absorption (C_T/C_S) changes with both time and concentration. The efficiency is maximum at 0.1 and 1.0 μ M KI levels. At these concentrations for the time studied, the absorption rate was unchanged; as the concentration increased or decreased the absorption rate fell. The presence of these relationships suggests that there are limiting sites on the membrane which provide a rate limiting process. At low concentrations (0.01 μ M) the solution is so dilute that the absorption is dependent on aqueous diffusion. At higher concentrations the transfer mechanism at the membrane or within the root becomes rate limiting.

In a separate experiment in which plants were pre-treated with iodide for varying periods of time it appears that the content of iodide within the plant does not affect the absorption rate into the plant. Uptake of iodide is relatively constant during light and dark periods, whereas water absorption is highly light dependent.

To obtain more information on mechanisms involved in the uptake by plants of iodide gas, coleus leaves were exposed on one surface to $I_2^{1/3}$ and the air on the opposite surface of the leaf was recovered and tested for the presence of $I^{1/3}$. Iodine-131 was detected in the air on the lower surfaces adjacent to the lower surface of the leaf and the first approximation of the permeability constant of the intact coleus leaf to $I^{1/31}$ gas is in the order of magnitude 0.1 cm /hr. Coleus has no stomatal openings on the upper leaf surface so that passage of $I^{1/31}$ across the leaf would appear to be due to passage through the waxy outer cuticle of the upper surface. In reports from other laboratories it has previously been assumed that penetration of $I^{1/3-1}$ into leaves was dependent on passage through the stomatal openings. While the present information is tentative, it does raise doubts regarding the interpretation uptake of $I^{1/31}$ by plants.

The loss of I_2^{131} from plants was shown in field tests to occur by a desorption of I131 from the leaf. To define more carefully some of the factors associated with this desorption, plants have been exposed to I_2^{131} and then held in the laboratory at different temperatures while the amount of I131 lost to the atmosphere was measured. Higher temperatures accelerate the loss of I^{131} from vegetation, but this accelerated removal persists only during the first 24 hours. An observed transfer of the I^{131} to the soil area of the plant is as yet uncertain, and further tests will be required to determine whether this was a translocation within the plant or a problem of external transfer of I^{131} from the leaves to the soil area.

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Columbia River Ecology

In plankton samples, average concentrations of Zn^{65} and Cr^{51}/g dry organic matter increased in December, while P³² concentrations were unchanged. Values for December were 460 nc, 2500 nc, and 89 nc, per g for the respective isotopes. Daily transport in November amcunted to about 40 tons dry weight of plankton per day. In terms of activity transported by the plankton, this amcunted to approximately 0.4 c Zn^{65} , 0.07 c P³², and 2.3 c Cr^{51} per day.

In periphyton samples, the amount of chlorophyll observed in early December was approximately 1/5 that of the November average. Total dry weight and the weight of dry organic matter per unit substrate area increased by approximately 67% during December. Concentrations of Zn^{65} , Cr^{51} , and P^{32}/g of dry organic matter in December differed little from the November averages, being 250, 1230, and 140 nc/g dry organic matter, respectively. It appears that all measurements for periphyton are at their minimum yearly values at this time.

Terrestrial Ecology

Populations of adult darkling beetles were measured in sagebrush and greasewood combunities during the last year. During the period of mass emergence of beetles in September and November there were minimal populations of 6-7 beetles per square meter in both habitats. Mineral analyses and biomass data are being developed to determine the extent of mineral transfer through insect portions of the communities.

Vegetational collections are continuing in the Blue Mountain and Cascade areas. Samples of three elk and two deer were obtained in the Cascades through cooperation of the Washington State Department of Game. Snow and water samples are being obtained at the sites adjacent to fallout collectors in these areas.

Chromosome Studies

Peripheral leukocytes of miniature swine were successfully cultured in in vitro systems and yielded adequate numbers of cells that are suitable for detailed micromorphological study of the chromosomes. Studies are in progress to determine if this test system may provide a sensitive means of detecting damage in animals with body burdens of bone-seeking radionuclides. Preliminary analysis of the microphotographs of the chromosomes of the miniature pig indicate a diploid chromosome number the same as that of the domestic pig (2N=38). (It is anticipated that several months will be required to complete culturing and analysis of microphotographs of chromosomes from representative irradiated and non-irradiated animals.)





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Biotelemetry

A body temperature telemetering system developed by Nucleonic Instrumentation was calibrated and tested in sheep. Accuracy of the system is within acceptable clinical standards. Refinements are being made in the system to permit continuous monitoring and print-out of results.

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BIOLOGY LABORATORY

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TECHNICAL INTERCHANGE DATA BIOLOGY LABORATORY

I. Speeches Presented

a. Papers Presented at Society Meetings and Symposia

Sullivan, M. F. Bile salt influence in intestinal radiation injury. Symposium on Intestinal Function. San Juan, Puerto Rico, January 15-18, 1964.

Hungate, F. P. Life in Greece. Health Physics Society, Richland, Washington, January 31, 1964.

b. Seminars (Off-Site and Local)

Park, J. F. Biclogical effects of inhaled radioactive particles. U.S. Naval Research Unit, Washington State University, Pullman, Wash. January 6, 1964.

- c. Seminars (Biclogy)
 - Park, J. F. Chronic toxicity of inhaled Pu in dogs. January 8, 1964.

Liu, D.H.W. Toxicity of industrial chemicals to fish. January 15, 1964

Eastlick, H. L., Department of Zoology, Washington State University, Pullman, Washington (Exchange Seminar Program). Methyl cholanthrene induction of neoplasms in chickens. January 17, 1964.

Wilson, D. O. Deposition of I¹³¹ on plants and soil under simulated fallout conditions. January 22, 1964.

Ballcu, J. E. Plutonium deposition in soft tissues. January 29, 1964.

d. Miscellaneous

Hanson, W. C. Radicecological studies of northern Alaska. Richland Church of the Nazerene Men's Group. January 6, 1964.

II. Articles Published

a. HW Documents

None

b. Open Literature

Thompson, R. C. 1963. Factors and conditions modifying the absorption and retention of chronically ingested radiostrontium. In Transfer of Calcium and Strontium Across Biological Membranes, Academic Press, Inc., New York, p. 393-404.

 b. Open Literature (Continued) 1963.
Eberhardt, L. L. /Problems in ecological sampling. Northwest Science 37: 144-154.

III. Visits and Visitors

- a. Visits to Hanford
 - Gordon H. Orians and class in advanced ecology from the University of Washington, Seattle, toured the Biology facilities on January 2-3 and discussed various aspects of ecological work with Dr. F. P. Hungate and associates.
 - W. E. McCormick of the BF Goodrich Co., Akron, Ohio, and Presidentelect of the AIHA discussed the AIHA monograph with Dr. Bustad on January 2.
 - B. A. Butt, D. O. Hathaway, and J. F. Howell of the Entomology Research Division, USDA, Yakima, used the 250 KV X-ray machine on codling moths and consulted with Dr. Hungate, January 7.
 - H. L. Eastlick, Zoology Department, Washington State University, Pullman, presented a seminar and toured facilities with E.M. Uyeki. (January 17)
 - M. Asa, West Coast Scientific Cc., Oakland, California. Demonstrate equipment. (January 27)
 - Clinton Powell, NIH, Bethesda, Md. Tour with H. M. Parker on Jan. 28.
 - Drs. George Duvall and Smith Murphy, Physics Department, WSU, Pullman, discuss mutual research interests with Drs. Hungate and ^Bustad. (January 29)
- b. Visits Off-Site

1/2-3	-	в.0). Stuart discussed the Hanford Symposium and radioactive
•			aerosol generation and sampling with Drs. Stannard,
			Casarett, and Morrow at the University of Rochester.
1/6	-	J.	F. Park addressed the Navy Research Unit at WSU, Pullman.
1/6-7	-	С.	E. Cushing and W. H. Rickard travelled to the Wooten
, .			Game Pange near Dayton for fallout sampling.
1/7-10	-	D.	G. Watson and L. L. Eberhardt collected samples for
•			fallout studies in Yakima and Packwood.
1/9-28	-	1.	C. Hanson was in Alaska on the Eskimo counting program.
1/17	-	W.	H. Rickard and L. L. Eberhardt went to Packwood for
			fallcut sampling.
1/24	-	I.	Newcomb collected samples at the Wooten Game Range.
1/13	-	Μ.	F. Sullivan discussed facilities at Los Alamos Scientific Lab.
1/15-18	-	Μ.	F. Sullivan presented talk at Symposium in San Juan, Puerto
1/20	-		Rico, and visited Argonne National Lab on return.

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 b. Visits Off-Site (continued)
1/28-30 - W. J. Bair and H. A. Kornberg attended the Conference on Radiochemical Toxicity Problems at the University of Chicago.
1/30 - M. P. Fujihara and C. O'Malley collected samples at Priest Rapids Dam.

IV. Achievements

None

V. Honors and "ecognitions

None

VI. Professional Group or Organization "ssignments

None



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HU-80560

APPLIED MATHEMATICS OPERATION

MONTHLY REPORT - JANUARY, 1964

ORGANIZATION AND PERSONNEL

Mr. H. D. Huber joined the group on January 2, 1964, as a statistician.

OPERATIONS RESEARCH ACTIVITIES

Development of a PERT type analysis in support of the Containment Systems Experiment was continued. As plans progress, time and dollar estimates will become available and the network can be completed. A first completed PERT network should be available by the first week of February.

Work continues on the development of an effective information system associated with IPD maintenance practices. Real-time tests of the usefulness of certain subsystems are being made as an experimental evaluation of broad applicability of the over-all system.

In connection with a proposed AEC project on the economic-biological interactions consequent on nuclear war, meetings and consultations were held with AEC-TAB personnel and others from NRDL and Rand.

In connection with current AEC-GE efforts at diversification and community development for the Tri-Cities, work has begun to extend the previous study and modeling of the Tri-City Area Economic Structure.

STATISTICAL AND MATHEMATICAL ACTIVITIES FOR OTHER HAPO COMPONENTS

N-Reactor Department

Statistical procedures which are to be part of a program specification being developed as an improved method of apportioning total production of a reactor were completed and submitted.

A previously proposed experimental design relative to a forthcoming study of temperature variation in pre-extrusion ovens was modified. The resulting quasi-factorial lattice design appears to accommodate the present set of test objectives and restrictions.

A linear model was developed for the analysis of a set of data relevant to heat treatment effects on the geometry of fuels.





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A data collection procedure was designed leading to a discriminant analysis approach to precise estimates of the "true" levels of "standard" units.

Irradiation Processing Department

An analysis of core size, type of blank, type of quench, delay time, and quench temperature in the heat treatment of dingots and ingots on the preirradiation warp of fuel element cores is being completed. Comparisons are also being made between ingots and dingots and between types of (d)ingots.

The analysis of PT-546 is being continued which compares the postirradiation incremental changes in the dimensional characteristics of fuel elements canned by the hot-die-sizing and AlSi canning processes.

A test to determine the effect of the spire-core and can-core annuli on the AlSi bonding layer is being analyzed. The test consists essentially of determining which annuli thicknesses give the best bond integrity measurements.

Assistance is being given in a study of the reliability of the Panellit gauge scram system. Particular emphasis is being placed on the system logic, data collection, and component and system reliability.

An analysis is being made of an experiment to determine whether the life of a Diversey cleaner can be prolonged by control of the pH factor.

A study is being made to determine how much certain geometries affect the counting rate in an alpha counter.

A test to assess the effect of plating procedures upon bond strength measurements of hot-die-sized fuel elements is being analyzed. The test will also compare the effects of different size tubings.

Assistance was given in determining the arrangements of six-inch and ten-inch fuel elements in tubes which would be most effective in assessing the incremental warp and dimensional changes for fuel elements of these sizes.

Results from PT-572A, which studies the behavior of self-supported fuel elements in smooth process tubes, are being analyzed.

Chemical Processing Department

The required demonstration of weapon component chemical and isotopic specifications was made for both "A" line and "C" line components for the fourth quarter of CY-1963.











The apparent difference in yield strength measurement between RFP and HAPO has practically disappeared in recent measurement comparisons, with HAPO's values increasing to those of RFP.

An apparent decrease in plutonium content of weapon components occurred for material to be shipped during first quarter of 1964. Cum-sum charts (daily averages two components on "C" line and four components on "A" line) are being maintained to aid in locating specific times of process average shifts.

The minimum variance inventory method for the control of Z-Plant component MUF is being applied by NMM. The PPO operation, except for an isolated onemonth period, has shown a highly consistent behavior using total receipts as the basis. However, fabrication does not appear to be as straight forward a system.

An EDPM program, based on the method of "direct search", which evaluates parameters in nonlinear mathematical models has been written, debugged, and tested on several small models. It is anticipated that this program will be used to investigate proposed models arising from several industrial engineering studies.

STATISTICAL AND MATHEMATICAL ACTIVITIES WITHIN HL

2000 Program

Continued assistance was provided on formulating and fitting mathematical models of radionuclide retention in a reactor effluent stream.

The EDPM program which is to provide numerical data for the theoreticallydeveloped "shoe-box" model of ground water flow is now in the debugging phase.

Discussions were held with regard to the possibility of developing a technique for studying flow in heterogeneous porous media analogous to the hydrodynamical concept of "streak functions".

Several problems of heat transfer were treated in connection with studies being made on underground waste-storage facilities.

Further work was done on the debugging of boundary value approach to the solution to the system of simultaneous nonlinear differential equations expressing mass transfer in a C-type extraction column as a function of column position.









The total amount of isotopic deposition on certain tubes is being estimated by fitting observed deposition rates per unit tube length with harmonic-type functions and integrating between appropriate limits.

Further work is being done on fitting density functions to observed frequency distributions of particle sizes. The previously fitted Weibull distributions have proved unsatisfactory in several respects.

3000 Program

Test tapes were successfully run on the prototype numerically controlled Sheffield rotary contour gauge. During the next month, it is anticipated that numerous magnetic tapes will be prepared, each designed to test specific response characteristics of the functioning mechanisms.

4000 Program

Study continued on the problem of obtaining a quantitative geometrical description of a proposed experimental reactor.

Mathematical analyses have been completed on a model to describe the expected behavior in a carbon burnout experiment. This model will now be studied by obtaining solutions with the aid of an EDPM program.

Additional understanding of the complex process of particle packing has been gained by the successful analysis of the geometry of multiple spirals - a phenomenon which has been repeatedly observed when spheres are stored in cylindrical containers.

Discussions were held with Dr. Moises Levy of the University of Pennsylvania on a mathematical model which has been developed in an attempt to describe wave transmission in a visco-elastic material.

Work continued on a formal Hanford report describing the quantitative metallographic techniques developed during the past several years for estimating the properties of the second phase spherical particles embedded in a matrix. The machine language program developed for analyzing data collected on the Ziess particle size analyzer is being modified to handle several methods of sample replication prior to electron microscopy and to present more meaningful graphical displays of both the raw data and estimated matrix distributional characteristics.

Statistical analysis was begun of quantitative metallographic data obtained on the Ziess particle size analyzer of second phase particles in dilutely alloyed dingot uranium. The evaluation should help determine the dependency of density and











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volume fraction of these second phase particles on alloy composition, the time and temperature of solutionizing anneal, and the time and temperature of precipitation anneal.

A study was begun to review a "fretting corrosion" problem associated with the PRTR. The immediate objective of this effort is the development of a subsampling scheme (relative to 85 pressure tubes), in accord with the scheduled outage periods, which will optimize certain critical information.

5000 Program

A new method of calculating channel group standard probabilities for use in the GEM program of the IRA system was used to recalculate these probabilities for a number of the standards used in the system.

Work continued on the calculation of the power function of the Poisson index statistic used to check stability of counting instruments. Power function calculations were completed for the alternative of a single shift during the counting periods assuming a fixed denominator and a normal approximation for the Poisson variables.

The formal Hanford report HW-76279, "Fixed Time Estimation of Counting Rates with Background Corrections" was released for unrestricted distribution during January.

A subroutine to edit the spectra which are input to the IRA 335 calculation pass was debugged and incorporated into the IRA system. An alternative index to the chi-square ratio is being programmed for the GEM calculation which will take a count of the relationship between standard and sample estimates.

Work continued on the actinide research program. Considerable interest in crystal indexing techniques developed at Hanford has arisen from offsite installations.

6000 Program

The final data obtained from the EDPM "triangular" diffusion model have been submitted to the program originator for evaluation and study.

A statistical analysis was initiated of adult progeny data from neutron exposed <u>Tribolium castaneum</u> which were three weeks old. The purpose of the study is to investigate the effects that neutron exposure of one member or both members of a mating combination and temperature have on the number of adult progeny produced.



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Data from a study to investigate the effect of treatment with DTPA has on the removal of Np²³⁹ and Pu²³⁹ from various rat tissues is being analyzed. The effect of Np²³⁹ on the removal of Pu²³⁹ is also being studied.

Statistical analysis of data from a study to investigate and compare the beetle populations of sagebrush and greasewood communities was started. The beetles of primary interest are the <u>Pelecyphorous</u> and <u>Stenomorpha</u>.

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REACTOR PROGRAM - 04 PROGRAM

PLUTONIUM UTILIZATION PROGRAM

Fueling Overmoderated Reactors

The effects of mixing UO₂ (natural enrichment) and ThO₂ in the same fuel channel in a reactor with a soft neutron spectrum (that is, wellmoderated) were calculated using $U^{2}35$, $U^{2}33$, and plutonium as enrichments. For comparison, a slightly-enriched uranium case (with no thorium present) was also calculated for the same neutron spectrum. All of the burn up calculations were made with the MELEAGER code, and fuel costs were calculated by the QUICK code which also determined the minimum fuel cost, optimum enrichment, and fuel exposure.

A significant effect of combining these fuels is to increase the fertility of the reactor. Since both U^{238} and thorium cross sections are highly dependent upon resonance absorption, and since the resonances do not overlap significantly, the resonance captures in each material can occur without reducing absorptions in the other. Thus in a reactor that is well moderated this can effect a reduction in "p", the resonance escape probability, without adjusting the lattice.

The resulting reduction in the fuel cost for the mixed system in a reactor with a soft spectrum is listed in Table I. This mixed system also achieves some reduction in fuel cost over pure thorium systems because of natural uranium serving as a limited but low cost neutron source. Of course, very slightly moderated reactors could possibly not tolerate the simultaneous fertility of $U^{2}3^{8}$ and thorium to use the natural uranium as an economic driver. Thus, the lowering of fuel cost from using a mixture of thorium with natural uranium may be limited to reactors that are otherwise considered overmoderated.

There are a number of ramifications in making economic comparisons of $U^{2}33$, $U^{2}35$, and plutonium fueling systems because the relative prices of these isotopes may not provide a "closed" system. For example, plutonium priced very high would yield low fuel costs for the plutonium producer cycles, but this system is unreal in the sense that no one would buy the plutonium at this price because at this price, plutonium enriched systems have unfavorably high fuel costs. In most paper studies involving just plutonium this paradox is avoided by using the "indifference" price for plutonium. This is the price of plutonium that yields the same fuel cost whether one is a producer or a user of plutonium. With $U^{2}33$ in the picture a far more complex situation is involved, since $U^{2}33$ is often priced relative to uranium cascade produced $U^{2}35$ as an alternate to $U^{2}33$

TABLE I

MINIMUM FUEL COSTS USING VARIOUS FUELS IN A WELL-MODERATED REACTOR

Economic Bases For All Cases:

Plutonium priced at \$10/gram fissile.
U²33 at \$14/gram fissile (a).

- 3) Jacketing at \$30/pound fuel.

Minimum	Fuel	Cost,	Mills	/kwhe
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		MILLS/ KWHE					
Fissile Fuel	Slightly Enriched Uranium	Natural Uranium	Thorium	Natural U + Thorium (c)			
u ² 35	1.00		1.36	0.872			
U ² 33		1.13	1.14	0.822			
Plutonium(b)	1.05	1.15	0.902			

- (a) Reactor mixtures of U^{233} contain some U^{234} and υ²35.
- (b) Plutonium Composition 76% Pu^{239} , 18% Pu^{240} , 5% Pu^{241} , and 1% Pu^{242} .
- (c) 50% natural uranium and 50% thorium.

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enrichment for thorium. This generally yields U^{233} values greater than the price of U^{235} , as well as fuel costs for these reactors greater than if slightly enriched uranium were employed and plutonium were sold at its indifference price. For example, HW-75007 (page 12.11) indicates a U^{233} value of \$13.90/gram and a corresponding fuel cost of 2.6 mills/kwh_e for the APWR, compared with a plutonium value of \$12.10/gram fissile and a fuel cost of 2.15 mills/kwh.

To provide additional perspective, new calculations have been completed for six fueling schemes in the several reactor simulations studied in HW-75007 (and described in HW-72217), and for the well moderated reactor just described. The schemes involve U^{233} , U^{235} , and plutonium enrichment of U^{238} and thorium. Table II lists these resulting fuel costs for U^{233} priced at \$14/gram fissile and plutonium at \$10/gram fissile. Based on this comparison it appears that U^{233} would not be used interchangably in these machines unless its price were lowered, since essentially all of the schemes using U^{233} enrichment have higher fuel costs than the others, except for U^{235} enriched thorium. A U^{233} -thorium breeder of reasonably short doubling time such as the molten salt designs being analyzed by ORNL may be able to use \$14/gram U^{233} and provide a "closed" cycle, but the over-all power costs from such a system must be competitive with other reactor types to capture a market.

Study of Conversion of Small Production Reactors to Power Reactors

Initial study of converting the Hanford small production reactors to power reactors without modification of the existing lattice has shown that the lowest fuel costs for a given set of economic parameters should be obtained using slightly enriched uranium metal as the fuel.

Other studies involving alternate loading schemes for the small production reactors were begun using the ALTHAEA code to indicate neutron sharing for complex loads. The first load studied used plutonium-aluminum driver (76% Pu-239, 18% Pu-240, 5% Pu-241, 1% Pu-242; 4.5% plutonium in aluminum) and thoria target. Alternate tubes of driver and target were assumed. In the central zone, the flux in the target was 5% higher than in the adjoining driver due to the larger macroscopic cross section of the plutonium in the driver. The breeding ratio was 0.73 gram of Pa²³³ plus U²³³ produced per gram of plutonium burned. This loading produced about 0.4 gram of higher isotopes per kilogram of plutonium charged to the reactor.

Seed Blanket Studies

To aid making generalizations, a representation of an idealized blanket with constant reactivity as a function of burn up was prepared. Figure 1 illustrates the specific power level as a function of radial distance from

TABLE II

COMPUTED MINIMIZED FUEL COSTS FOR SEVERAL REACTOR SIMULATIONS

USING SIX DIFFERENT FUELING SYSTEMS

Jacketing Costs, \$30/pound fuel Current US-AEC Price Schedule--U²³³ \$14/gram fissile** and Plutonium \$10/gram fissile, Fuel Cost mills/kwh_e

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	Fueling Schemes					
Reactor Simulations	U-235 in Thorium	U-233**in Thorium	Plutonium* in Thorium	U-233** in Natural Uranium	Plutonium* in Natural Uranium	Slightly Enriched Uranium
Pressurized Water Simulation (Stainless Steel Jacketing	1.95	1.71	1.46	1.53	1.34	1.55
Graphite Gas-Cooled Simulation (Low Temperature)	1.54	1.33	1.32	1.41	1.28	1.23
Boiling Water Simulation (Zirconium Jacketing)	1.52	1.26	1.20	1.19	1.10	1.13
Heavy Water Simulation (Insufficiently moderated for natural UC ₂ operation)	1.41	1.17	1.07	1.07	0.99	0.98
Well-Moderated Reactor with zirconium tubes)	1.36	1.15	1.14	1.13	1.05	1.00
	Notes: * Plu	tonium Compo	osition 76% Pu	239, 18% Pu ²⁴⁰	, 5% Pu ²⁴¹ , a	and 1% Pu^{2h2}

** Mixtures of U^{233} contain some U^{234} and U^{235}



the center of a 10 cm radius seed in a cylindrical seed-blanket module for various values of k_b, the infinite multiplication factor of the blanket. The computer code was set up to limit the maximum specific power at any point in the reactor to 200 watts/cc. Two curves are given for the blanket with reactivity near 1.0 to illustrate the influence of macroscopic cross section of the seed. For Curve 1, the macroscopic cross section in the seed is 1.4 times that of the blanket and the maximum specific power in the reactor occurs on the seed side of the seed-blanket interface. For Curve 2, the macroscopic cross section in the seed is only 80% of the blanket value and the maximum specific power in the reactor occurs on the blanket side of the seed-blanket interface. Although other seed-blanket configurations have not been run, one may infer from the data that the percent of the power produced in the blanket as the thickness of the blanket varies would be essentially as shown in Figure 2 when the seed macroscopic cross section is 1.4 times that of the blanket. This corresponds to about 0.14 gram of U-235 per cubic centimeter of fuel, and under these conditions the seed radius must be about 10 centimeters in order for the seed to be critical when surrounded by blanket with low reactivity. From these curves, it is evident that for any given blanket reactivity, there is a definite limit to the fraction of the power that can be produced in the blanket and, in fact, eventually, increases in blanket thickness decrease power generated in the blanket.

Figure 3 illustrates the fraction of the reactor power that can be generated in a blanket as a function of the fractional reactor volume occupied by blanket. It should be noted that the larger the reactivity of the blanket, the larger the maximum fraction of the reactor power that can be generated in the blanket. Also, for higher blanket reactivities the maximum occurs at a larger fraction of the blanket reactor volume. However, the total reactor size is increased more rapidly than power output as blanket is added. Figure 4 shows the ratio of the power produced in the blanket to that produced in the seed as the volume of the blanket is increased. This illustrates the limitation in the gain in total reactor power as reactor size is increased by addition of blanket material. For example, if the blanket reactivity is about 0.9 adding seven volumes of blanket to one volume of seed will add two units of power. Figure 5 illustrates the decrease in average reactor specific power as the blanket volume is increased. Using the same assumptions, the specific power for a reactor using graded enrichment could be maintained at 200 watts per cubic centimeter.

Plutonium Fuels

In fuels containing U^{238} or $\mathbb{T}h^{232}$, $\mathbb{P}u^{241}$ is generally considered superior to $\mathbb{P}u^{239}$ as an enrichment material. This is because of its lower \mathcal{A} , * higher absorption cross section, greater energy yield per fission, and higher

^{*} **d** = ratio of number of neutrons absorbed by capture to the number absorbed by fission.



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average neutron yield per fission. However, in fuels that contain only plutonium and which rely on Pu^{240} as the fertile material in the fuel, Pu^{239} is superior from the standpoint that neutron captures in Pu^{239} produce the fertile isotope Pu^{240} , whereas capture in Pu^{241} produces the parasite Pu^{242} . The importance of this fact is that greater exposures are obtained with plutonium fuels with a low Pu^{241}/Pu^{239} atom ratio. It is, of course, also important to have a fissile-to-fertile atom ratio low enough so that sufficient fertile material is present.

In Figure 6 exposure per initial gram fissile is plotted as a function of Pu^{241}/Pu^{239} atom ratio. Maximum exposure is obtained at Pu^{241} atoms/ Pu^{239} atoms = 0. The fissile-to-fertile atom ratio was constant and equaled 2.33 (corresponding to plutonium containing 30 percent Pu^{240}).

In Figure 7 the fissile-to-fertile atom ratio is 1.0 -- corresponding to plutonium batches containing 50 percent Pu^{240} but with the Pu^{241} atoms/ Pu^{239} atoms varied as for the data plotted in Figure 6.

Fuel cycle costs were 1.86 mills/kwh_e for the ratio Pu^{241} atom/Pu²³⁹ atom = 0 in Figure 6 where the exposure was 0.965 Mwd/cc. Fuel cycle costs were 1.49 mills/kwh_e for the maximum exposure point of Figure 7.

Standard economic parameters of \$0.61/cc FEFJ, 12-1/2% economic interest, 4-3/4% AEC interest, 33% thermal-to-electrical conversion efficiency, \$10.24/gram fissile plutonium price, \$0.21/cc separations cost were used. The effective fuel density (plutonium density in the fuel) was 1 gram/cc.

Successive Plutonium Recycle

A set of simple equations for calculating plutonium value as a function of composition and reactor parameters has been completed using a statistics code applied to the plutonium value calculated for the various reactor types appearing in the document <u>Pu and U233 Values Computed for Successive</u> Recycle in Five Simulated Reactor Types, HW-76195.

The values were first fitted for a single reactor type and fueling scheme using only the plutonium composition. The resulting general equations usually contained a constant plus some coefficient which was multiplied by the Pu^{242} and/or the Pu^{240} percent composition in the discharged fuel. An example equation for a PWR is shown below.

Pu Value, $\frac{1}{242}$ = $\frac{100}{100}$ (percent Pu²⁴²)

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EXPOSURE PER INITIAL GRAM FISSILE AS A FUNCTION OF THE Pu²⁴¹ to Pu²³⁹ RATIO FOR A FIXED FISSILE-TO-FERTILE RATIO

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Each of the many equations was checked for statistical validity and accuracy of fit, which led to selection of the best equation. The best equation usually changed with fueling mode, but was the same for any reactor type with the same fueling mode.

The coefficients from the equations for specific reactors were then fitted against the reactor parameters. These parameters included moderator index (SDPV), nonfuel cross section (SNF), neutron temperature, and a term mainly dependent on fuel element diameter (SCA). Again many equations were tried and checked for statistical validity and, again, good fits were obtained over a range of reactor types for a given fueling mode.

Examples of these results are shown in Table II for self-produced plutonium recycle with graded irradiation. The constant term appearing in the equation on Table II is modified by SNF (flux weighted macroscopic cross section of the nonfuel materials) and SDPV, (the moderator index prepared from the moderator slowing down power; moderator volume, and fuel volume). As the SNF term increases, plutonium value increases. This is probably due to plutonium's large cross section which allows it to compete more easily for neutrons than U^{235} in a reactor with a high nonfuel cross section. As SDPV becomes smaller, the neutron spectrum hardens and the value of the constant increases. This indicates that the value of Pu^{239} , Pu^{240} , and Pu^{241} as a unit increases in value as the spectrum hardens. (It is suspected that this is due to the increase in Pu^{241} production.) The second part of the equation containing Pu^{242} reduces the plutonium value as Pu^{242} concentration increases. This is because Pu^{242} is essentially a parasite in thermal reactor fuel. As SDPV decreases, the spectrum becomes harder and the Pu^{242} cross section increases which increases its influence as a parasite, further reducing the value.

The ninth column in Table JT, Iteration Error, is the allowed error in the iterative process of calculating the plutonium values which were fitted. In some cases this error is larger than the fitting error.

Plutonium Recycle and Conservation

The investigation into the rate at which demestic uranium resources are being depleted is being conducted in two steps. First, the YUKON code has been developed to determine the total ore requirements for a hypothesized nuclear power economy over the next century, as described in the December 1963, Monthly Report. With this hypothetical economy and an assumed nonoptimum reactor, lower grade (more expensive) ore will be needed early in the next century whether or not plutonium is recycled if present thermal burners supply the entire amount of power. If platonium is not recycled, the low grade uranium ores will not last until 2060 and extraction of uranium from such materials as granite would appear necessary. Plutonium recycle obviates this difficulty for the foreseeable future (neyond 2100).

The second step is to improve the accuracy of the above analyses by calculation of optimum conditions for several ore prices with and without plutonium recycle, and with the MELEAGER-QUICK chain for typical thermal reactors (BWR and HWR). The ore use and power production are being integrated over the hypothesized future.

After completing the above steps, the calculation will be repeated using various breeder reactors to determine the over-all resource conservation attainable. A final step is to examine the conservation characteristics of thermal reactors in conjunction with various breeders in combinations that may prevail in the future.

One of the primary considerations in the study of breeder reactors is the total amount of fuel involved in the recycle operation; this amount of fuel is made up of the fuel in the reactor plus the fuel undergoing separation and fabrication outside of the reactor. The longer the out-of-reactor time, the larger the total fuel inventory and conversely, except that "dead" time may be encountered if the out-of-reactor time is reduced so far below the in-reactor time that the reactor is not ready for discharge when recycled fuel is ready. Furthermore, the larger the fuel inventory, the longer the doubling time. The dependence of doubling time on out-of-reactor time is indicated in Figure 8, which has been revised slightly from last month.

Figure 8 was made on the assumption that the breeding complex includes only one reactor. It is worthy of note that by simultaneous use of several reactors, the minimum out-of-reactor time could be reduced, since a batch of reprocessed fuel could be recycled into the first reactor ready to receive it, instead of having to stand idle waiting for a particular reactor.

CODE DEVELOPMENT

Calibration of JASON-MELEAGER Codes

Further study was continued on the calibration of JASON-MELEAGER using some published data from PCTR experiments. From this and other sources it is hoped that "universal" calibration of MELEAGER will be possible that can be used for general surveys. Heretofore, MELEAGER's calibration factors were adjusted for each reactor simulation for even approximate surveys. It was found that JASON predicted values of thermal utilization, f, 1.3% lower than the PCTR value at the smallest lattice spacing and 0.46% higher than the PCTR value at the largest lattice spacing. These values of f with the appropriate ϵ and SDPV were then transferred to MELEAGER where k_{CO} was calculated.





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With a spectral index calibration parameter RAYK l = 1.0, the values obtained for k_{∞} from MELEAGER were higher than the corresponding PCTR values. The greatest deviation occurred at the smallest lattice spacing where the value of k_{∞} was 2.8 percent larger than the experimental value. (See Figure 9.) When RAYK l = 0.5, the greatest deviation from the experimental value occurred at an intermediate lattice spacing, the MELEAGER value for k_{∞} was 2.1 percent below the PCTR value.

Thus, for graphite moderated systems, it appears that some intermediate value for RAYK 1 will yield values of k_{CO} from the MELEAGER code that are in good agreement with experimental results. Determination of this intermediate value of RAYK 1 is currently in progress.

ALTHAEA Code

The timing routine has been incorporated in the program so that the computing time spent in each subroutine, as well as the tape reading and writing time, can be measured. The calculation of control requirements in the diffusion subroutine has been revised to improve conveyance for the cases in which the flux shape is unusually sensitive to control application. A new constant was added to input setting an upper limit on specific power in each region. The flux will now be normalized to provide the desired reactor power or the specific power in the limiting region, whichever is lower.

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J. W. Woodfill Manager-Programming



RADIATION PROTECTION OPERATION REPORT FOR THE MONTH OF JANUARY 1964

A. ORGANIZATION AND PERSONNEL

Effective January 1, F. Swanberg, Jr., transferred to Programming and H. V. Larson assumed the duties of Manager, Internal Dosimetry. Effective the same date, Iral C. Nelson became Manager of External Dosimetry, and R. O. Budd, Senior Engineer, Internal Dosimetry. W. E. Parker returned to Environmental Monitoring effective January 13 after a one-year training period in Radiation Monitoring. Effective the same date, J. R. Berry transferred from Environmental Monitoring to Radiation Monitoring. J. W. Mathis transferred from temporary to permanent status in Composite Dose Studies and Records. Three temporary employees in CDS&R, G. D. Larson, C. R. Bowers, Jr., and E. M. Epperson terminated during the month. T. A. Fleming formerly of HUPO was assigned a temporary position in CDS&R until June 30, 1964. J. N. Morgan, a Tech Grad, accepted a rotational assignment in Radiological Development and Calibrations.

B. <u>ACTIVITIES</u>

Occupational Exposure Experience

There were eight new plutonium deposition cases confirmed by special bioassay analysis during the month. Six resulted from inhalation of contaminated air during the fire in the Redox Final Products Concentration Building (233-S), and one case resulted from a contaminated injury received in the Weapons Manufacturing Building (234-5). All seven were estimated to be depositions of less than 1% of the maximum permissible body burden (MPBB plutonium, with bone as reference, is $0.04 \ \mu c$). The eighth case, estimated to be a deposition of 3% of the MPBB, was detected by routine bioassay sampling. A re-evaluation of a deposition case originally estimated to be less than 1% of the MPBB resulted in its removal from the list of confirmed deposition cases. The total number of individuals who have received internal plutonium deposition at Hanford is 33^4 of which 2^42 are currently employed.

In January there were nine CPD incidents involving nine employees, two HL incidents involving two employees, and one HUPO incident involving five employees which required special bloassay sampling for plutonium analysis. The following is a brief description of the more significant incidents.

A CPD operator received a plutonium nitrate contaminated injury on January 2, 1964, at the Weapons Manufacturing Building (234-5). The employee was cleaning a vacuum filter when his right thumb was

pierced by a wire mesh screen. Examination at the plutonium wound counter indicated 7.3 x 10^{-3} µc plutonium present in the wound. After excision, the wound count was less than the detection limit of 1 x 10^{-4} µc. The employee was not a previous deposition case.

A CPD operator received a plutonium oxide contaminated injury on January 14, 1964, at the Weapons Manufacturing Building (234-5). The employee was cleaning a tantalum plug used in casting when a sliver penetrated the hood and surgeons gloves and entered his left thumb. Visual examination revealed a small black sliver in the thumb reading >40,000 d/m which could not be removed in the field. Examination at the plutonium wound counter, after excision, indicated 2.8 x 10^{-4} µc. The excised tissue, counted by the wound counter showed 0.295 µc. The employee was a previous deposition case estimated to be <10%.

Five HUPO laundry workers were exposed to airborne concentrations estimated to 1.2 x 10^{-11} µc Pu/cc for approximately eight hours while working without proper respiratory protection at the 200-W Laundry (2724-W).

There were two new plutonium contaminated injuries during the month, both requiring excision. In January there were five plutonium contaminated injuries; three requiring excision.

In addition to the incidents involving plutonium, there were two incidents involving seven HL employees, two incidents involving three IPD employees, and one incident involving ten CPD employees that required evaluation for possible intake of radioactive material. The more significant incidents are summarized below:

A strontium and cerium contamination spread resulted from changing an agitator in the 244-CR Tank Farm Vault in 200-E Area. At the end of day shift, nine CPD employees were found with facial contamination up to 1,000 c/m, which was easily removed. Nasal smears, checked in a laboratory counter, indicated contamination from 30 c/m to 600 c/m for five individuals. Examination at the Whole Body Counter indicated that no detectable deposition had occurred in any of the five employees.

Two persons were working in the Shielded Analytical Laboratory (325-B) with irradiated thorium oxide when a spill occurred. An air sample indicated a concentration of $\sim 1 \times 10^{-9} \ \mu c \ FP/cc$. Examination at the Whole Body Counter detected a deposition of only trace amounts of Pa-233.

During inspection of a UO_2 -PuO₂ fuel element, suspected of having a cladding rupture in the storage basin of PRTR (309), a utility light shorted and arced to the fuel element. Fission gas escaping from the

rod gave readings >5 rads/hour. The three operators in the storage basin evacuated immediately. As a result of CP measurements of 300 mrads/hr in the vessel, personnel at that location evacuated also using the emergency air lock to avoid the storage basin area. Both the storage basin and containment vessel were kept on fresh air status for approximately 2-1/2 hours until the ventilation system cleared the gases. Whole Body Counter examinations of the three operators and two radiation monitors detected no significant particulate contamination.

Environmental Experience

Concentrations of fallout materials in the air of the Pacific Northwest continued to decrease during January. The monthly average of 0.6 pc β/m^3 was the lowest recorded since the Fall of 1961. Averages for the months of November and December, 1963, were 1.0 and 0.9 pc β/m^3 , respectively.

Maintenance work associated with the reactor effluent line to the 107-F Retention Basin interrupted the effluent flow for several days. On the afternoon of January 19, high winds from the S.S.W. blew contaminated material out of the nearly dry basin, in spite of efforts with spray equipment to prevent such an occurrence. Particulate contamination greater than 80,000 c/m was found along the west side of the basin, surveys near the basin showed about 300 feet of blacktop road contaminated to about 1 particle every 20 to 30 square feet, and an estimated ten acres of land between the 107-F Basin and the Columbia River was found to be contaminated to a lesser degree. No contamination was found from ground surveys made along Highway 11-A and in the vicinity of Radar Hill (a general northeasterly direction from 100-F Area). Surveys along the roads immediately across the river from 100-F Area detected no contamination. Private cars were temporarily restricted from the Biology farm area as a precautionary measure.

Studies and Improvements

Title II drawings for the Consolidated Service Facility (Project CAF-961) were reviewed for compliance with the established radiological design criteria. Written recommendations resulting from this review were adopted intact and were incorporated into the final project proposal.

Initial calibrations of the simulated G.I. tract dose monitor were continued using distilled water spiked with radionuclides As⁷⁶ and NP²³⁹. The As⁷⁶ response was similar to that previously observed for Zn⁶⁵ and Sc⁴⁶. The Np²³⁹ response was similar to that observed for Cr⁵¹ and was about one hundredth the magnitude of the response for Zn⁶⁵ and Sc⁴⁶. Some difficulty due to poor bonding between the scintillation crystal and the light pipe was discovered near the end of the calibration study. The bond was repaired and calibration samples of Np²³⁹ are scheduled for re-run to confirm the earlier observations.

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New preamplifiers were incorporated in the neutron spectrum measurement electronic system and the system was calibrated. The new preamplifiers reduce the system noise level although the resolution as determined with monoenergetic fast neutrons is approximately the same as that obtained with the old preamplifiers. The neutron spectrum from the PuF_h neutron source was measured with 2 inches of lead between the source and the detector to reduce the gamma radiation interference. Two experiments are still required to properly subtract the gamma radiation interference, though the lead shielding does reduce the interference considerably. When the detector is filled with He³, the resultant spectrum is a combined neutron, gamma, and silicon activation spectrum. The silicon activation is due to (n,p) reactions in the silicon surface barrier detectors. When the detector is filled with air, the spectrum is due to gamma radiation and silicon activation only. The gamma radiation interference and silicon activation spectrum can be subtracted from the combined spectrum. A background spectrum was also measured from the PuBe source with a lead brick as shielding for the gamma radiation.

The spectrometer was taken to the K-West reactor and set up in a relatively low dose rate neutron field. The spectrum was measured at this location with He³ in the detector. The background run with air in the detector is now in progress. The gamma radiation spectrum was also measured at this location with a sodium iodide crystal. A relatively large amount of Co^{60} gamma radiation was present in the spectrum. This Co^{60} gamma radiation caused increased gamma radiation interference in the neutron spectrum measurements.

The addition of a nitrogen purge system recommended by the manufacturer to increase the sensitivity of the thermoluminescent dosimeter reader was completed. Several sets of LiF dosimeters exposed last month were read with nitrogen flowing through the read-out chamber. The dosimeters were read with voltages of 1300 and 1500 volts applied to the photomultiplier. The dosimeters read at 1300 volts did not show much greater sensitivity to neutrons than the dosimeters read before the reader was modified. At 1500 volts, the dosimeters were much more sensitive than before the modification. For one exposure, the difference between the neutron and gamma dosimeters was 56 units per rad of fast neutrons. These dosimeters were exposed to 4.72 MeV neutrons from the positive ion accelerator. If the neutron-gamma difference remains consistently as large as 56 units per rad, the dosimeters should be capable of accurately detecting one rem of fast neutrons. A more complete set of neutron exposures will be obtained as soon as positive ion accelerator time is again available.

A series of autoradiographs were completed which concluded our first 2,000 hour exposure of nuclear track film to air sample material. The alpha tracks showed extensive fading when examined under a microscope. The alpha tracks were examined, counted, and compared to the results from

the 200 hours exposure for identical filter areas. The camera used to make the exposures permitted reproducibility in filming of the same area with excellent precision. This filming precision permitted examination of the alpha tracks from the same particle at both the 200 and 2,000 hour exposure times. It was concluded that while the tracks fading made it more difficult to count the tracks, sufficient evidence of each track remained to make it legible. The smallest particle of plutonium oxide detected with a 2,000 hour exposure was about 0.05 microns in diameter. The particle distribution curves remained similar to those plotted previously with no significant change in mass mean diameter although the count mean diameter became considerably smaller. This is what one would expect since a few large particles easily offset the particles with a diameter between 0.05 and 0.1 microns.

The significance of a single point sample result in relation to the average river characteristics at a sampling location is frequently in question. Study of reactor effluent effects has shown that one or two points in a given cross section will routinely be very close to the mean temperature of the section. In order to test whether the same condition holds for other effluent characteristics, concurrent samples were taken for chemical and radiochemical analysis during a temperature and gamma energy analysis traverse at the 300 Area cross section. Results are not yet available.

The atmospheric monitoring equipment at 100-N Area was activated on December 30, 1963. A radiological design criteria manual chapter, "Design Criteria for Portable Air Samplers", was issued.

The air sampling station at Great Falls, Montana, was closed. This is one in a series of moves to realign the Hanford air sampling network. As another move in the realignment, permission was obtained from the FAA for the installation of an air sampler at their facilities at the Pendleton airport.

Collection of integrated sanitary water samples was begun and the collection of integrated raw water samples was discontinued at Pasco on January 20. The sanitary water sample, like the one at Richland, is being collected by means of a solenoid valve and timer. At the same time, the use of resin columns to sample water for strontium and cobalt analyses was moved from the field to the laboratory. This should improve the measurements since conditions governing the operation of the column can be much more closely controlled in the laboratory.

The 327 Building stack monitoring equipment was evaluated to determine if the stack effluent sample presently take is representative of the stack discharge. The study consisted of comparing the results of samples taken from five probes which traverse the stack. It was found that the result of the sample that we routinely take is generally higher than the average of the stack effluent.

Research Studies

Effect of Reactor Effluent on the Quality of Columbia River Water (02)

Work this month was principally associated with definition of temperature difference on cross sections of the river and traverses were made at several locations to obtain the necessary data. Measurements of this type are necessary at all seasons in order to relate the data recorded by instruments at fixed points to the mean temperature of the river water flowing past the cross section.

Mechanisms of Environmental Exposure (02)

Individuals identified by the creel census as unusually heavy consumers of fish from the Columbia River were matched with records of persons who have received whole body counts. Counts were available for four persons who claimed to have eaten fish once each week and for one person who claims to have eaten fish twice a week. The highest body burden of Zn^{65} detected was 21 nc. (When an RPO staff member ate local whitefish once a week for over a year under controlled conditions, his burden of Zn^{65} reached 130 nc.)

Nuclear Facilities Monitoring Guide

Drafting of the Guide continued to be primarily concerned with describing the principles and methods of evaluating the significance of radiation doses received via environmental pathways. The first draft of the Guide is now approximately 30 percent completed.

C. RELATIONS

Safety meetings were held throughout the Section during the month. There were no security violations, and one minor injury. A security film, entitled "Road to the Wall" was shown to several RPO groups.

Five new suggestions were received this month. Two suggestions are to be reopened, making a total of seven to be evaluated. Six were evaluated during the month. Three of the six were rejected, and three were adopted. Suggestion awards were made to K. B. Elledge - \$10 award; H. N. Larson -\$7.50 award; and J. W. Mathis - \$20 award.

Radiation orientations were presented to Biology, Plutonium Metallurgy, and Electrical Utility personnel.

An orientation on Contamination Control Procedures was presented to twenty employees of Technical Shops assigned to 306 Building.



All four sessions of the fifth round in the Radiation Monitoring Refresher course were held. Total attendance was 56, of whom nine were CPD employees, and one AEC employee. These sessions dealt with basic principles of construction and operation of radiation monitoring instrumentation. Interest in the sessions was generally excellent, but discussion revealed a somewhat lower level of knowledge in this area than was expected. The material presented is believed to have helped the Radiation Monitors appreciably in gaining a better understanding of the instrumentation.

Two three-hour training sessions on Disaster Monitoring were presented for IPD Processing personnel at the Reactor Personnel Certification facility. Total attendance was 16. This completes the series committed for presentation.

A short talk on emergency signals, evacuation, and individual response in such situations was presented to 300 Area Maintenance personnel. Attendance was about 75.

A series of weekly training sessions, conducted by the Specialist, Environmental Monitoring, for the EMO monitors, was begun this month. The sessions examine, in detail, the different tasks of Environmental Monitoring. Four meetings were held in January.

- D. SIGNIFICANT REPORTS
 - HW-76525 12 "Radiological Status of the Hanford Environs for December, 1963", by R. F. Foster.
 - HW-78395 "Evaluation of Radiological Conditions in the Vicinity of Hanford, April-June, 1963", by R. H. Wilson.



External Exposure Above Permissible Limits	January	1964
Whole Body Penetrating	0	0
Whole Body Skin	0	0
Extremity	0	0
Hanford Pocket Dosimeters		
Dosimeters Processed	2,553	2,553
Hanford Beta-Gamma Film Badge Dosimeters		
Film Processed	10,777	10,777
Results - 100-300 mrads	160	160
Results - 300-500 mrads	16	16
Results - Over 500 mrads	3	3
Lost Results	37	37
Average Dose Per Film Packet - mrad (ow) - mr (s)	5.3 19.7	5.3 19.7
Hanford Neutron Film Badge Dosimeters		
Slow Neutron		
Film Processed	1,755	1,755
Results - 50-100 mrem	0	0
Results - 100-300 mrem	1	l
Results - Over 300 mrem	0	0
Lost Results	3	3
Fast Neutron		
Film Processed	530	530
Results $= 50=100 \text{ mrem}$	22	22
Results = 100-300 mrem	76	76
Results - Over 300 mrem	2	2
Hand Checks	3	3
India Checks		
Checks Taken - Alpha	38,228	38,228
- Beta-Gamma	54,160	54,160
Skin Contamination		
Plutonium	1	1
Fission Products	50	50
Uranium	1	1
Tritium	0	0
Thorium	0	0

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HW-80560

HW-80560

Whole Body Counter	Number of Examinations						
Subject	747-A WBC	<u>1964</u>	Mobile WBC	1964	-		
GE Employees Regular Incident Cases Terminations New Hires Special Studies	60 21 3 27 0	60 21 3 27 0	218 1 0 0 10	218 1 0 10			
Non-Employees	<u> </u>	<u> </u>	<u> 0 </u> 229	<u>0</u> 229			

Bioassay

<u>Analysis</u>		Current Reporting Limit	Results <u>Reportin</u> Jan.	s Above ng Limit 1964	Samples Assayed Jan。 1964			
	Plutonium	2.2x10 ⁻⁸ uc/sample	53	53	163	163		
	Fission Prod	. 3.1x10 ⁻⁵ µc/sample	0	0	0	0		
	Strontium	3.1x10 ⁻⁵ µc/sample	0	0	0	0		
	Tritium	5.0 µc/sample	75	75	181	181		
	Uranium	0.14 ugm/1	0	0	0	0		
	Special Stud	ies	0	0	0	0		

Calibrations

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	Number of Un	its Calibrated
	January	1964
Portable Instruments		
CP Meter	1,048	1,048
Juno	275	275
GM	561	561
Other	240	240
Audits	97	97
	2,221	2,221
Personnel Meters		
Badge Film	650	650
Pencils	45	45
Other	294	294
	989	989
Miscellaneous Special Services	65	65
Total Number of Calibrations	3,275	3,275



Environmental Monitoring

Samples	January	1964
Air		
Filters	392	392
Scrubbers	274	274
Water		
Raw	45	45
Sanitary	74	74
Process	25	25
Vegetation	100	100
Test Well	328	328
Fish	199	199
Waterfowl	77	77
Food Products	66	66
Beef Thyroids	31	31
Measurements		
Control Plots	38	38
Aerial Monitoring	0	0
Ionization Chambers	232	232

Carl M. Unruh

for the Manager RADIATION PROTECTION

CM Unruh:ald



FINANCE AND ADMINISTRATION

ACCOUNTING

Cost Accounting

A revised FY 1964 Financial Plan was issued by Washington-AEC during the month. In summary, it includes funds for the nonproduction programs assigned to Hanford Laboratories as follows:

Research and Development	
04 Program	\$13 754 000
05 Program	1 403 000
06 Program	3 380 000
08 Program	240 000
Equipment	
04 Program	587 000
05 Program	143 000
06 Program	301 000
08 Program	45 000

We were informally advised that the above 04 Program Research and Development amount would be adjusted to reflect a net increase of \$150,000 by increasing the Fuels and Materials Program by \$200,000 and decreasing the Plutonium Recycle Program by \$50,000.

The operating cost control budget was adjusted in January as follows:

- 1. Addition of the \$20,000 allocation received for boron technology studies, which is a diversification study authorized by RLOO-AEC.
- 2. Reduction of \$70,000 in the service assessment budget to conform with the latest control allocation from the General Manager HAPO.
- 3. Addition of \$100,000 in the Physics and Instruments Laboratory budget for equipment work in progress to provide a more realistic budget for this function.

Special accounting codes were established during January for the activities described below:

.8T Participation by M. F. Sullivan in a meeting on the "Intestine and Its Response to Radiation," sponsored by Harvard Medical School, Biophysics Laboratory, under a grant from the National Institute of Health. This meeting was held at the Puerto Rico Nuclear Center at San Juan, Puerto Rico, January 15 through 18, 1964. Billing will be for travel and subsistence.

.8V Consultation with APD, Palo Alto, California. J. J. Fuquay consulted on limits and controls for radioactive contamination. Billing will be made for two days at \$175 per day plus travel and subsistence. R. A. Schneider consulted on analytical measurements for radioactive materials. Billing will be for four days at \$100 per day plus travel and subsistence.

The call letter for preparation of the Budget for FY 1966 and Revision of Budget for FY 1965 was issued to Hanford Laboratories' management. The budget is scheduled for completion April 1, 1964, including the research and development proposals.

The following organization and program codes were established during the month:

Organization Codes

737D - Landlord Maintenance 737N - 314 and General Area Maintenance

Program Codes

- .10 Boron Technology (02 Program, Production Miscellaneous)
- .22 Radioactive Shipment Hazards (04 Program)
- .29 Critical Flow at High Temperature and Pressure (04 Program)
- .30 Upstream Boiling Burnout (04 Program)

General Accounting

Approval No. AT-316, AEC Monograph on Iodine 131 - L. K. Bustad, C. C. Gamertsfelder, and J. K. Soldat, was sent to the AEC, January 23, 1964. An addendum to Approval No. AT-198 is being prepared covering the shipment of additional miniature swine, which are excess to our needs, to Hammersmith Hospital, London, England.

The following revised OPGs were issued during January 1964:

OPG No.

Ti	t.1	e
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22.1.5	Biology Laboratory Organization
22.1.12	Test Reactor and Auxiliaries Organization
2.2	HAPO Organization
3.9.1	Safety Shoe Purchase
3.4.19	Weekly Salaried Employee Appraisal Plan

Classification activities for the month included the review of 886 Purchase Requisitions, 715 Work Orders, 16 Appropriation Requests, and 11 Stock Adjustment Requests for capital expense determination, compounding, work review, and for reimbursability.

Hanford Laboratories' net material investment at January 1, 1964, totaled \$24.9 million as detailed below:

SS Materi Reactor a Spare Par Yttrium	al and Other Specia ts	al Material	(In thousands) \$ 23 668 982 349 26
Subto	tal.		25 025
Reserve:	Spare Parts Yttrium	\$79 26	(105)
Net In	nventory Investr	nent	\$ 24 920

The cumulative value of nuclear material consumed in research by Hanford Laboratories during FY 1964 (at January 1, 1964) is \$560,678 comprised as follows:

02	Program	\$ 17	099
03	Program	156	294
04	Program	<u>387</u>	285
	Total	\$560	678

The following is a summary of Hanford Laboratories' inventory activity excluding nuclear material for the first six months of FY 1964 at December 31, 1963:

	Balano 7-1-63	ce <u>}</u>	Recei	.pts	Consu tic	mp- on	Oth	<u>ier</u>		Bala <u>12-3</u>	unce 31-63	Fo Be 6-	oreca llanc -30-6	st e 4
Heavy Water Zirconium Platinum All Other Reactor &	\$ 400 247 191	599 466 419	\$375 56 7	927 921 102	\$ (89 (43 (531) 600) (908)	\$(240 (6	940)-1) 719)-3)	\$	44 6 260 190	055 787 - 2 894	\$ 2)	381 236 204	948 506 118
Materials	89	<u>573</u>	1	384	(6	<u>664</u>)	<u></u>	82		84	<u>375</u>		99	143
Subtotal	929	057	441	<u>334</u>	(140	<u>703</u>)	(247	<u>577</u>)		982	111	····	921	71.5
Spare Parts Less Reserve	327 (81	859 267	89	454	(66	541)	(1	985)- 4)		348 (79	787 208).	-5)	328 (82	000 000)
Subtotal	246	592	89	454	(66	<u>541</u>)	(1	<u>985</u>)-4)		269	579		246	000
Total Inven- tories	<u>\$1 175</u>	649	<u>\$530</u>	788	<u>\$(207</u>	<u>244</u>)	<u>\$(249</u>	<u>562</u>)	<u>\$1</u>	251	690	<u>\$1</u>	167	<u>715</u>

- (1- Scrap returned to SROO.
- (2- Does not include 3,840.4 lbs. scrap material and 10,388.3 lbs. of R&D material (charged to Cost) controlled by quantity only at the Laboratory Storage Pool.
- (3- Scrap shipped to the NYOO for reclaiming.
- (4- Spare parts excessed.
- (5- The spare parts reserve at 12-31-63 is short \$7,989 of the required 25%. This reserve will be adjusted during June 1964 business; currently the deficit is applicable to PRTR Cost.

A certification type inventory of Other Special Materials in the custody of Hanford Laboratories' material holders was conducted as of December 31, 1963. The inventory revealed a net deficit of \$387, primarily due to material lost to environment during flame spraying operations, through loss as Pu contamination and from corrosion in hot cell processes. The necessary approved documents were obtained from material holders to remove dollars from the inventory account.

Two SS Material forecasts were prepared during the month:

- SS Material Projects R&D Other Than in Production (for all programs except 02 Program). The period covered by this forecast was FY 1965.
- 2. A ten-year SS Material Planning Estimate for all material within the O4 Program. The period covered by this report was FY 1965 through FY 1974.

Total Hanford Laboratories completed Plant and Equipment Investment at December 31, 1963 is shown below:

Section	Amount	Area	An	Amount		
Hanford Laboratories Management	\$ 47 987-1)	100-B	\$	103	550	
Finance and Administration	2 747 308	100-D	2	132	571	
Programming	181 120-1)	100-F	3	891	638	
Test Reactor & Auxiliaries	20 194 344	100-H		18	268	
Physics & Instruments Lab.	5 902 490	100-K	1	058	246	
Reactor & Fuels Laboratory	27 353 777	100-N		l	385	
Chemical Laboratory	12 421 778	200 -E	1	471	235	
Biology Laboratory	3 952 085	200 -W	5	263	692	
Radiation Protection	2 204 985	300	58	711	366	
Applied Mathematics	168 073-1)	1100		36	424	
Other HAPO Departments	951 270 -1)	700		500	534	
		Off-Site	l	265	007	
		White Bluffs		659	387	
		Riverland		2	443	
		General				
	and the state of the	Reservation-	2 <u>) 1</u>	009	471	
	<u>\$76 125 217</u>		<u>\$76</u>	125	217	
		U	NCLAS	SSIF	[ED	

- (1- Represents value of occupied building space.
- (2- Includes plant and equipment inside the perimeter barricade, excluding the limited areas.

In addition to the above, Hanford Laboratories has RDX equipment valued at \$1,137,674; POO equipment (property belonging to the Air Force and Canadian Atomic Energy) valued at \$146,900; office equipment, photographic equipment, ADP (IBM rental equipment), firearms and binoculars, and automotive and construction type equipment utilized by Hanford Laboratories personnel but the accountability responsibility of other HAPO departments.

The value of Hanford Laboratories equipment located in buildings assigned to other HAPO Departments is:

IPD	\$4 064 834
CPD	426 447
HU&PO	6 338
NRD	1 385
	\$4 499 004

Laboratory Storage Pool activity is summarized as follows:

	Current Month		FY t	o Date
	Quantity	Value	Quantity	Value
Beginning Balance Items Received Items Reclaimed by Custodians Equipment Transfers Items Disposed by PDR Items Disposed by Excess	1 762 243 (14) (53) (6) <u>(108</u>)	\$1 443 305 32 050 (27 688) (13 810) (99) (68 061)	1 480 1 238 (151) (238) (146) (359)	<pre>\$ 811 520 946 405 (76 004) (97 390) (11 389) (207 445)</pre>
Ending Balance	1. 824	<u>\$1 365 697</u>	1 824	<u>\$1_365_697</u> _1)

(1- Includes 172 items valued at \$120,485 on loan at January 31, 1964.

During the month, 161 items valued at \$54,960 were loaned and/or transferred in lieu of purchases. A total of 666 items valued at \$261,752 has been redirected to useful purposes this fiscal year. Operating costs for FY 1964 (ϵ t December 31, 1963) were \$9,033.

Total value of equipment and material in custody of the Laboratory Storage Pool at January 31, 1964 was \$2.3 million including Reactor and Other Special Materials of \$302,943; SS Material, \$154,800; and Other Materials, valued at \$483,358.

During the month, the total platinum, gold, and silver (valued at \$14,205) holdings of one Chemical Laboratory custodian were transferred to the Laboratory Storage Pool.

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The heavy water inventory at the end of January 1964 showed a loss of 701 pounds valued at \$9,623 for the PRTR. Heavy water scrap generated during the month amounted to 2,127 pounds, resulting in a \$2,574 charge to operating cost. Heavy water accumulated at January 31 for return to SROO amounted to 11,006 pounds valued at \$137,818. Fifty-five drums (27,445 pounds) of heavy water were received during the month, having a value of \$375,448 Fund and \$172,629 Nonfund.

Action on projects during the month follows:

New Money to HL

21

CAH-922 Burst Test Facility for Irradiated Zirconium Tubes (\$15 000) (Contingency funds not required by GE for work assigned to GE.)

Construction Completion and Cost Closing Statements Issued

*CAH-958 Plutonium Fuels Testing and Evaluation Laboratories, 308 Bldg. CGH-992 Additional Fuel Loading Equipment, 308 Bldg. *CAH-995 Air Conditioning Modifications, 309 Bldg.

*AEM Services only

The following contracts were processed during the month:

DDR-184	Van's Metal Spinning Company
SA- 321	The Swedish Hospital
SA-322	Schwarzkopf Microanalytical Laboratory
SA-327	William E. Beckmeyer

Personnel Accounting

Effie Seaman took optional retirement effective February 1, 1964.

Personnel statistics follow:

Employee Changes	Total	Exempt	Nonexempt
Employees at beginning of month Additions and transfers in Removals and transfers out	1 811 18 	792 13 <u>7</u>	1 019 5 14
Employees on payroll at end of month	1 808	<u>798</u>	1 010

Overtime Payments During Month	January	December
Exempt Nonexempt Total	\$ 9 828 18 498 \$ 28 326	\$ 10 736 35 206 <u>\$ 45 942</u>
Gross Payroll Paid During Month		

Exempt	\$ 780 954	\$ 779 533
Nonexempt	557 595	706 727
Total	\$1.338 549	\$1 486 260

Participation in Employee Benefit	Janua	ry	Dece	mber
Plans at Month End	Number	Percent	Number	Percent
Pension Insurance Plan - Personal	1 599 411	99.3	1 585 415	99.4
- Dependent	1 392	99.9	1 377	99.9
U.S. Savings Bonds Stock Bonus Plan Savings Plan Savings and Security Plan Good Neighbor Fund	158 66 1 257 1 299	39.0 3.6 89.4 71.7	158 68 1 241 1 294	39.5 3.8 88.3 71.7
Insurance Claims Employee Benefits	Number	Amount	Number	Amount
Life Insurance Weekly Sickness and Accident Comprehensive Medical	-0- 15 76	\$ -0- 1 237 4 762	-0- 9 63	\$ -0- 987 4 522
Dependent Benefits Comprehensive Medical Total	135 226	12 876 \$18 875	<u>121</u> 193	<u>11 877</u> \$ 17 386

TECHNICAL ADMINISTRATION

Employee Relations

Seven nonexempt employees were hired; no open requisitions remain.

Suggestion plan activity included 55 submissions, 25 adoptions, and 26 rejections.

Information and Presentations

Visitors Center activity:

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January attendance	1. 034
Average attendance per day open	4 <u>1</u>
Cumulative attendance since 6-13-62	61 319
Conducted groups	8 (totaling 63 people



Plant tour activity:

	Number	Total People
General Public Relations Tours	3	23
Special tours	2	15
Cumulative visitors since		
1-1-64 (all tours)	-	38

Documented information flow during the month was comprised of 1,310 titles (13,164 copies) received at Hanford and 104 titles (7,739) copies sent offsite.

The major effort (copy layout work, pictures, etc.) have been completed on the color brochure being prepared to assist Tri-City diversification efforts. Remaining to be done is a host of final checks, minor revisions, and securing approvals.

News releases were prepared and distributed on PRCF, the use of sugar in chemical separations, the use of a digital computer to control solvent extraction, and the recovery and use of technetium-99.

Professional Placement

Overall recruiting results for January are as follows:

Offers extended	10
Offers accopted	12
Offers rejected	18
Added to roll	10

Advanced Degree - Eight Ph.D. applicants visited HAPO for employment interviews. Five offers were extended; two acceptances and one rejection were received. Four offers are currently open.

<u>BS/MS (Direct Placement)</u> - Four offers were extended. Four acceptances and no rejections were received. There is one offer currently open.

<u>BS/MS (Program)</u> - One offer was extended. Six acceptances and 17 rejections were received. Twenty-seven offers are currently open.

<u>Technical Graduate Program</u> - Seven Technical Graduates were placed on permanent assignment. Four new members were added to the roll and none terminated. Current Program members total 67.

FACILITIES ENGINEERING

At month end, Facilities Engineering Operation was responsible for nine active projects having total authorized funds in the amount of \$7,592,500. The total estimated cost of these projects is \$10,839,000. Expenditures on them through December 31, 1963 were \$2,038,000.

The following summarizes project activity in January:

Authorized projects at month end ------ 9 New projects authorized ----- 0 Projects completed ----- 0 New projects submitted to the AEC ----- 2 CAH-126, Waste Transport System CAH-128, Heat Transfer Apparatus for Model Studies New projects awaiting AEC approval ----- 3 CAH-114, Critical Mass Laboratory Ad Lition CAH-126, Waste Transport System CAH-128, Heat Transfer Apparatus for Model Studies Project proposals being prepared ----- 5 CAH-123, Laboratory Fire Protection System Modifications to 5201 Building Geological and Hydrological Wells - FY 1964 Atmospheric Physics Building 327 Building Services Addition

The status of active projects follows:

<u>CAH-916 - Fuels Recycle Pilot Plant</u> - Construction is 28 percent complete compared to a scheduled 23 percent. Concrete work is continuing at and below grade. Piping and electrical work are proceeding on schedule. Some structural steel has been erected above grade. The contractor is encountering difficulty with the subcontractors' providing the shielding windows, sleeves, and plugs. These difficulties could delay construction progress.

<u>CAH-922</u> - Burst Test Facility for Irradiated Zirconium Tubes - Construction is 99 percent complete compared to a scheduled 100 percent. The directive completion date of January 31, 1964 was not met. It will be necessary to extend it about two weeks. The project is complete except for a minor amount of electrical wiring, painting, equipment testing and decontamination, and rehabilitation and installation of the prototype containment vessel.

<u>CAH-962 - Low Level Radiochemistry Builing</u> - Detailed design is completed. A construction bid package is being prepared. A directive dated January 13, 1964, authorized total project funds in the amount of \$1,200,000. The invitation to bid on construction of the building should be issued early in February.

<u>CAH-977</u> - Facilities for Radioactive Inhalation Studies - Design is complete. The Commission has not issued a project proposal revision requesting authorization of construction funds. No progress is being made.

<u>CAH-982 - Addition to Radionuclide Facilities - 141-C Building</u> - The architectengineer completed the design and estimated the construction will cost \$136,000. This is \$39,000 more than his estimate upon completion of Title I design. This will result in an estimated total project cost of \$170,000.

<u>CGH-999</u> - Plutonium Recycle Critical Facility Conversion to Light Water -Beneficial use of the facility was attained on December 30, 1963. Since that time it was necessary to fabricate new control rod snubbers and rod followers as a result of early operating difficulties. All work is now completed. It is anticipated that a project proposal revision will be submitted requesting authorization to increase the scope to provide capability within the PRCF for handling and testing irradiated fuel pieces. Therefore, it is not planned to issue a Physical Completion Notice at this time.

<u>CAH-100 - High Temperature Lattice Testing Reactor</u> - The Company completed its review of Vitro's Title I design package on December 13, 1963. Comments were relayed to Vitro by the Commission on December 27, 1963. On January 10, the Commission informed the Company the current estimate for design and construction of this facility is \$2,700,000, which exceeds the available funds by \$200,000. A meeting was held on January 15 among Commission, Vitro, and Company representatives to review the estimate and consider possible scope changes that will reduce the total project cost. Vitro is evaluating the information discussed, and the Commission will then decide upon the next step to be taken.

<u>CAH-114 - Critical Mass Laboratory Addition</u> - The project proposal requesting design funds was forwarded to Washington-AEC accompanied by a letter recommending its approval.

<u>CAH-116 - PRTR Decontamination and D₂O Cleanup</u> - Vitro Engineering Corporation is performing Title I design. Several meetings have been held with Vitro and Commission representatives to discuss general requirements and technical aspects of the work.

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CAH-119 - PRTR Storage Basin and Experimental Facilities Modifications -Vitro Engineering Corporation began design on January 27, 1964. The directive was issued November 15, 1963.

<u>CAH-123 - Laboratory Fire Protection System - The project proposal has been</u> rewritten and will be rerouted for approval.

CAH-126 - Waste Transport System - The project proposal was transmitted to the Commission for authorization on January 29, 1964

CAH-128 - Heat Transfer Apparatus for Model Studies, 185-D - The project proposal was transmitted to the Commission on January 29, 1964.

Engineering Services

Engineering work was performed in support of design and construction on active projects, project proposals, preliminary planning, and design criteria for new projects. Principal work items included: (1) field liaison, review of shop drawings and approval of submitted materials on CAH-916, FRPP; (2) review of A-E design on CAH-962, Low Level Radiochemistry Building; (3) review of A-E design on CAH-982, Radionuclide Facilities; (4) review of A-E preliminary design on CAH-100, HTLTR; (5) study of 327 building addition and scope and design criteria for modifications to 5201 building and facilities for the split table assembly being transferred from KAPL; (6) preliminary scope for operation of Critical Facility with irradiated fuel, CGH-999, and (7) scope and design criteria for Atmospheric Physics Building.

Scope design and design criteria are being prepared for the Containment System Experiment. Discussions have been held with the J. A. Jones Company and with potential suppliers of the major equipment pieces. Design of the containment barrier was completed by Vitro Engineering.

Engineering and consulting work provided to research and development personnel included: (1) engineering assistance on experimental neutron spectrometer, 105-KE Building; (2) engineering assistance on high temperature gas loop, 314 building; (3) engineering assistance on HTLTR mock-up; (4) engineering and test work for installation of 20,000 psi pressure bonding autoclave; (5) engineering assistance on fast reactor concept study; (6) engineering assistance on critical mass experiments; (7) engineering of Biology transfer cart and hood; (8) engineering assistance was provided to CPD for an audit of the 233-S ventilation system and consulting assistance on Project CAC-880, CAC-121 instrument design and dry-air system, 234-5; (9) engineering work for installation of 329 motor generator; (10) development of engineering information for purchase of computer trailer; (11) engineering for equipment packaging for arctic Biology program; (12) engineering for power vibrating feeder index; (13) engineering for installation of waste flow meters; (14) engineering on liquid sodium loop instrumentation; (15) engineering for installation of induction heater, 327 and 292-T; and (16) engineering for repair of rolling mill.

Plant Engineering

Engineering service was provided on numerous maintenance and laboratory modification and improvement jobs. Major items were: (1) engineering inspection of Atmospheric Physics towers; (2) engineering for installation of 325 building emergency electrical generator; (3) engineering of modifications to 325 building counting room air conditioning system; (4) engineering to adapt double filtration filter boxes for use in 325-B with activated charcoal media; (5) ventilation study of 144-F building; (6) work on engineering standards; (7) engineering for chemical sewer to 306 building; (8) analysis of ventilation, room 138, 308 building; (9) safety modification of 314 crane; and (10) load analysis of 321 building crane.

Pressure Systems

Specifications for the reactor simulator vessel (2500 psig) for the containment systems experiment were developed.

Designs were reviewed for the 1200° F liquid metals loop.

The C-1 loop was charged with fuel elements and is in complete operation. No difficulties have been experienced.

Facilities Operation

Costs for the month of December were \$174,513, which is 105.4 percent of the forecast for the month. Fiscal costs for the first six months were 107 percent of the predicted. Maintenance costs continue to be high. Curtailment of unusual maintenance has been initiated as a control measure.

The following table summarizes waste disposal operations:

Item	November	December
Concrete waste barrels disposed to 300-Wye burial ground	4	2
Concrete waste barrels disposed to 200-W plutonium burial ground	0	5
Loadluggers of dry waste disposed to 300-Wye burial ground from the 325 building	3	3
Loadluggers of dry waste disposed to 300-Wye burial ground from 300 area sites other than the 325 building	19	16

Item	November	December
Loadluggers of dry waste disposed to 200-W plutonium burial ground from 300 area sites	.5	?
Crib waste volume (gallons)	400 000	355 000

A work order has been issued to CPD to install six vertical burial tubes in 200-W for disposing milk pails containing Pa waste.

None of the retention basins exceeded the activity levels of 1.0×10^{-5} $\mu c-\beta/ml$ or 5.0 x $10^{-6} \mu c-\alpha/ml$.

The Class A inspection of the wheel assemblies for the three tankers is complete. Except for the broken cross-axle on tanker $\#5\frac{1}{2}$, nothing unusual was found. All the brakes were relined, brake drums were turned down, and air hoses and valves were repaired or replaced.

Twenty hours of decontamination work was done for 308 building. About 75 casks were decontaminated this month. Fifty-six of these were cleaned in the decontamination tank.

High pressure air revisions were completed at 308 building. All compressed air for the hood process work is presently being supplied by the 384 system. The 308 Joy compressors are on standby.

The 326 building air flow system was calibrated and updated. Survey of all HL buildings is scheduled with resurvey scheduled each year to maintain complete air flow data.

Drafting

The equivalent of 138 drawings was produced during the month for an average of 30 man-hours per drawing.

Major jobs in progress are: (1) high temperature gas loop; (2) HTLTR mockup core; (3) plutonium powder processing line; (4) fast super pressure power reactor concept; (5) thoria processing line; (6) inhalation studies hood asbuilts; (7) rotating crystal spectrometer; (8) capacitor discharge test apparatus; (9) 209 building piping service drawings; (10) modifications to 309 chilled water system; (11) refraction compounds glove box; and (12) dynapak furnace hood. Work was also produced in support of engineering reported under previous sections of this report.

Activity during the month on construction work (J. A. Jones Company) being performed for Hanford Laboratories components is given below:

		Unexpended Balance
Orders	outstanding beginning of month during the month (including	\$2 57 209
stpp J.A.	l. and adj.) Jones extenditures during month	356 735
(inc	ludes C.O. ccst)	242 201
Balanc	e at month end	371. 743
Orders	closed during month	145 103

In addition, work on four maintenance work orders issued to plant forces and having a face value of \$14,032 was supervised.

Major nonproject jobs in progress during the month were: (1) building modifications and laboratory furniture, 141-H; (2) install platform, stairway and monorail, 308; (3) relocate small dynapak and install new dynapak, and renovate, room 125, 308; (4) construct block addition for gas loop, construct office addition and install HTLTR mock-up, 314; (5) construct thoria laboratory, room 417, install emergency generator, modify room 520, install 440 V distribution panel and filters and exhaust ducts, construct high temperature labs and mezzanine offices, install floor tile in ceramics laboratory, enclose north entry to basement, enlarge men's restroom, and construct retaining wall, 325; (6) install lighting, 3717-B; (7) enclose mock-up area and construction manhole and retention waste bypass line, 309; and (8) construct roof, 3718.

Containment systems experiment facilities have been started in 222-T. The crane maintenance platform is being constructed and materials are being purchased for the ventilation barrier wall.

Fabrication work continues on the waste solidification program equipment for 324 building. Racks 2A and 1B are scheduled for test installation in 321 building in February.

Plans have been completed for initiation of the FY 1964 well drilling program. Major work involves improving existing wells and trial work will be accomplished by J. A. Jones contract to determine method and procedures.

Manager Finance and Administration

W Sale: JVM: whm



Daganom

REACTOR DEVELOPMENT - 04 PROGRAM

PLUTONIUM RECYCLE PROGRAM

Plutonium Recycle Test Reactor

Operation

Reactor output for January was 1482 MWD, for an experimental time efficiency of 78% and a plant efficiency of 68%. There were five operating periods during the month, two of which were terminated manually and three were terminated by scrams. A summary of the fuel irradiation program as of January 31, 1964, follows:

	Al-Pu		U02		Pu02-U02		Other		Totals	
	No.	MWD	No.	MWD	No.	MWD	No.	MWD	No.	MWD
In-Core Maximum Average	0		6	982.2 251.7 163.7	79	9349.8 221.3 118.4			85	10332.0
In-Basin Buried	43	3726.4	26	2882.1	24	1150.2	ı	7.3	93 1	7758.7
Chem. Process.	<u>32</u>	<u>2309.3</u>	<u>35</u>	1965.8					67	4275.1
Program Totals	75	6035.7	67	<u>5830.1</u>	103	10500.0	1	<u>7.3</u>	246	22373.1

Note: (MWD/Element) X 20 = \sim MWD/TU for UO₂ and PuO₂-UO₂.

 $\mathrm{D}_2\mathrm{O}$ and indicated helium losses for January were 700 pounds and 133,907 scf., respectively.

Equipment Experience

A total of 59 reactor outage hours were charged to repair work. Main items were:

Instrume	32 hours				
Valves					ll hours
Process	Tube	to	Nozzle	Gaskets	6 hours

Preventive maintenance required 324 hours or about 7% of the total maintenance effort.

A recorder has been installed in the control room to record the four communications channels. This will provide the ability to review reactor communications following an unusual occurrence.

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Improvement Work Status (significant items)

Work Completed

Modification for Reuse of Damaged Nozzle Caps 125-Volt Battery Disconnect Contactor Pressurizer to Stack Valve Relocation Boiler Feedwater Pressure Transmitter Snubber

Work Partially Completed

In-Line Gas Sampling Process Tubes Level Indicator Inlet Gas Seal Replacement Shim Rod Shroud to Top Cap Modification Improved RTD Connector Sealant and Bracing Instrument Power Transfer System Installation of New Alarm Annunciator Thermistor Probe Installation in FEEF Flow Monitor Tubing Snubber Installation Indication of DC Solenoid Failure Holdup Tank - High Level Alarm

Design Work Completed

DT-1 Storage Tank Sight Gauge Emergency Personnel Air Lock Door Operators Containment Valve Bypass for Sump Pump-out Lines

Design Work Partially Completed

Additional Fuel Storage and Examination Facility Vibration Snubbers for Earthquake Protection Decontamination Building and D₂O Cleanup Facility Flux Wire Scanning System Supplemental Emergency Water Addition Permanent Installation of Closed Circuit TV Rupture Monitoring System Modifications

Process Engineering and Reactor Physics

A study is being conducted to determine the feasibility of using soluble boron in the moderator of PRTR to supplement or replace the existing shim system. Results to-date indicate that an initial concentration of about 10 ppm boron in D₂O will be required to compensate for temperature effects and for equilibrium xenon poisoning. Corrosion studies have been started to determine the effect of concentration of boric acid on calandria shroud tubes just above the operating moderator level.

Allowable tube powers were reviewed for the various types of PRTR fuel elements based on light water injection capacity and amount of plutonium segregation. New derating factors were assigned to the different types of mixed oxide fuel elements and the administrative control limit of 1300 KW relaxed. Allowable tube powers now range from 1300 KW to 1800 KW.

A review of the capacity of the light water injection system for higher specific power fuel elements was completed. The present LWI system is inadequate for elements having specific powers of 54.8 MW/ton and 73.2 MW/ton for certain types of primary ccolant leaks.

Preparation of a preliminary safeguards analysis for reactor power levels up to 120 megawatts was started. This review will be concerned with the hazards analysis of operation at higher reactor power levels. Analog studies were initiated.

Procedures

Operating Procedures issued Revised Operating Procedures issued Revised Operating Standards issued Temporary Deviations to Operating Standards Process Specifications accepted for use Equipment Standards issued	issued	1 5 11 13 6 2
Drawing As-Built Status:	January	Total
Approved for As-Built In Drafting In Approval Deleted or Voided Scheduled for review Total	81 7 1	1 262 29 1 <u>82</u> 1 374 <u>213</u> <u>1 587</u>
Personnel Training:		Manhours
Qualification subjects Specifications, Standards, Procedures Emergency Procedures Maintenance Procedures		138 74 18 <u>43</u> 273

Status of Qualified Personnel at Month-End:

Qualified Reactor Engineers	8
Provisionally Qualified Reactor Engineers	1
Qualified Lead Technicians	6
Qualified Technicians	18
Provisionally Qualified Technicians	4

Experimental Reactor Services

The status of the various test elements at the end of January 1964, is shown below. Those elements which had reached their assigned goal exposure or had been permanently discharged for other reasons prior to January 1, 1964, have been deleted from the table.

Test No.	Channel Location	Number	Description	Date Initial <u>Charge</u>	Dis- charged	Approximate Accumulated <u>MWD</u>
14	1956	5097	Moxtyl-Swaged	4/2/62		138.1 repad
14	1352	5098	Moxtyl-Vipac	5/8/62		219.0 repad
14	1758	5099	Moxtyl-Vipac	5/8/62		154.3 repad
48	1156	5150	Moxtyl (1/2" x 1/2" pads)	8/1/62		150.8
47	Basin	5121	Unautoclaved LX AlPu	6/13/63	1/27/64	101.9
47	Basin	5194	Unautoclaved LX AlPu	7/6/63	1/27/64	97.7
47	Basin	5193	Unautoclaved LX AlPu	7/6/63	1/27/64	97.6
54	1542	5116	Moxtyl (clip-on pads)	5/8/62		154.9
54	1554	5118	Moxtyl (clip-on pads)	5/8/62		221.3
61	1249	5185	Moxtyl-Physics	5/28/63		127.2
61	1354	5186	Moxtyl-Physics	5/28/63		116.4
61	1556	5192	Moxtyl-Physics	6/13/63		128.1
67	1152	5119	Moxtyl (Repaired wire)	10/20/63		59.0
67	1457	5117	Moxtyl (Repaired wire)	10/20/63	~-	101.5
80	Basin	5210	Moxtyl(1% PuO2, Swaged)	11/8/63	1/11/64	42.6
80	Basin	5213	Moxtyl(1% Pu02, Swaged)	11/11/63	1/27/64	53.5
80	1544	5214	Moxtyl(1% PuO2, Swaged)	11/18/63		47.6
85	1847	5230	Moxtyl(1% PuO2, Vipac)	1/30/64	~~	0.0

Three fuel elements failed during the month and were removed from the reactor.

A total of 23 fuel elements were examined in the basin during the month. One rod was cut from each of six elements and shipped to Radiometallurgy for testing.

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Flutonium Recycle Critical Facility

Startup work for the H₂O moderated PRCF continued during the month, which included pre-operational testing, completion of project exceptions, administration, preparations and some repair work. At month-end the fuel follower sections for the rods are the last major item still needed.

Modifications were made to the PRCF-EBWR core consisting primarily of enlarging 60 holes in the lattice plates to accommodate thimble tubes. The thimbles will be used to accomplish void formation tests. Provision was also made for a flux foil penetration near the center of the core.

One Nuclear Safety Specification was issued and three revised Process Specifications were accepted for use.

Fuel Element Rupture Testing Facility

FERTF Test 4-0, using a defective Al-Pu element, was completed. Considerable difficulty with airborne radioactive contamination was encountered in the loop annex cell, equipment room and tunnel. Air activity problems were evaluated while FERTF Test 4-1, utilizing another defective Al-Pu element, was being performed. As a result, vent lines to the pump hoods were installed upon completion of FERTF Test 4-1, thereby eliminating the air activity problems.

Processing of Spent PRTR Fuels

A preliminary schedule was prepared for the chemical processing of 35 Al-Pu foel elements in March.

TECHNICAL SHOPS OFERATION

Total productive time for the period was 20,130 hours. This includes 16,059 hours performed in Technical Shops, 2,899 hours assigned to J. A. Jones Company, 1,045 hours assigned to offsite vendors, and 127 hours to other project shops. Total shop backlog is 14,991 hours, of which 90% is required in the current month with the remainder distributed over a three-month period. Overtime worked during the month totaled 231 hours or 1.1% of the total available hours.

Distribution of time was as follows:

	Manhours	% of Total
N Reactor Department	2 681	13.3
Irradiation Processing Department	3 602	17.9
Chemical Processing Department	482	2.4
Hanford Laboratories	13 365	66.4
Hanford Utilities and Purchasing Operation	-	- '

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LABORATORY MAINTENANCE OPERATION

Total productive time was 23,300 hours of 24,600 hours potentially available. Of the total productive time, 91% was expended in support of Hanford Laboratories components, with the remaining 9% directed toward providing service for other HAPO organizations. Craft overtime worked during the month was 1.5% of total available hours. Manpower utilization (in hours) for January was as follows:

Α.	Shop Wor	k .			2	400
в.	Maintena	nce			8	400
	1. Pre-/	entive Maintenance	2	500		
	2. Emer	gency or Unscheduled Maintenance	1	100		
	3. Norm	al Scheduled Maintenance	4	200		
c.	R&D Assi	stance			12	500

mon Rice

Manager Test Reactor and Auxiliaries

WD Richmond:bk



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.

INVENTOR

E. R. Astley L. M. Finch E. R. Astley L. M. Finch R. J. Hennig W. W. Schulz

TITLE OF INVENTION OR DISCOVERY

A New Method of Controlling Reactors

Nuclear Reactor Skewed Process Tube Design and Arrangement

Tertiary Amine Solvent Extraction Recovery of Plutonium and Neptunium from Purex Process 1WW Solution

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Manager, Hanford Laboratories






DATE FILMED 4/13/93

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