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BATTELLE-NORTHWEST MONTHLY ACTIVITIES REPORT

MARCH 1965

APRIL 15, 1965

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PACIFIC NORTHWEST LABORATORY

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For The

U. S. ATOMIC ENERGY COMMISSION

Under Contract No. AT(45-1)-1830

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of 165 pages.

PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
MARCH 1965

Compiled by
Department Managers

April 15, 1965

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By Authority of W.H. Sullivan

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By J. C. Sullivan

Verified By J. C. Sullivan

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RESEARCH AND DEVELOPMENT

1. Reactor and Materials Technology

Production Reactor Support - The effect of aluminum, iron, and silicon additives on the hot hardness and elevated temperature stress cycling behavior of uranium are being studied experimentally. Aluminum additions produce the greatest resistance to plastic deformation at 400 and 500 C.

Two Al-Li target elements with an inner aluminum can and an outer Zircaloy-2 can that were irradiated to an exposure of 6 gas volume ratios showed no changes in dimensions or density.

Corrosion of Zircaloy by molten aluminum is nearly uniform with a penetration rate of 0.4 mil/min at 700 C and 3.5 mils/min at 900 C. The penetration rate slows with time. Addition of nickel or silicon to aluminum effects a distinct reduction in corrosion rate. Flame sprayed alumina coatings on Zircaloy are notably protective at 700 C.

Copper-40 at. % Cd alloy rapidly disperses in molten Al-Li alloy at 700 C. Cadmium oxide or gadolinium poison showed no significant reaction under the same conditions.

The rate of tritium evolution through 30-mil aluminum cladding increased from 1×10^{-3} cc/day to 3.4×10^{-3} cc/day in a 14-hr run at 350 C. At 375 C, a rupture occurred in the cladding, releasing gas contained in the target center hole. Visual examination of the cladding failed to indicate the area of rupture.

Corrosion failures of the copper wire connections in the resistance temperature detectors (RTD) monitoring the N-Reactor outlet water temperature were investigated. Cause of failure was identified as corrosion of the copper wires by residual soldering flux. Proper removal of the flux eliminates the problem.

Test results confirm the hypothesis that hydriding of Zircaloy-2 can occur in the presence of water if conditions for protective oxide formation

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are unfavorable due to the low temperature or asperities on the metal surface.

The N-Reactor heat exchanger decontamination using the APSULF procedure resulted in low DF's (~ 1.6) on stainless steel components due to excessive AP solution dilution.

Half-plant reactor tests were started to evaluate (1) corrosion and film formation in raw river water, and (2) effectiveness of a silicate inhibitor.

Graphite burnout monitors reveal a maximum burnout of 3%/1000 operating days 60 in. into the stack. Lesser values were noted at 95 and 155 in. and at the centerline.

Effective-gamma-dose-rate studies have been made, using the decomposition of methane as a criterion.

Examination of two Zircaloy-2 tubes discharged from KW-Reactor disclosed massive hydriding on the inside surfaces in an 8 ft section at the downstream end of both tubes. Programs have been started to determine the extent of hydriding in other tubes in the reactor, the mechanism by which the hydride is formed, effects on the mechanical properties of the tubes, non-destructive methods of detection and a way to recover tubes partially affected by the problem.

Twelve boiling burnout points were obtained with an electrically heated model of the outer flow annulus in an N-Reactor fuel column with the fuel eccentric toward the bottom of the process tube. A preliminary comparison with the results of earlier burnout experiments, using a similar test section with the two tubes concentric, showed the burnout heat flux for the eccentric model to be as much as one-third lower for comparable mass velocities and bulk outlet enthalpies.

Analyses of pressures in K-Reactor downcomers during a transient coolant temperature increase showed that pressures would be at least as high

as the saturation pressure corresponding to the bulk coolant temperature. This would mean that a temperature increase of 10 C above a base of 97 C could cause pressures in the downcomer above its strength capabilities.

Hydraulic tests have been run on old production reactor fuel elements to establish the effect of support modifications on flow. Such tests have also been run on toggle valves for the K-Reactors.

Plutonium Recycle Program - A thermocoupled, short 2% element is being fabricated for irradiation testing at 1600 kW in the Rupture Loop during April.

Three additional PRTR high power density rods were fabricated using 2 wt% PuO_2 - UO_2 Salt Cycle fuel material and segmented rod fabrication techniques. This element is currently undergoing irradiation in the Rupture Loop. An additional recycle fuel rod has been successfully irradiated in the Materials Testing Reactor (MTR).

A test element designed to permit impingement of UO_2 on Zircaloy cladding was successfully irradiated for 24 days in the MTR. A second similar element has been shipped to the MTR for irradiation.

A fuel rod was successfully rejuvenated, after irradiation in the MTR, using Pu-Zr wire enrichment. It has been reassembled into a 4-rod cluster for further irradiation in the MTR. Fuel rods for PRTR elements are now being fabricated, using a Pu-Zr wire through the center of cored UO_2 pellets.

The Fuel Element Fabrication Code is being modified to accept process flow sheets which have been developed for a 1 ton per day mixed oxide, vibrationally compacted fuel process. The flow sheet for a pellet mixed oxide process is being developed.

PRTR HPD Core - The maximum burnup of the seven UO_2 -2 wt% PuO_2 high power density elements currently in the PRTR is 2814 MWd/ton_U.

A status report on the transition to the PRTR High Power Density (HPD) Core was issued.

Corrosion and Water Quality - Zircaloy-2 coupons, autoclaved (400 C, 1500 psi), vacuum annealed (gettered system to completely dissolve the oxide) and reautoclaved, showed postanneal oxidation rates 1.3 to 2.5 times normal rates on etched Zircaloy-2.

Plutonium dissolution tests have shown that concentrated (85%) H_3PO_4 at 156 C dissolves high-fired UO_2/PuO_2 without excessive attack on stainless steel or Zircaloy-2.

Out-of-reactor flow tests with extensively fragmented ceramic cores of irradiated PRTR fuel elements have shown that the fragments do not escape from large defects (1/16 x 6 in. slit) under ordinary flow conditions. Impressed vibration, however, has resulted in a significant increase in washout from an irradiated, swage-Vipac fuel rod.

Two tests to study corrosion in crevices of electrically heated rods with geometries similar to those on the surfaces of PRTR fuel elements have shown excessive oxide formation and Zircaloy-2 penetration (~7 mils) at bundle bands. The corrosion is believed to be the result of concentration of LiOH by localized boiling.

No significant corrosion was noted on the dump valve or shim well walls during pilot plant tests prototypical of PRTR Phase I Boron Shim operation.

PRTR Pressure Tubes - Stress rupture tests on Zircaloy-2 indicate that the time-to-rupture increases and then decreases with increasing fast neutron exposure.

Crack propagation tests indicate that the stress necessary for propagation of a given size crack is increased by temperature and by fast neutron exposure.

Reactor Components Development - The influence of changes in steam generator pressure on rate of fretting has been determined.

Plutonium Ceramics Research - Preliminary tensimetric studies in AmO_2 suggest a two-phase region near the sesquioxide composition, as in the PuO system. PuO_2 and niobium were found to be chemically compatible in the molten state.

Examination of stainless steel-20% PuO_2 extruded cermet pins indicates a dense continuous matrix with very little damage to PuO_2 particles. Both high density (82% TD) and low density (76% TD) extrusion billets appear to produce good pins. High density and low density stainless steel-20 vol% PuO_2 cermets being irradiated in the MTR attained burnup of 1.6% of plutonium. Low density cermets heated to 900 C in vacuum for 1000 hr were more dimensionally stable than high density cermets heated at the same conditions. Pneumatic impactation, without extrusion, was used to directly produce integrally clad and capped stainless steel- UO_2 cermet pins.

Approximately 8 kg of mechanically mixed and impacted UO_2 -20 wt% PuO_2 is being prepared for irradiation in EBR-II in a cooperative program with Argonne National Laboratory.

Ceramics (Uranium) Fuel Research - A relatively partial molar enthalpy of uranium in UO_{2-x} between 1800 and 2400 C is estimated to be 20 kcal/mole. This value indicates that UO_{2-x} is a defect-type compound. The movement of tungsten markers in molten UO_2 during short irradiations in which the fuel elements were rotated, suggest that molten UO_2 has a higher viscosity than that experienced in laboratory tests. The cause of the apparent high viscosity is not known, but this effect could be very important to increasing reactor safety during over-power transients.

A reaction zone in the stainless steel cladding on UN-20% PuN fuel capsules does not appear to be a nitride, oxide, or carbide.

The differential thermal analysis apparatus for use to 1700 C was installed in a plutonium glove box. Plutonium ceramics metallography facilities were changed from air to a nitrogen atmosphere. Ion pump installation on the electron microscope was completed.

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Swelling Studies - The third uranium swelling capsule (P-4) of a series of controlled pressure-temperature capsules was charged in a reactor and is operating successfully at 1000 psi and 625 C.

Initial density results on uranium specimens from Capsule P-3, irradiated at 575 C and 1000 psi to about the same burnup as Capsule P-2 (0.13 at. % BU, 450 C, and 1000 psi) show that pressure inhibits swelling at the higher irradiation temperature also.

Irradiation Damage to Reactor Metals - Components for a bench model of a temperature controlled liquid metal irradiation capsule have been fabricated and are presently being assembled.

In-reactor and out-of-reactor creep tests on annealed AISI 304 SS at 650 C and 10,000 psi exhibited rates of 4.4 and 2.2×10^{-6} /hr, respectively, after 900 hr of testing.

Irradiated specimens of BONUS transition joints were tensile tested. They failed in the Zircaloy-2 as did the control specimens. However, the ductility was substantially reduced.

Evaluation of an empirical equation derived from experimental data continues to show excellent correlation between experimental and predicted results for a wide variety of mechanical tests.

ATR Gas Loop Studies - Conductivity tests on foil insulation showed a decrease in the effect of flow and indicate that the leakage flow around the insulation may be the primary cause of the flow dependency.

Components of the second generation gas heater for the model gas loop are now being fabricated.

The rate of evaporation of Haynes Alloy 25 at 1120 C and a pressure of 10^{-4} torr has been decreased by a factor of 2 with the addition of 10 torr hydrogen pressure.

The initial rate and transition to secondary linear oxidation rates for niobium are masked by gas-phase diffusion to the sample. However, the

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rates obtained after a film is formed compare well with those presented in the literature.

Graphite Studies - The first measurements of the radiation-induced reaction of carbon dioxide and hydrogen have been obtained. At 350 C the yields are about 3.0 molecules carbon monoxide and 0.1 molecule of methane per 100 eV of energy absorbed. Water is also a product, but equipment is not available to measure the small amounts formed. Values for the yields of this reaction are needed to predict the effects of traces of oxidants in a high-temperature graphite reactor.

Preliminary results indicate that the in-reactor breaking strain in graphite depends on the strain rate. Low-strain-rate samples achieve higher strains before fracture.

Calculations are under way to develop a theoretical model for displacement damage in boronated graphite.

Metallic Fuel Development - Th-U fuel elements being irradiated in the ETR have attained an exposure of 11,000 MWd/ton with minimum swelling. Irradiation of a second set of elements has been started.

Fuel rods containing submicron dispersions of uranium carbide in uranium are being examined after irradiation to 0.3 at. % BU at a maximum temperature of 500 C. Three samples had brittle clad failures and little swelling, while a single sample with 2 to 5 micron carbide particles showed some increase in diameter.

Plutonium and U²³³ Fueling of a Fast Compact Reactor - Engineering calculations are essentially complete, and the final report is in preparation. Reports are being prepared on two computer programs used in this study.

Fast Flux Test Facility Studies - Authorization was received from the USAEC to expend \$500,000 of FY-1965 funds for conceptual design of a "Fast Flux Test Facility" and to determine its R&D requirements.

The FY-1967 PAC item for FFTF was revised to \$75 million.

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Radiation Effects on Metals - Results of studies on thermally-activated deformation in polycrystalline molybdenum suggest that the controlling mechanism for dislocation motion in the temperature range 223 to 350 K is the motion of jogs in screw dislocations. In the irradiated material, however, the rate-controlling obstacles are thought to be small clusters of interstitials.

Irradiation of rhenium foils to 7×10^{18} nvt ($E > 1$ MeV) at 60 C causes the lattice parameter to increase. The initial preirradiation value of the lattice parameter is regained by annealing the foils at 800 C.

Examination of high magnification electron micrographs, made from rhenium that was irradiated to 7×10^{18} nvt, disclosed the presence of spot defects. This is the first electron microscope observation of neutron damage in a cph metal.

Molybdenum foils irradiated to 1×10^{19} nvt ($E > 1$ MeV) show a barely perceptible decrease in lattice parameter after annealing at 200 C.

The intensities of X-rays diffracted from electropolished surfaces of molybdenum crystals are not affected by irradiation to 10^{19} nvt or low temperature annealing.

Alpha-plutonium samples of significant size have been prepared, which, within certain limits, behave crystallographically as a single crystal.

Customer Work - Tests of graphite-5 wt% B_4C exposed to nitrogen containing 5000 ppm water at 1100 C are being conducted in support of HTLTR. Thermocouples sheathed in molybdenum showed no corrosion effects when tested in the first run of the 314 mockup.

The second run of the HTLTR mockup has reached a core temperature of 260 C. Both the prototype control rod and safety rod have functioned properly up to this temperature.

The balance of the rods required for the plutonium loading of the EBWR were shipped to Argonne National Laboratory. One hundred fifty

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spare rods are being fabricated. Strain gage measurements were made to determine stresses encountered in EBWR Zircaloy-2 during vibrational compaction.

Special Al-Pu core EBWR elements were fabricated for physics testing. Thermal cycling tests being conducted on similar elements indicate both a lengthening and shortening of the rods.

EBWR fuel rods have satisfactorily achieved exposures to 4.8×10^{20} fissions/cm³ (17,000 MWd/ton of fuel). Fifty-seven full-size EBWR rods are under irradiation, and, of these, 21 have attained an exposure of 0.25×10^{20} fissions/cm³ (860 MWd/ton of fuel).

Eighty-four Saxton rods were fabricated and are now being etched and autoclaved in preparation for shipment in early April.

Service work on preparation of potential reactor poison materials was conducted for General Electric Company. Pellets of C-5 wt% Sm₂O₃ and C-0.5 wt% Gd₂O₃ were prepared by cold pressing.

2. Physics and Instruments

Analysis of exponential and PCTR experiments in support of N-Reactor coproduct studies continued. Discrepancies between theory and experiment have been greatly reduced by the use of transport theory instead of diffusion theory. For tube-in-tube fuel in graphite-water lattices, however, a comparison of diffusion and transport theory calculations showed good agreement in k_{∞} values. The final report on the exponential assembly experiments has been written.

The surface contamination tester developed to monitor uranium contamination on finished fuels has been installed, checked out, and is undergoing production evaluation by N-Reactor fuels quality control personnel.

Laboratory spectrophotometric analysis of the arc associated with end cap welding of experimental Al-Si fuels readily distinguished elements whose surfaces were contaminated with trace quantities of water, oil, and paraffin.

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Criticality experiments were continued with PuO_2 -polystyrene compacts to evaluate effects of the plastic coating material on the blocks and to study the neutron spectrum by foil counting techniques. The fuel contained 8 wt% Pu^{240} .

Pulsed neutron source measurements were made on a bare assembly to obtain comparisons at different Pu^{240} contents. For a change of Pu^{240} content from 2.2 to 8.08 wt% the β/λ value changed from 1763 to 1357 sec^{-1} and the neutron lifetime changed from 1.2 to 1.6 μsec .

Subcritical neutron multiplication measurements were started with 1.96 wt% U^{235} enriched tubular fuel elements to provide data for nuclear safety guidance in processing and storing of enriched N-Reactor fuels.

A Monte Carlo calculation was made to determine critical mass of Pu^{238} metal in spherical geometry and reflected by 10 cm of stainless steel. A value of 2.7 kg was calculated to be critical at a density of 19.6 g/cm^3 .

The use of a copper crystal monochromator for the triple-axis neutron spectrometer shows promise for improved performance.

Work continued on the analysis of the differential scattering of neutrons from zirconium hydride.

Progress was made on analytical computer programs for fast-neutron total cross sections and preparations were made for another measurement series in April.

The mass spectrometer equipped for ion-counting was able to place an upper limit at 95% confidence of 0.3 ppm U^{233} in near-natural uranium samples.

A detailed analysis of a typical Pu-Al critical assembly was made using HFN code to determine the effect of small changes in the cross sections used. Results showed that in addition to the obvious importance of the thermal absorption and fission cross sections, the fast group parameters are also quite important.

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Physics code development work continued with improvements resulting in reduced programming time for INPUT/LISTIN/UNIT. Studies were initiated to evaluate codes for describing the decay resonance radiation in gases and to prepare a code for high energy photon transport.

Survey calculations are being made to determine optimum driver target configurations of Pu-Al drivers and ThO₂ targets. Planning of PCTR experiments is under way.

Flux flattening for the Pu-Al-polyethylene Phoenix fuel experiment was successful in the PCTR. Mathematical analysis shows the necessity for specific treatment of the lattice voids rather than homogenized treatment as a reduction in density and also shows the angular importance of neutrons incident on a thin plate.

An improved method of calculating the effect of grain size on resonance absorption is now operative. Although originally of interest in BeO-PuO₂ systems, the calculational method could be also used with mixed oxide fuels.

An analysis of fuel cycle costs for H_x Pu in 50-200 MW PWR's indicates the attainment of plutonium fissile/U²³⁵ value ratios greater than unity.

Physics analysis of a fast fuel test facility continued. Reports were issued covering neutron statics, i. e., critical size and reflector control worth and also a preliminary investigation of kinetics properties of the FFTF.

The second run of the HTLTR mockup was started. Prototype control and safety rods as well as modified heater elements are being tested. Work has started on setting up a system for testing materials in 1100 C nitrogen carrying various concentrations of water vapor.

HTLTR reactor calculations to provide data for driver fuel loading, fuel specifications, and safeguards analysis are continuing. Calculations were made to predict the temperature coefficient compensation that can be obtained by placing Gd₂O₃ in the reactor.

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Calculations for the PRTR HPD core were extended to include a concentric ring model in addition to a solid rod equivalent of the 19-rod cluster. It was found that the calculated η_f is insensitive to the model used. Planning for an HPD PRCF experiment was started.

Calculations using multithermal groups in HFN have been performed for a series of EBWR-fueled approach-to-critical lattices. The Δk_{eff} 's between one and five groups varies between 0.2% (0.55 in. lattice spacing) and 3% (0.80 in. lattice spacing).

Several revisions have been made to the VESTA code to improve its usefulness including the possibility of studying fueling breeders with U^{235} to make additional plutonium available. To solve the problem of likely exchange of fuel between different reactors wherein the relevant decisions will be based primarily on economic factors, linear programming appears to offer a useful technique for simulating the situations involved; namely, best decisions regarding economic optima in view of practical constraints. A simple demonstration problem has been selected and sample calculations have been made with the LP-90 code. Plutonium values have been related to some generalized reactor parameters; namely, the amount of parasitic material in the reactor and the amount of moderation.

Initial analyses of reduced density fuels using plutonium enrichment have been made. Additional Monte Carlo calculations were performed to establish a satisfactory means of applying the RBU code to ascertain fuel geometry effects in plutonium values.

A series of in-core neutron flux monitor tests at KW-Reactor demonstrated titanium encased MgO cables generate markedly fewer noise currents than cables previously used.

A hypersonic transducer for nondestructive testing uses was produced by diffusing copper into indium doped CdS. The transducer appears to have two resonant frequencies: 25 Mc, corresponding to the diffusion depth, and

1.8 Mc, corresponding to the total crystal thickness. The higher frequency may be increased by reduced diffusion time.

A relationship was developed with which radiation dose rates can be calculated for two sides of a plane boundary between two different media, one of which contains radioactivity.

A "thermal shorting switch" installed into the fast radiation calorimetry system allows equilibrium to be obtained in 3 to 4 hr rather than the usual 20 to 30 hr.

A new technique for molding tissue equivalent plastic is being used with good success in making tissue equivalent ionization counters.

X-ray filters of metal salts and of a precise thickness were made by forming a gel of an aqueous solution of the salt. These filters (Ross filters) provide increased resolving power for specific X-ray energies.

The feasibility of routine field sampling using Xe^{133} as an atmospheric tracer was demonstrated. Significant samples of xenon were obtained out to 1 mi from the source in a dual zinc sulfide-xenon release. Modification of the sampling equipment eliminated the malfunctioning that occurred in the initial test held previously. Field preparation time was reduced 2 hr over the time required for the previous test.

3. Chemistry

The velocity of the impinging stream was shown to be the controlling variable in the abrasive erosion caused by a thoria slurry agitated by spargers or air lift circulators.

A protective slag cover or an inert atmosphere is necessary to avoid oxidation reactions in the reduction of bismuth nitrate to metal with carbon.

Molten bismuth was successfully sprayed with a two-fluid nozzle; a particle size less than 50μ can be achieved.

Calculations indicate inclusion of internal fins in a container for solid-state radioactive materials will markedly improve heat transfer and permit a significant increase (6-fold) in loading.

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A "bang-bang" technique for computer control during startup of a chemical process was incorporated in the GE-412 and tested on a column pulsing system. Steady state was achieved in about 1.5 sec versus 15 min by conventional techniques.

The presence of stainless steel corrosion products markedly increases the corrosivity of thoria dissolvents to AISI 304-L SS.

Spray calciner-melter runs were made using a simulated Nuclear Fuels Service's first cycle waste with added phosphoric acid as feed. Deposition of solids on calciner walls was minor at M^+/P ratios of 3 and 2.7 but increased markedly at M^+/P ratios of 2.3 and 2. Filter plugging was more severe at the lower ratios.

Nine fuel pins fabricated from material produced by Salt Cycle process treatment of irradiated UO_2 - PuO_2 fuel are now being irradiated in the PRTR.

Five hundred impactions have been made with the 1120-F Dynapak machine without failure of any equipment that would require remote maintenance in a hot cell.

An irradiated N-Reactor target element was heated inductively in a partially shielded quartz furnace to observe (with colored movies) the rupture of the can and flow of molten core.

Low but detectable concentrations of Sr^{90} have been noted in recent ground water samples from wells monitoring the Redox S-7 process condensate crib. A change to use of the replacement crib was recommended.

Oxides of plutonium formed by ignition of plutonium rods at 600 C were found to be coarser than reported in the literature.

Scouting studies are in progress to evaluate methods for removing sulfate ion from the Waste Solidification Engineering Prototype glass melter off-gas condensate. Precipitation as calcium sulfate and crystallization as sodium sulfate were tested.

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The three-part containment vessel for the Containment Systems Experiment was completed and pressure testing is imminent. The design of the reactor simulator vessel is on schedule.

A new plotter program was written for the off-gas disposal to ground research study which permits plotting of various flow pattern developments considering soil anisotropy.

The "STREAM" and "GENORO" computer programs are being used with actual field data to obtain knowledge on the fitting and calculating of soil permeabilities from multiterm polynomials.

In the studies of anion-soil reactions, attempts to fit dichromate and phosphate breakthrough curves to chromatographic equations were not successful.

Based on a review of theories of turbulent deposition of particles, a model approach is planned to include better estimates of particle eddy diffusion coefficients, particle approach velocities, and particle concentration at the edge of the boundary layer.

Removal of polonium into 8 wt% H_2 -Ar during reduction of bismuth oxide at 1000 K was slow. Physical changes in bismuth oxide on heating at 1000 C did not correlate with time at temperature.

Thermogravimetric studies on Pu(IV) oxalate decomposition disclosed (1) less carbon in the product with increased calcination time and temperature, (2) the desirability of air flow and undesirability of CO_2 flow during calcination, and (3) an increased rate of calcination on storage (aging) of the oxalate.

Pressurization (from water radiolysis) in storage of radioisotopes on zeolites is being studied under conditions simulating actual storage conditions.

The manganese removal process for upgrading promethium concentrates was successfully applied in Strontium Semiworks equipment.

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DTPA was found to be the most satisfactory of six complexants tested for the stripping of yttrium from D2EHPA-TBP-solvent.

Spray calcination runs have been started to evaluate this unit operation for producing thoria.

Cold pressed thoria target elements have been completed for reactor charging.

Sulfate has been found to have little effect on phosphate glass characteristics. Increasing the (simulated) fission product content to 75% increased the glass melting point. Glasses made from simulated NFS waste have been prepared.

Electrochemical purification of Salt Cycle melts is believed superior to recrystallization techniques.

The second multidimensional gamma ray spectrometer is now operative and appears to be slightly better than its predecessor. A computer program for data reduction was written.

Application of multidimensional gamma ray spectroscopy to activation analysis of blood permitted measurement of Cs, Fe, Co, Zn, and previously unreported Ag, Sb, and Sc. Eight radionuclides were also measured in Alaskan caribou liver, as were seven isotopes in Pacific Ocean euphausiids. These measurements are important to ecological studies.

A dual tracer Xe^{133} -ZnS experiment was performed in studies of atmospheric diffusion.

A new dye, FDC Blue No. 2, was found effective in protecting agarose-water jellies from radiation effects.

Preparation of the Radiochemistry Facility A-Cell for promethium purification is proceeding. The chemical conversion function, recently intended for A-Cell, will now be housed in lightly shielded gloved hoods as originally planned.

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Compatibility studies of promethium stand-ins and various metals were delayed because of failure of sealed Vycor containers.

Promethium metal and fluoride were compared as to their reentry burn-up characteristics in computer studies. From other calculations, it was concluded that "super-alloy"-clad promethium would survive a launch pad missile fuel fire-ball.

4. Biology

Tests of the toxicity of a spent reactor tube decontaminant on chinook salmon fingerlings demonstrated a variety of mortality responses obtainable with different species of fish. These studies emphasized the importance of on-site tests of such materials with local fish.

Two swine which had received $125 \mu\text{Ci Sr}^{90}$ /day for about 3 yr were killed, with symptoms highly suggestive of leukemia. In addition to the four confirmed cases of leukemia in this group, there are now four probable cases awaiting histopathologic confirmation. It is of particular interest that there is a close genetic relationship among six of these eight swine.

Pregnant ewes fed Cs^{137} daily since December have body burdens about nine times the average daily intake. This is only about one-half the level observed in male sheep 1 yr ago.

Immediately following exposure to uranium ore dust the uranium and thorium of the deposited ore particles in rat lungs were virtually in secular equilibrium. As earlier reported, this ratio of thorium to uranium increases about 9-fold after several months' retention in the lung, indicating the need for separate consideration of these radioelements in evaluating the hazard of exposure to uranium ore dust.

A technique was developed for measuring kinetics of amino acid transport in N. crassa. Preliminary results suggest that L-tryptophan transport does not require an external energy source.

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Periodic sampling of the shrub, Mountain Maple, of the Blue Mountains during 1964 showed no increase in leaf concentration of Cs^{137} during the period the leaves were on the shrub. Either the major accumulation occurs at the time the leaves are first unfolded, or the rate of contamination through the year is offset by concomitant loss of activity from the leaves.

Statistical analysis of Eskimo Cs^{137} body burdens was begun. Assuming a constant Cs^{137} intake during several periods of the study, one can estimate an effective half-life for Cs^{137} of about 70 days.

5. Radiation Protection

Development work on the action of fissionable material in contact with Lexan plastic continues to show promising possibilities for the use of this system as a personnel neutron dosimeter and for the detection of extremely small quantities of fissionable material.

On two occasions during the month the distribution of reactor effluent in the Columbia River was studied with the use of a dye tracer. One test showed downstream concentrations of effluent from B Area; the other the time of travel from D Area.

Preliminary contacts were made to use the mobile whole body counter to obtain data on local school children and fishermen.

Studies directed toward increasing the neutron sensitivity and decreasing the detection limit for LiF gave encouraging results and indicate this material has a good probability of meeting the performance criteria for personnel neutron dosimeters.

TECHNICAL AND OTHER SERVICES

The levels of radioactivity found in the Columbia River and in fish were normal. The concentrations of fallout materials in the atmosphere increased slightly and reflects the expected Spring influx.

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Study of the possible consequences of a flow stoppage in the Rupture Test Loop of PRTR indicated that very prompt action by the operator would be necessary to save the fuel element.

The AEC Division of Biology and Medicine was provided assistance in evaluating the performance of criticality dosimeters used at various AEC contractor sites. The Hanford-developed criticality dosimeter was among those used at Oak Ridge with bursts from the Health Physics Test Reactor.

A previously used sampling procedure for steam generator tube bundles was evaluated for use in an April 1965 outage.

Modifications leading to increased efficiency and versatility have been completed on the computer program which calculates the transient heat transfer characteristics of a source-laden slab of material.

The computer program for studying certain functions in inelastic media has been modified so that input, fitted, and attenuated pulse data are plotted automatically.

A study was initiated to assess seven separate indices of plutonium burnup employed in interpreting certain PRTR experiments and develop a "best" single index.

A nonlinear model was developed to express the relationship between the extent of cracking and the force applied to reactor models.

The GEM program is being modified so that the weights used in the least-square estimation of nuclide decay rates reflect instrument instability as well as counting variation. Mathematical work on this problem has been completed.

Specifications are being written for a computer program to convert counting data for gastrointestinal studies into corrected counts-per-minute when more than one radionuclide is present.

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An exponential model was formulated to express the relationship between the gross beta and P^{32} retained by Columbia River whitefish.

SUPPORTING FUNCTIONS

Plutonium Recycle Test Reactor output for March was 1067 MWd for an experimental time efficiency of 65% and a critical efficiency of 52%. There were seven operating periods during the month, two of which were terminated manually, four were terminated by scrams, and one extended through month-end. A summary of the fuel irradiation program as of March 31, 1965, follows:

	<u>Al-Pu</u>		<u>UO₂</u>		<u>PuO₂-UO₂</u>		<u>Other</u>		<u>Program Totals</u>	
	<u>No.</u>	<u>MWd</u>	<u>No.</u>	<u>MWd</u>	<u>No.</u>	<u>MWd</u>	<u>No.</u>	<u>MWd</u>	<u>No.</u>	<u>MWd</u>
In-Core			3	1 174.5	76	16 401.3			79	17 575.8
Maximum				414.7		463.7				
Average				391.5		215.8				
In Basin	7	572.5	30	3 806.2	62	9 580.6			99	13 959.3
Buried							1	7.3	1	7.3
Chemical										
Processing	<u>68</u>	<u>5 465.8</u>	<u>35</u>	<u>1 965.8</u>	<u>—</u>	<u>—</u>	<u>—</u>	<u>—</u>	<u>103</u>	<u>7 431.6</u>
Program										
Totals	<u>75</u>	<u>6 038.3</u>	<u>68</u>	<u>6 946.5</u>	<u>138</u>	<u>25 981.9</u>	<u>1</u>	<u>7.3</u>	<u>282</u>	<u>38 974.0</u>

(Note: MWd/Element x 20 $\approx$ MWd/ton_U for UO₂ and UO₂-PuO₂)

Heavy water loss and indicated helium loss for the month were 468 lb and 118,000 scf, respectively.

A total of 1963 craft manhours were utilized during the March reactor outages as follows:

Repair	736
Improvement	535
Operations Assistance	363
Preventive Maintenance	329

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A standard 58-in. length Mark I fuel element in the Fuel Element Rupture Test Facility containing six outer rods of 2% PuO_2 in UO_2 fuel material obtained by the Salt Cycle process was charged for the reactor startup of March 17. The estimated power generation of the Salt Cycle element was 875 kW at full reactor power.

Arrangements have been made with the Redox Operation of GE-CPD to conduct, in late May or early June, a chemical processing campaign on spent PRTR fuels. Approximately 8 kg of plutonium containing 20% Pu^{240} would be recovered from the selected fuel elements.

A total of 25,868 hr of shop work in the Technical Shops Section was produced with a backlog at the end of the period totaling \$195,081 or 21.675 hr. To meet emergency requests for service, it was necessary to work 1409 overtime hours or 6.5% of the total hours produced in Technical Shops.

Total productive time in the Laboratory Maintenance Section was 30,200 hr of 32,400 potentially available. Craft overtime worked during March was 3.4% of total available hours. Manpower utilization (in hours) was as follows:

A. Shop Work		2 800
B. Maintenance		10 500
1. Preventive Maintenance	3 000	
2. Emergency or Unscheduled Maintenance	2 000	
3. Normal Scheduled Maintenance	5 500	
C. R&D Assistance		16 900

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REACTOR AND MATERIALS TECHNOLOGY DEPARTMENT
MONTHLY REPORT - MARCH 1965

TECHNICAL ACTIVITIES

A. FISSIONABLE MATERIALS - 02 PROGRAM

1. Metallic Fuel Development

Alternate Uranium Composition. Operating conditions for driver fuel elements of a single tube design and extended goal exposure require development of a fuel material more swelling resistant than the 150 Fe - 100 Si N-fuel composition used in the current tube-tube design. Five alloys for N-driver single tubes ranging in composition from 150-350 ppm Fe, 100-350 ppm Si, and 250 and 800 ppm Al were beta and gamma heat treated by the salt-salt process, and the following evaluation tests are in progress: (1) examination of microstructure and bond zone, (2) hot hardness tests at 300 and 400 C, (3) determination of recrystallization temperatures after 21.4% reduction in area, and (4) stress cycling tests at 400 and 500 C.

These tests will be completed in April. Hot hardness data and stress cycling tests completed to date are shown in the following table. It is planned to run these tests to 5000 cycles for each material and examine the microstructure for evidence of intergranular cavities or voids. As one basis for comparing these materials, the number of intermetallic compounds and their composition for each alloy was calculated and are shown in the second following table. Several simplifying assumptions were made, the most limiting being that the compounds were all of equal size and that all had been precipitated from solution. Such a comparison, however, does indicate that if keying of dislocations by second phase particles is primarily responsible for elevated temperature deformation behavior and irradiation performance, the materials would fall in three groups: Alloy 1; Alloys 2, 3, and 4; and Alloy 5. Notice that the hot hardness of alloys 2, 3, and 4 are similar. Slight increases are generally observed for gamma treated material in which all the alloy addition is taken into solution at heat treating temperature. The stress cycling data to date shows only that those alloys containing aluminum have greater resistance to plastic deformation at 400 and 500 C.

ELEVATED TEMPERATURE MECHANICAL PROPERTY TESTS OF URANIUM FUEL MATERIALS

Alloy No.	Uranium Base Material			Heat Treatment	Hot Hardness ⁺ 300 C	Kg/mm ² 400 C	No. of Cycles	Stress Cycling	
	Fe ppm	Si ppm	Al ppm					Total Strain - % in 1.125 Gage*	
								400 C **	500 C ***
1	150	100	-	Beta	94.6	60.3	500	.71	.53
				"			1000	.98	1.69
				"			5000	4.62	
				Gamma	93.1	60.8	500	.44	1.51
				"			1000	.62	1.51
				"					
2	250	350	-	Beta	98.5	70.8	500	.27	.89
				"			1000	1.15	2.66
				"					
				Gamma	101.7	75.1	500	.35	.27
				"			1000	2.04	.62
				"					
3	250	250	250	Beta	94.8	71.7	500	.27	.09
				"			1000	.44	.27
				"					
				Gamma	96.9	75.0	500	.09	.09
				"			1000	.18	.35
				"					
4	350	-	800	Beta	105.0	68.8	500	0	.35
				"			1000	.18	.35
				"					
				Gamma	101.2	74.6	500	.27	.09
				"			1000	.35	.62
				"					
5	350	350	800						

+Average of tests run at 400, 600, and 800 pound loads, 4-hour loading, 10 mm WC ball.

*Deformation measured on specimen after cooling to room temperature.

**Stress level 30,000 psi - cyclic stress $\pm$ 3000 psi.

***Stress level 15,000 psi - cyclic stress $\pm$ 3000 psi.

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CALCULATED NUMBER OF INTERMETALLIC COMPOUND PARTICLES IN URANIUM BASE ALLOYS

Alloy	Number of 0.2μ cube particles/cc x 10 ¹²				Volume Percent Compound	Total Particles 10 ¹² /cc*
	<u>U₃Si</u>	<u>U₆Fe(Si)</u>	<u>UAl₂(Fe)</u>	<u>Total</u>		
1	0.098	0.674	--	0.772	0.62	0.098
2	1.11	1.15	--	2.26	1.81	1.11
3	0.69	1.09	0.33	2.11	1.69	0.99
4	--	0.89	1.29	2.18	1.74	1.29
5	1.08	1.40	1.25	3.73	2.99	2.33

*Assuming U₆Fe(Si) grows rapidly to a size that is not effective in gas or dislocation pinning and not effective in strengthening or hardening.

Studies are in progress to evaluate the irradiation performance of uranium base alloys in terms of composition, fuel fabrication history, corrosion behavior, and irradiation history. Forty test capsules and five temperature monitoring capsules are being irradiated in five process tubes. The irradiation includes 120 test specimens of 17 uranium base alloys containing small additions of Zr, Nb, Mo, Al, Si, Fe, P, and C in various conditions of heat treatment. All thermocouple systems are operating satisfactorily, and the initial data shows the maximum uranium temperatures at full reactor power are in the range 470-490 C. A goal exposure of 3000-4000 Mwd/ton is planned for these fuels.

Target Element Development. A study to determine the effects of a high temperature reactor excursion on Zr-2 clad Al-Li alloy target elements is continuing. Examination of three irradiated targets which failed when subjected to heating tests up to 930 C was completed. In addition to metallographic results reported in the previous monthly concerning the reaction of the molten Li-Al core and the Zr-2 cladding, further examination has shown evidence of hydriding at various locations in the Zr-2. However, there are no indications of association of the hydrides with the cladding failures. Evidence from the radio-metallurgy examination of the heated targets and from high temperature pressure testing of unirradiated Zr-2 tubes leads to the conclusions that Li-Al target elements fail due to the internal gas pressure resulting from the release of helium and tritium from the target alloy during heating.

Two Al-3.5 wt% Li target elements irradiated in a pressurized water loop to an exposure of 6 GVR* have been examined. The Al-Li metal cores were double contained with an inner can of 0.030-inch thick aluminum and an outer can of 0.030-inch thick Zr-2. The Zr-2 cans were 8.9 inches long with an outside diameter of 0.755 inch. Measurements show no changes in dimensions or density. Metallographic sections from one of the rods shows evidence of a microstructural difference in the aluminum can, at the core-can interface, to a depth of approximately 0.001 inch. However, the original interface is readily visible, and it is doubtful that bonding has occurred. Comparison of the pre- and postirradiation metallography of the Li-Al alloy shows no major changes in microstructure.

Zircaloy-2 Corrosion in Molten Aluminum. The corrosion of Zr-2 in molten aluminum is being studied in the temperature range of 700 C to 1100 C. Corrosion of the Zr-2 is nearly uniform, and the approximate penetration rates at 700 and 900 C are 0.4 and 3.5 mils per minute, respectively.

Experiments to date show that the corrosion rate slows somewhat with time. This results from the buildup of Zr-Al intermetallics in the molten bath and, more importantly, on the surface of the Zr-2 coupons. The presence of an initial autoclave film also slows the corrosion rates slightly. The following data illustrates the previous statements.

Temp., °C	Time, min.	Average Penetration Rate - mils/min	
		Autoclaved Surface	Etched Surface
700	5	0.7	1.0
"	30	0.18	0.42
"	50	0.35	0.45
"	80	0.25	0.27
"	100	0.31	
900	5	4.6	3.1
"	9	3.0	3.2

Experiments are conducted in clay crucibles using resistance heating at 700 and 800 C and induction heating from 900 to 1100 C. Air atmosphere covers the molten aluminum.

Protective Coatings for Zircaloy-2. Flame sprayed coatings and nickel plating as well as nickel and silicon additions to the molten aluminum are being evaluated as deterrents to the corrosion of Zr-2 by molten

*GVR = Gas Volume Ratios.

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aluminum. Nickel and silicon additions of 3 and 12 wt%, respectively, effect a distinct reduction in corrosion rate. Flame sprayed coatings that are protective in pure aluminum at 700 C are tungsten carbide, zirconia, molybdenum, tantalum, and most notably alumina.

Coatings that do not perform well are nickel plate, steel, nickel, aluminum-iron bronze, aluminum and copper. The coatings which perform well at 700 C will be re-evaluated at 1100 C.

Elevated Temperature Reactions Between Target Core and Various Poisons. Experiments are being conducted to determine the possible interactions at temperatures from 500 to 700 C between the 1-1/2% Li-Al target alloy and nuclear poison materials, including a Cu-40 at% Cd alloy, cadmium oxide, and gadolinium. The results so far indicate no significant reactions between the target core and either cadmium oxide or gadolinium at temperatures to 700 C and times to 15 minutes, but a definite dispersion of the Cu-40 at% Cd alloy in the target core occurs at 700 C in times as short as five minutes. In 15 minutes at 700 C a target core sample completely dissolved a rod of the cadmium alloy equal to about 2/3 of the core volume or about 1/2 of the total volume.

N-Reactor Fuel Support Development. Fatigue properties of 1/4, 3/16, and 1/8 inch wide tube-tube inner supports are currently being obtained as a part of a mechanical testing program designed to evaluate the effects of strip thickness and width on the properties of fuel element supports. Test data will be used as a basis of establishing an optimum support design for both inner fuel tubes and target elements.

Forged "N" Outer Tube Support Development. Die quenching during the forming cycle of forged supports does not entirely eliminate the problems during part stripping. The die is being modified to utilize the rigid cross section (tee) of the support, and the mechanical stripping features of the press to remove the formed part from the die cavity.

Original design relied on the remaining feed wire on either side of the forged part for stripping. With no die quench this section of wire remained too hot and had no strength for stripping. A six-second die quench cools the wire sufficiently to strip the part. The bending moment imparted to the support tabs, through the feed wire, tilts the tabs enough to cause inconsistencies in tab welding behavior during assembly with fuel pieces. The die is being modified to utilize the press knockout working against the tab forming surfaces to provide the stripping action. This modification will reduce the bending moment on the tabs to a negligible amount.

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ENCLOSURE 8

Tritium Diffusion Through Aluminum Cladding on Aluminum-1% Lithium Target Elements. Experiments are being conducted to determine tritium evolution rates through aluminum clad target elements with anodized and nonanodized outer surfaces.

An anodized 30-mil C-64 aluminum cladding, previously heated at 350 C for 14 hours, was subjected to an additional 14-hour run at 350 C. The initial rate (0 to 2 hr) was about equal to the previous initial rates ($\sim 1 \times 10^{-3}$ cc/day from an 8-inch, 0.68-inch OD cladding). Between 7 and 14 hours, the rate was higher (3.4×10^{-3} cc/day). Total tritium in the core is 145 cc, STP.

The core was heated to 375 C. Sometime during the run (>1 hr $<3\frac{1}{2}$ hr) the cladding ruptured, releasing the gas contained in the 0.25-inch diameter center hole. (1 atm helium + gas released in 350 C and 375 C runs). Total pressure in the can at the time of rupture was between 35 (initial helium pressure) and 43 psi. The run was terminated and the target was removed from the furnace. Visual examination failed to indicate the area of rupture.

2. Corrosion and Water Quality Studies

RTD Corrosion. Corrosion failures of the copper wire connections in the resistance temperature detectors (RTD) monitoring the N-Reactor outlet water temperature were investigated. Cause of failure was identified as corrosion of the copper wires by residual soldering flux. Proper removal of the flux eliminated the problem. No other corrosion problems on the RTD assemblies were noted.

Low Temperature Hydriding of Zircaloy-2. Zircaloy-2 coupons with various pretreatments have been exposed to wet hydrogen (97% H_2 - 3% H_2O) for four months at 100 and 200 C. A hydride case was observed on some samples at both 100 and 200 C in the area of freshly punched holes or areas that were vapor blasted. Hydride layers up to 42 mils were observed. No hydriding was observed on preautoclaved specimens. These results confirm the hypothesis which initiated the experiments, that hydriding can occur in the presence of water if conditions for protective oxide formation are unfavorable due to the low temperature or asperities on the metal surface.

Decontamination of N-Reactor Heat Exchanger. Preparations and laboratory work for the decontamination of N-Reactor heat exchangers were completed during the past month. Time and temperature limits were determined for the inhibited sulfamic acid solution specified. The solution is stable at 70 C for approximately 12 hours; after this, the inhibitor breaks down and the solution becomes extremely corrosive

to work-hardened areas of 304 stainless steel, causing excessive intergranular attack in cold worked areas, and to 400 series stainless. Reducing the temperature to 50 C after four hours at 70 C, the solution was found to remain stable in excess of six days. A foreign ion effect was also observed in the corrosion characteristics of the acidic solution. Chromium ion was observed to accelerate the corrosion rates on Type 400 stainless steel and in cold worked areas.

Decontamination of steam generators 2-A and 2-B in N-Reactor was started March 15, 1965. Radiation readings were obtained prior to decontamination on the primary and the secondary sides of the steam generators as well as on the cell piping. During the decontamination several operational difficulties occurred which diluted the alkaline permanganate solution to 90 g/l of NaOH, which after about one hour was further diluted to 40 g/l, rather than the prescribed 180 g/l. The acid solution was circulated at the desired concentration of 90 g/l. The net result was that the film and contamination were removed effectively from the carbon steel portions, but ineffectively from the stainless steel portions. The actual extent of removal is difficult to estimate because the low readings were affected by high backgrounds. The average of 10 readings with a bazocka on the cell piping (carbon steel) indicated a decontamination factor (D.F.) of 9; the D.F. on the steam generator was about 1.6.

The results of this decontamination are in agreement with all previous experiments and evaluations. To have an efficient decontamination of stainless steel, it is necessary to treat with concentrated (180 g NaOH and 30 g KMnO_4 per liter) alkaline permanganate for one to two hours at 105 C. The alkaline permanganate during N-Reactor decontamination was too dilute to be effective. To decontaminate carbon steel, the preliminary treatment with alkaline permanganate is not required; only the acid step is needed. During N-Reactor decontamination the acid remained at sufficient concentration and the carbon steel was effectively decontaminated.

N-Reactor Analytical and Sampling Equipment Review. A review of the N-Reactor analytical system and sampling procedures was performed during the previous reporting period. One chloride analyzer for the primary system was placed in operation. The sensitivity of the analyzer was doubled so that it reads 0 to 200 ppb full scale instead of the previous 0 to 400 ppb. Work is continuing on the remaining two automatic chloride analyzers.

The hydrazine analyzers are potentiometric and have not operated satisfactorily, presumably due to the inability to provide the required

inlet temperature and pressures. This problem was temporarily obviated by converting a hardness analyzer to a wet chemical hydrazine analyzer. The converted analyzer was calibrated and put on line monitoring eight points in the secondary system. Work is continuing on the potentiometric hydrazine analyzers.

In general, both analyzers appeared to operate satisfactorily while they were on line. The increased sensitivity of the chloride analyzer will be valuable during future operation. Some of the burden on the manual section of the laboratory will be relieved with these analyzers.

Hydride Formation in K Process Tubes. Significant hydriding was found in two Zircaloy-2 tubes at KW Reactor. Tubes 3065 (installed August 1961) and 3075 KW (installed August 1963) suffered brittle fracture during removal after being identified as water-leakers. Vacuum analyses showed high hydrogen content (400-1200 ppm H_2) in the rear nine feet of the tube and low hydrogen content (<50 ppm H_2) in the remainder of the tube. The hydrogen was concentrated on the internal surfaces, with layers of pure zirconium hydride, 2-4 mils thick. Immediately below the zirconium hydride layer was a thin layer of metal with a high concentration of hydride; but the hydrogen content of the remainder of the metal, including the outer surface, was within normal limits.

The reason for the heavy hydriding is not known, although several possible causes have been suggested. The tubes were not autoclaved prior to charging and, therefore, did not have the protective zirconium oxide film; any thin oxide films present may have been damaged by scraping, exposing active metal; fluorides (from a proprietary decontaminating agent) may have been present; the zirconium was in intimate contact with other metals. Damage to the film or localized corrosion may have facilitated the diffusion of hydrogen (from corrosion reaction) into the metal.

Work on this problem includes: (1) examination of additional Zircaloy tubes from KW, KE, and C Reactors, to determine the approximate extent of hydride formation throughout the reactors; (2) analyses of Zr-2 coupon samples that have completed 3 to 12 months of exposure in CSDU tests in 1706 KE single-pass facilities; (3) nondestructive examination (by Probolog [eddy-current] and other methods) of most Zr tubes in the K Reactors; (4) testing of Zr samples with various suspect surface pretreatments; (5) installation of Zr coupons and pieces of actual process tubing (with various surface pretreatments) in downstream in-reactor locations; and (6) extension of present testing of hydriding in steam H_2 mixtures in Chemical Metallurgy Unit laboratories)

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to include lower temperatures representative of the single-pass-cooled reactors. Coolant Systems Development Unit will direct the work of Items 2, 4, and substantial portions of Item 5, and will also assist in evaluation of data from Item 1. Corrosion probes to monitor extent of Zircaloy hydriding are being procured and will be installed in one of the in-reactor tubes.

Corrosion and Film Formation in Raw Water. A half-plant test of reactor cooling with raw Columbia River water was started at F-Reactor, to continue until reactor deactivation, to determine corrosion, effluent radioactivity, and filming effects. Coupon corrosion samples were installed in the rear spacer pattern of selected F-Reactor cooling channels; anticipated exposure periods are one and three and one-half months.

Silicate Inhibitor - Reactor Test. A 1/2 plant test of sodium silicate (20 ppm added SiO_2) was started at D-Reactor to further determine effluent radioactivity and corrosion effects with this additive to the cooling water. Coupon corrosion samples and corrosion probes were installed in various locations, including rear spacer pattern of selected cooling channels, effluent water sampling lines, and inlet systems. Anticipated exposure periods are one and four months.

3. Gas Atmosphere Studies

Graphite Burnout Monitoring. Graphite burnout monitors discharged from Channel 1880 at KW-Reactor showed a burnout profile with these peaks: (1) about 3% per 1000 operating days (%/KOD) at 60 inches into the stack; (2) 1.5 %/KOD at 95 inches; (3) 0.9 %/KOD at 155 inches; and (4) a dome-shaped peak of 0.5 %/KOD at the centerline of the stack.

Except for the first peak at 60 inches, this same general profile has appeared in the previous two tests at KW. This first peak is attributed to the reaction of oxygen and graphite. Due to the stack over-bore patterns, the maximum temperature appears in the vicinity of 90 inches, where the thermally-induced reactions with H_2O or CO_2 cause the second peak to appear.

The composition of the gas atmosphere is a mixture of He and N_2 with small amounts of H_2O . Analysis by mass spectrometry also showed 1% CO_2 , a product of graphite reaction with O_2 . The dome-shaped peak near the centerline is attributed to the radiation-induced reaction of C and CO_2 . (The flux profile of the reactor is not affected by the skewed temperatures and follows the usual cosine-like curve.)

At this time no explanations can be given for the peak located at 155 inches.

Since the early part of January 1965, F-Reactor has operated on a production test (IP-725) which permits a 50 C increase in graphite temperature above the 735 C limit.¹ On the basis of acceptable rates from monitors which were discharged February 3, 1965, it is expected that the temperature limit will be raised another 50 C in the near future.

Profiles of the burnout rates showed the absence of any O₂. The only peak appeared near the stack centerline where the temperature and the flux were at a maximum. The peak was 1%/KOD in Channel 1067 and 2%/KOD in Channel 3580. Monitors, which were protected from the gas atmosphere by a 3/8-inch-thick wall of graphite, oxidized at the same rate as exposed monitor, indicating a homogeneous reaction.

Monitors in Channel 3461 at B-Reactor had rates below 2%/KOD. There was an O₂ peak of 1.2%/KOD at 60 inches.

Measurement of the Effective Gamma Dose Rate in KE and KW Reactors by Methane Decomposition. The determination of the effective radiation dose rate in nuclear reactors has been continued. The dose rate was measured by comparing the rate of methane decomposition by reactor irradiation with the decomposition rate in Co-60 field of known flux.

In a radiation field, methane decomposes yielding hydrogen, ethane, and small quantities of other products. In a Co-60 gamma flux of 2.5×10^6 r/hr, based on ceric sulfate dosimetry, the 100-ev yield of hydrogen from methane in pyrex ampoules is 7.5.

Six methane samples were irradiated in the "quickie" facilities in KE or KW reactor. Five of these samples gave reasonably consistent decomposition rates. The average dose rate for these five experiments is $2.0 \pm 0.2 \times 10^6$ r/hr.

4. Process Tube Development

Fracture Studies. Examination of two Zircaloy-2 tubes discharged from KW-Reactor disclosed hydriding of the inside surface of the tubes from a point approximately coincident with downstream end of the fuel column to the extreme downstream end of the tube. Samples

¹A. Russell, "Production Test IP-725 Increased Graphite Temperature Limit - F-Reactor," RL-REA-14, December 1964.

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taken from several points upstream of the location described showed no increase in hydrogen content above amounts normally present. Downstream of that location, hydrogen content increased rapidly with distance to a level of about 1300 ppm in the vicinity of the downstream flange. The two tubes were virtually identical in quantity and distribution of hydride.

The hydrogen in the affected areas is present as a case of massive hydride at the inside surface varying in thickness up to about 0.005-inch depending upon the location of the sample relative to the end of the tube. On any given section of the tube the hydride case appears to be quite uniformly distributed around the circumference.

Programs are in progress to determine the mechanism by which the hydride is formed, the extent of hydriding in other tubes in the reactor, nondestructive methods for detecting hydrided tubes in situ, effects of the hydriding on mechanical properties of the tubes, and possible methods of recovering tubes partially affected by the problem without removal from reactor.

5. Thermal Hydraulic Studies

N-Reactor Boiling Burnout Studies. Twelve burnout points were obtained using an electrically heated model of the outer flow annulus in an N-Reactor fuel column with the fuel element eccentric toward the bottom of the process tube. Eleven of the burnouts were shown by abnormal temperature rises indicated by thermocouples used as burnout detectors. The twelfth point resulted in physical burnout of the model.

The model consisted of two horizontal tubes, an outer unheated tube having the same inside diameter as an N-Reactor process tube and an inner electrically heated tube having the same OD as the outer fuel tube in an N-Reactor fuel element. Cooling was provided by water flowing in the annulus between the two tubes. The construction of the model included a 2-ft unheated flow-calming length at the inlet end, followed by a 9-inch length of heated tube concentric in the process tube, and a 23-inch length of heated tube eccentric toward the bottom of the process tube. The degree of eccentricity approximated that which would result from shearing off the fuel self-supports except for the weld tabs. The over-all geometry of the model simulated that which would exist in an N-Reactor fuel column with the supports sheared from one fuel element.

The 12 burnout points were obtained at mass velocities of 0.5, 1, 2, 3, 4, and 5×10^6 lb/(hr)(sq ft), with inlet temperatures of 590 F

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(310 C) and 550 F (290 C)(about 10 and 50 F subcooling at the system pressure of 1500 psig). Outlet conditions ranged from about 30 F subcooling to 25% steam quality.

Burnout behavior during these experiments was flow dependent. At mass velocities up to 2×10^6 lb/(hr)(sq ft), burnout produced a sudden temperature increase of several hundred degrees. At higher mass velocities, unstable temperature rises of 50 to 150 F (25-85 C) occurred.

Burnout generally occurred within two inches of the downstream end of the heated section, at or near the bottom of the annulus. The two burnouts at a mass velocity of 0.5×10^6 lb/(hr)(sq ft), however, occurred at positions from 6 to 18 inches upstream from the downstream end.

A preliminary comparison with the results of earlier burnout experiments, using a similar model with the two tubes concentric, showed the burnout heat fluxes for the eccentric model to be as much as one-third lower for comparable mass velocities and bulk outlet enthalpies. Modifications are being made to a digital computer data handling program to allow a more accurate comparison.

A model simulating the geometry of the inner annulus in a column of proposed N-Reactor co-producer fuel elements has been assembled and installed in the Thermal Hydraulics Laboratory high pressure loop. This model, consisting of two concentric tubes with the outer tube heated electrically and the inner tube unheated, will be used to investigate burnout in the annulus between the fuel tube and target cylinder in the co-producer fuel.

N-Reactor Flooding Experiment. Plans are being made for laboratory experiments to determine the distribution of water in the graphite stack following an N-Reactor process tube rupture. The problem arises because of the possibility of a reactivity change produced by water collecting in the stack, with certain advanced fuel types being considered for the reactor. Calculation of the reactivity change requires a knowledge of the amount and distribution of the water. Construction of a model of the stack and development of a suitable method of measuring the water distribution following its introduction through a simulated broken process tube will be required for these experiments. Investigations of various methods of construction and measurement are in progress.

Predictions of Pressures in a K-Reactor Downcomer. The analysis was continued to determine the pressures in a K-Reactor downcomer during

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a transient temperature increase of the reactor coolant. Such an event could occur during a power surge and possibly cause damage due to overpressurization of the downcomer with boiling of the bulk coolant.

Four cases of temperature transients were considered at a total reactor flow rate of 210,000 gpm. Tentative conclusions were made from the results of analysis and are as follows:

1. Analytical predictions of pressure after boiling of the coolant starts in the downcomer can be made with only limited accuracy. The improvement of this calculation would cost considerable time and money and would involve the use of models and possibly tests on the reactor downcomer.
2. It was estimated that the pressure at the bottom of the downcomer approach section during a 10 C temperature transient above 97 C would be at least 9 psig. This compares to a strength limitation of 4 psig for the downcomer.
3. In general, the pressure in the downcomer will equal or exceed the saturation pressure corresponding to the temperature of the bulk coolant since there is no way for vapor to escape once boiling starts. As a result, the downcomer must be made strong enough to withstand the pressures anticipated for a particular coolant temperature or else the reactor must be operated at a level below that at which the coolant can reach saturation conditions.
4. The entrance elbow to the downcomer causes a significant restriction to flow by cavitation occurring at the entrance to the elbow. Redesign of the elbow would not eliminate pressure increases during temperature transients but would allow a significant increase in flow capability, if desired.

Hydraulic Tests for the Present Production Reactors. A comparison was made of the hydraulic characteristics of OIIIE I&E fuel elements centered in the process tube by arch bumpers, elliptical bumpers, and bridge bumpers. The arch bumper is being considered for use on OIIIE and OIIIN I&E fuel because it is easy to fabricate and attach to the fuel. The results, based on an analysis of previous data as well as recent tests, indicate that the pressure drop per arch

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bumper is about 28% less than that drop associated with the older bridge bumper. (Flow increase would be approximately 0.5 gpm per process tube.) Conversely, the flow resistance is approximately 17% greater than for the rounded elliptical bumper (flow reduction ± 0.4 gpm). It is estimated that the pressure differential over the length of the fuel column could be reduced to that of a corresponding elliptical bumpered charge by reducing the arch material stock thickness by 5 mils (15%).

Two types of toggle valves which are used in the Panellit pressure lines of K-Reactor were hydraulically compared. One assembly, which is presently in use, contains a needle-type stem while the other (new) assembly utilizes a double tapered stem in conjunction with a valve body which appears to be identical with the older valve.

The primary purpose of the test was to determine if the stems were interchangeable and what, if any, differences in hydraulic characteristics could be expected with various valve openings.

These valves are not used for regulating fluid flow, but rather for providing a means by which orifice or venturi pressure readings can be monitored with a Heise gauge or a Panellit gauge, either individually or simultaneously. In some cases, where the conditions permit, they may also be used for suppressing undesirable (or erroneous) pressure fluctuations originating in the reactor hydraulic system.

Two types of tests were conducted -- one in which the flow response to various valve openings was found and the other in which the relative effectiveness of suppressing induced pressure oscillations with various valve openings was determined. The tests were conducted with the original assemblies and with the stems interchanged. The results indicate that:

1. The stems can be interchanged between valve bodies and that the hydraulic characteristics of an assembly will depend only on the type stem used.
2. The double tapered stem is less sensitive to small variations in valve openings than the needle stem; i.e., a valve opening of $5/8$ turn for a double tapered stem is equivalent in flow resistance to an opening of $1/8$ turn for a needle stem.
3. A double tapered stem is only slightly more effective in suppressing pressure oscillations than the needle type. (A needle valve opening of $1/32$ turn is equivalent to an opening of $1/16$ turn for the tapered stem.)

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4. Some suppression of a 30-40 psi induced Panellit pressure fluctuation was evident up to approximately 1/4 turn for the tapered valve stem, whereas the needle stem showed some suppression only to about 1/8 turn open.

A hydraulic system in which step flow changes (transient pressure responses) can be accurately measured, is being fabricated. The system will be utilized for determining the several variables associated with pressure oscillations and the corresponding transient response of the reactor pressure monitoring system.

6. Graphite Studies

N-Reactor Irradiations. The irradiation program for N-Reactor graphite continues to progress satisfactorily in the GETR. The first of the fifth-generation capsules, H-4-5, was removed from the reactor after successfully completing 126 effective days (5 cycles) of reactor operation. The capsule was disassembled without incident, and all samples and flux monitors were recovered and are being returned. The final fifth-generation capsule, H-6-5, was installed.

As discussed under EGCR Graphite Irradiation revision of the neutron exposures for all samples in the previous capsules has been completed. For the N-Reactor H-4, -5, -6 capsules the change in flux varied from -14.7% to +17.7% with a mean value of +1.2%. The maximum neutron exposure on the long-term samples is now estimated to be 8×10^{21} nvt.

7. NRD Shielding Studies

The N-Reactor shield analysis report was completed. Information presented in the report includes:

1. Measured and calculated thermal, epithermal, and fast neutron flux distributions, and gamma ray and neutron dose distribution throughout the N-Reactor reflector and top shield.
2. Foil activation data and active detector response from which experimental results were calculated as related to position and experiment.
3. Calculated neutron and gamma ray spectra for selected shield locations.
4. Nuclear heating rates in the top thermal shield and at a selected location in the top shield.

5. Measured temperature distribution in the primary shields.

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

Thermocouple Fuel Element. A thermocoupled short core PRTR fuel element is being fabricated for irradiation testing at 1600 Kw in the PRTR rupture loop during April. To minimize flux peaking and high temperatures near the end caps, all rods in the 19-rod cluster are incrementally loaded; i.e., 1 cm UO₂ adjacent to the end caps, then 1 cm or 3 cm of UO₂-1 wt% PuO₂ at rod top and bottom, respectively, and then the main UO₂-2 wt% PuO₂ core. Four rods are thermocoupled -- two with two chromel-alumel thermocouples in each end cap and two with a W-5 Re/W-26 Re thermocouple on each end extending into the UO₂-1 wt% PuO₂ zone. These measurements will be used to refine the computer program for calculating core and end-cap temperatures and to aid in interpreting photomicrographs of irradiated fuel structure.

A new system for leading the thermocouples from the reactor has been developed. Double sealing at the nozzle cap will be provided -- the primary seal being a unique, compact, compression ferrule design followed by a brazed secondary seal.

Recycle Fuel Elements - Salt Cycle. Nine additional fuel rod segments were fabricated, bringing the total number of segments fabricated, using salt cycle fuel material, to nineteen. The PRTR cask unloading facility was inoperable due to construction; therefore, the nine segments were transferred into the PRTR basin using special handling techniques and the segments were assembled into three full length 60-inch rods.

The three rods that had been previously fabricated and assembled were removed from the element underwater, and the new rods, containing improved quality fuel, were placed in the element. The element was rebanded and later charged into the PRTR rupture loop where it is presently being irradiated. The nine rods contain approximately 2 wt% PuO₂ compacted to 82 to 83% TD.

A salt cycle recycle fuel segment was successfully irradiated in the MTR. The rod segment, containing UO_2 -2 wt% PuO_2 and irradiated to about 2.9×10^{19} fissions/ cm^3 , will be returned for postirradiation examination after a short cooling period. The UO_2 - PuO_2 in this test was enriched and codeposited by the salt cycle process from PRTR fuel previously irradiated to approximately 1.4×10^{20} fissions/ cm^3 and is similar to fuel in rods now undergoing irradiation in the PRTR rupture loop which will be scheduled into the reactor core in the next cycle.

Molten Core Fuel Element Tests. A test element to study the behavior of molten core UO_2 fuel elements was shipped to the MTR for irradiation. The four-rod cluster, GEH-4-116, contains 41.0 cm fuel columns with a transverse hole 6.45 cm from the bottom of each column; the size of the holes range from 0.074 cm to 0.259 cm diameter, simulating four different cases of fuel cracking which might occur during the reactor cycle. The expected power generation rate is 1 Kw/cm with a maximum surface heat flux at the cladding outer surface of 218 w/ cm^2 ; maximum fuel temperature is 3200 C with 60% of the fuel radius molten on startup.

Test Element GEH-4-111 successfully operated for 23.7 effective days in the MTR. The element was designed to allow molten UO_2 to contact the cladding. The initial melt radius on startup was approximately 60% and the surface heat flux of the Zr-2 cladding was 198 w/ cm^2 . The 2.74 cm diameter by 27.9 cm long fuel element contained 1.6 wt% enriched UO_2 pellets, one of which had a 0.254 cm diameter transverse hole. The element is currently undergoing radiometallurgical examination.

Diffusion Test Element. The Zr-2 clad diffusion test element was successfully irradiated for one cycle in the MTR GEH-4 loop. Each fuel rod of the four-rod cluster contained 16 inches of sintered pellet fuel with 1/2 inch UO_2 -2 wt% PuO_2 pellets alternated between one inch increments of natural UO_2 pellets. The element was discharged from the reactor and will be returned for radiometallurgical examination.

Fuel Element Rejuvenation. Two special fuel rods were irradiated one cycle in the MTR, then returned to Radiometallurgy for rejuvenation (re-enrichment after partial burnup). One end of the rod was opened, and a 0.040-inch diameter x 8.9-inch long Pu-15 wt% Zr wire was inserted into the 0.060-inch center hole in the pellet loaded fuel rods. Then the rod end was welded shut. Only one of the two rods was successfully rejuvenated. The hole in the other rod was blocked, and the Pu-Zr wire could not be inserted in the element. The rod that was successfully rejuvenated has been reassembled with a new cold rod into a four-rod cluster ready for further irradiation in MTR.

Wire Enrichment of PRTR Elements. Fuel elements containing sintered UO_2 pellets enriched with 85% Pu-Zr wire have been prepared for irradiation in the PRTR.

Fuel Fabrication Process. The analysis of the over-all fuel fabrication process for particle packed fuel elements is continuing. A schematic flow sheet of the process has been prepared and the unit operations divided into five major units: (1) powder preparation, (2) pneumatic impaction, (3) particle preparation, (4) vibrational compaction, and (5) quality appraisal. The analysis of the process has shown that there are 38 dependent and independent variables to be considered. This complexity has made it necessary to selectively test by and between unit operations.

The initial design model to experimentally investigate factors influencing vibrational compaction calls for 32 treatment combinations. This test will distinguish the interaction at two levels between: (1) particle size, (2) particle shape, (3) O/U ratio, (4) acceleration, and (5) frequency. This initial test will then be followed by subsequent replications to distinguish the dominant process variables for control.

Fuel Cycle Analysis. The Fuel Element Fabrication Code is being modified to accept the flow sheet developed for a one-ton per day mixed oxide, vibrational compaction fuel process. A flow sheet has also been laid out for the manufacture of mixed oxide pellets. When the calculations have been completed, it will be possible to compare the vibrational compaction process with pellets using three different methods of enrichment -- U-235, low exposure Pu, and high exposure Pu.

PRTR High Power Density Core

Fuel Elements. Seven UO_2 -2 wt% PuO_2 HPD elements (including the salt cycle element) are currently being irradiated in the PRTR. The maximum burnup on one of the elements is approximately 2814 Mwd/ton₁ which has been accumulated at tube powers as high as 1350 Kw. Maximum fuel temperatures approximate 2600 C at the mid-plane of the elements, and localized fuel melting probably occurs in the bottom end cap region of some rods.

Postirradiation examination of one rod from HPD Fe-6000 (maximum tube power 1350 Kw) revealed a central void over essentially its full length, i.e., to within 0.25-inch of the bottom and terminating to a point approximately eight inches from the top end cap. The other three rods examined from FE-6000 have been sampled for internal gas and diametrically measured. The average operating temperature of each

rod is reflected in the gas volume collected, i.e., the least gas was collected from the center rod in the element and the most from the outer rod. Diameter measurements along the length of the three rods indicate that no dimensional changes occurred during irradiation.

Design Analysis. "Status Report No. 1 - PRTR High Power Density Core" was completed and is being issued at month-end. This is a progress review of the activities of all the participants in the PRTR High Power Density Core transition program.

The third draft of the PERT Chart for the PRTR Batch Core Program was issued as "Comment Issue No. 2," dated 3/15/65. This chart is being included in the status report.

Dose rate calculations were made for the boron removing ion exchange units to be used in the Phase 1 chemical shim test at the PRTR, based on experimental data from small columns through which moderator water was passed.

Boron requirements for the eight-month "batch cycle" operating run were calculated. For an assumed operating period involving a scheduled three-day shutdown every month but no scrams during the course of the run, a total addition requirement of 2.5 kg of natural boron was calculated, including that required for initial borating of the moderator. During the run a total of 2.1 kg boron, containing 230 gm B^{10} , will be removed from the moderator by ion exchange or equivalent means. Most of the addition-removal requirements are due to reactivity adjustments at startup.

A total of 212 gm B^{10} will be consumed in $B^{10} (n, \alpha) Li^7$ reaction during the run, forming 149 gm Li^7 . The use of IX-3 or an equivalent cation exchanger is required to keep lithium concentration, and pH of the moderator, within acceptable values.

The assumed operating pattern for the run consists of eight 27-day periods of operation (interspersed with 3-day shutdowns) and a final, 24-day ninth cycle. For the first seven cycles of operation and part way through the eighth cycle, the rate of B^{10} burnout will exceed the required decrease in B^{10} concentration to compensate for fuel burnup. Therefore, boron addition during operation will be required for the first seven cycles and the first portion of the eighth cycle. For the remainder of the run, boron removal during operation will be required.

Corrosion and Water Quality Studies

Film Dissolution Effect Studies. Studies continue to determine the anomalous effect of film dissolution on subsequent corrosion.

Autoclaved Zr-2 coupons having 20 mg/dm² oxides were vacuum annealed inside and outside a shield made from Zr-2 foil. The purposes of the experiment were (1) to investigate effects of gettering, and (2) to investigate effects of shielding from silicon contamination (from the quartz furnace tube) on post-anneal oxidation rates. Autoclaved coupons were annealed at two conditions: 700 C for 370 min and 800 C for 70 min. Either of these annealing procedures is more than sufficient to dissolve the surface oxide.

Post-anneal weight gains (in 400 C, 1500 psi steam) were accelerated 1.3 to 2.5 times, compared to etched specimens. There was essentially no difference in rate between samples suspended inside and outside the Zr-2 foil. All annealed coupons underwent a second transition 14 to 21 days after the anneal. Total weight gains were 150 to 218 mg/dm² 47 days after the anneal (54 days total).

The results are in agreement with previous results obtained in un-gettered vacuum anneals, discounting impurities as a source of accelerated post-anneal oxidation.

Decontamination After Failure of Plutonium Ceramic Fuel Elements. A solution capable of dissolving high-fired PuO₂ and UO₂·PuO₂ mixtures has been developed after an extended research program. The material, inhibited 85% H₃PO₄ at 156 C, dissolves these refractory oxides and is not excessively corrosive to Zircaloy and stainless steels. Previously, an extended literature search had revealed no satisfactory solution. In the laboratory ~200 tests had been made with many different reagents, but the H₃PO₄ is the most satisfactory of all tested. Tests are continuing to determine the corrosivity of the reagent to various materials of construction and to develop a modified reagent. However, if a fuel element does fail in the PRTR and it becomes necessary to remove the rupture debris from portions of the reactor, it is felt that this reagent may be used if suitable precautions are observed. Tests will also be conducted to determine how effectively this reagent dissolves other candidate core materials.

Ceramic Washout Tests. Testing has been completed on the fourth ceramic core washout test. Externally applied vibration caused the release of a considerable portion of the core material from the fuel pin. The element used was a 12-inch section of a longer rod irradiated in the PRTR to 224 Mwd/ton. It was fabricated by swage-pneumatic compaction and contained a UO₂-1% PuO₂ core. Prior to

testing in the IRP loop the element was predefected with a slit 6" long by 1/16 - 3/32" wide. During the nonvibration portion of the test the element was tested for two weeks at 300 C, 13 fps, and 1600 psi with temperature cycles to 270 C every three hours. Thermal cooling from 300 to 40 C was also performed three times. No washout of the core was observed after the start of the test when several grams were lost, probably due to loose material from the machining operation.

Testing was resumed at the same conditions except that external vibration was applied to the test section containing the fuel element. After four days with vibration, the element had lost 38 grams. An additional three days of testing resulted in 27.5 grams lost.

During testing of three other similar rods without vibration, the only one that released any activity was fabricated by the Vipac-Nupac process. That rod lost seven grams, primarily during three thermal cycles from 300 to 40 C.

PRTR Crevice Corrosion Tests. The second in a series of tests to study crevice corrosion using Zircaloy-2 clad cartridge heating units with surface crevices formed by spiral-wound wire wraps and a "cluster" band was terminated by heating unit failure after 60 days. The test was conducted in lithiated water at pH 10; temperature and pressure were 293 C and 1350 psig. Test assembly surface heat flux was 550,000 BTU/(hr)(ft²). The general appearance of the test assembly indicates that metallographic examination will reveal more extensive corrosion than that associated with the first test assembly (February 1965 Monthly Report); gray-white deposits and heating unit surface penetration were evident immediately adjacent to one of the areas where the "cluster" band contacted the heating unit, i.e., in the narrow portion of a heated crevice formed by the heating unit surface and the band. White corrosion product was found between the double wrap of the "cluster" band as well as on the surface of the band where it contacted the heating unit. Similar corrosion product has been observed on irradiated high power density fuel pins adjacent to bundle bands.

The February 1965 monthly report stated that microscopic examination of sections from the first test assembly revealed gray-white deposits, up to 0.005-inch thick, in crevices associated with the "cluster" band. No estimate of penetration (which can be different from oxide thickness) was given at that time because of the irregularity of the corroded surface. Measurements from photomicrographs now show that the heating unit surface penetration was at least 0.007 inch.

PRTR Boron Shim Development. The actuator and seat springs from a PRTR dump valve operating in a H_3BO_3 environment (10 ppm B, 60 C) were examined after six days of testing. There was no indication of corrosive attack on these components. Testing was resumed after the examination and total exposure to date is about 30 days.

A test of the boron shim control in the 314 Building Shim Test Loop was terminated after 23 days of operation at 60 C in borated water (10 ppm boron). There was no evidence of pitting corrosion in the aluminum shim well where contact was established with the shim rod. It was concluded that corrosion under Phase I PRTR conditions would not be severe. Examination of coupons and a shroud-tube bellows assembly exposed during this test did not reveal any severe corrosion except on carbon steel and Type 416 stainless steel coupons. These materials would have corroded rapidly even without H_3BO_3 present, however. It was concluded that there are no apparent problems that would warrant delaying the Phase 1 tests.

All components for the prototype for Phase 1 PRTR tests have been delivered to PRTR, and installation is about 60% complete. Only a small amount of additional outage work will be required since the majority of the remaining work can be performed during reactor operation. Functional performance tests will be conducted with H_2O before the prototype is connected to the moderator system. It should be possible to complete installation and the functional performance tests in time to begin testing in the moderator during the April outage.

Water Quality Studies for Phase 2 PRTR Short Core Test. Analyses of the Phase 2 boron shim installation for PRTR were made. It was concluded that:

1. Activity levels in the H_3BO_3 removal exchangers should be $<100 \text{ m}\mu\text{r/hr}$ at the unit surfaces except near the surface of the upstream bed where filtration of particulates could result in levels up to $300 \text{ m}\mu\text{r/hr}$.
2. Specifications for moderator quality controls and analyses should be changed from the present values. Recommendations for operating limits, analyses, and analytical frequency were prepared.
3. The existing moderator cleanup ion exchanger will adequately control the pH and lithium concentration during Phase 2 operation. The effect of proposed Phase 2 operations on useful lifetime of this exchanger will be small.

4. Boron elution from ion exchange units during dedeuterization will be very small. The boron concentration in the effluent D_2O will be controlled primarily by the boron concentration of the liquid in the bed when dedeuterization is started.
5. The Phase 1 H_3BO_3 prototype could be used for Phase 2 operations. However, the design is not optimum and resin replacement requirements would probably be significant.

A laboratory test was conducted to determine the activity pickup by the BIX exchangers during Phases 1 and 2 of the PRTR High Power Density Core operation. The laboratory data were applied to full-size BIX units. The resulting activities were less than 100 mr/hr at the surface of the unit except for the top of the column where a 300 to 350 mr/hr field might exist. The field at the top of the column can be reduced to an acceptable level by installing a lead (or other material) girdle. The specific activity of the coolant was reduced ~27% in passing through the column.

PRTR Pressure Tubes

Mechanical Testing. Four stress rupture tests and two crack propagation tests were performed.

Three of the four stress rupture tests indicated that the time to rupture increases and then decreases with increasing fast neutron exposure. This is in accord with two stress-rupture tests previously reported. All stress rupture testing has been done at 280 C. Two hoop stress levels have been used -- 66,000 psi and 64,000 psi.

One crack propagation test was performed at 100 C with a specimen whose average fast neutron exposure was 1.5×10^{20} nvt. The other test utilized a specimen whose average fast neutron exposure was 1.5×10^{21} nvt. It was performed at room temperature. These tests indicated that the stress necessary for propagation of a given size crack is increased by temperature and by fast neutron exposure. This is the first indication of a positive effect of fast neutron exposure on crack propagation. Earlier data indicated no significant difference between specimens having exposures of zero and 1×10^{20} nvt to 5×10^{20} nvt.

Four specimens of the Zr-2.5% Nb tubing have been burst tested. One room temperature and one 300 C temperature test was made with both the 30% and the 60% cold worked tubing. The results of these tests are summarized as follows: (1) the 60% CW material was only a few percent stronger than the 30% CW material; (2) the ultimate principal

stress was 115,000 psi at room temperature and 85,000 psi at 290 C; and (3) circumferential elongation was 1% for all specimens.

In these tests the ratio of hoop to axial stress was two to one. In each test the test specimen failed in a shear mode. In no case did the test specimen fail end to end as is frequently the case with highly cold-worked Zr-2 when tested at room temperature.

Work is under way to conduct stress rupture tests under biaxial stress conditions in which the hoop strain may be continuously measured without test interruption. The current approach is to employ two venturi flow meters, one as a flow measuring standard, and the second, an indirect measure of changes in OD of the test specimen. A test section for the 350 C flow loop in the 314 Building has been designed to provide the same approximate geometry, flow, and temperature condition existing in a PRTR process channel. Construction of the flow loop test section has begun.

Reactor Components Development

Fretting Corrosion Investigation. Final data have been received from the test to determine the increase of zirconium content of primary coolant following a rapid change in the steam generator pressure. A gradual increase in the HX-1 pressure from 335 psig to 365 psig is known to cause an increase in amplitude of inlet jumper vibration (greater than 20% on a monitored tube), and a simultaneous increase in the tube Zr content. This test was performed as a part of PRTR Test 101 to determine how dependent the zirconium content is on prior conditions. Such information would indicate whether or not the zirconium content follows the change in operating conditions closely or lags for some undetermined length of time. This would permit an assessment of the value of a zirconium monitor for detection of fretting.

The relative increase of vibration was not obtained in this test due to a malfunction of the amplifiers of the monitoring system; the test will be re-run. The analytical results, as usual, show some rather wide unexplained variances; they do indicate, however, that the zirconium content is affected rather rapidly (within about 25 minutes) after a change in reactor operating conditions.

Second Generation Shim Rod. No significant change in the operational characteristics of the second generation shim rod has been observed. The "A" rod functions normally, but the "B" rod continues to exhibit intermittent sticking on the up drive. Modification of the alternate

second generation driving head is under way at the PRTR. The modifications include drilled and welded heat sinks, conventional limit switches and actuators, and potentiometer readout of position.

Chemical Shim. Testing of PRTR moderator components was resumed in the environmental test facility. Conditions of the test are 50 ppm boron, 60 C, seven gpm average flow, and two gph through the shim well. The dump valve test continued at 60 C and 10 ppm boron.

Shroud Tube Removal. Development of PRTR calandria shroud tube replacement equipment is continuing. The first phase of development includes shroud removal equipment and tube-sheet plugging techniques. A second phase will include shroud installation procedures and equipment. Preliminary scope of a cutter to slit a shroud tube into a helical ribbon, prior to removal, has been started. Tests have established that a milling cutter will split, in one pass, a 6063 aluminum shroud tube (as-received or annealed condition).

Design Analysis

Plutonium Utilization. Burnup calculations for the investigation of plutonium burning in large thermal power reactors were initiated. Yankee core I has been assumed as a base case, and a "calibration" calculation was completed in which an attempt was made to predict the measured burnup in this core with uranium enrichment. Results obtained using the multigroup, multiregion code ZODIAC 2 were found to underpredict the reactivity loss with burnup by as much as 50%, although excellent agreement was obtained for the excess reactivity of the hot, clean core. The source of this discrepancy is being investigated before attempting to substitute plutonium enrichment in the calculation.

2. Plutonium Ceramics Research

Phase Studies. Buoyancy and convection corrections of the tensi-metric apparatus calibration were completed using ThO₂ and UO₂ specimens. The thermocouple correction was rechecked and found to be in agreement with the curve previously obtained. O/U ratios were determined under several sets of conditions selected to yield single and two-phase products. These ratios were within 0.01 units of published values. However, equilibrium was not attained in these experiments. The rate of approach to equilibrium was extremely slow, on the order of 0.002 (O/U)/hr at 1130 C.

AmO₂ was reduced in hydrogen in the thermobalance. A compound of constant composition was not formed. The O/Am ratio after one hour at 750 C reached 1.473 and was still slowly decreasing, implying

formation of AmO or Am metal. On cooling to room temperature, however, the O/Am ratio had increased to 1.525 and was continuing to increase at the rate of 0.002 (O/Am)/hr. X-ray diffraction of AmO₂ heated in oxygen showed a single fcc phase with $A_0 = 5.383 \pm 0.001 \text{ \AA}$. Heating a portion of the oxide to 900 C in hydrogen produced a rust colored material which comprised two phases; a hexagonal La₂O₃ type and a cubic phase. This suggests a two-phase region near the sesquioxide composition (similar to $\alpha + \beta \text{ Pu}_2\text{O}_3$) although the possibility of an incomplete structural transformation cannot be ruled out.

High Temperature Chemical Compatibility Studies. Chemical compatibility between ceramic fuel materials and metals potentially useful as cermet matrices or cladding is an important factor in nuclear fuel design. A rigorous compatibility test was conducted by arc melting PuO₂ with niobium in argon. Metallographic examination showed compatibility between PuO₂ and niobium in the liquid state. Perfect spheres of each of the materials, PuO₂ and niobium, were randomly dispersed in the other.

Stainless Steel-PuO₂ Cermet Extrusions. Examination of one of the stainless steel-20% PuO₂ cermet pins extruded last month shows a dense, continuous matrix with very little damage to the PuO₂ particles. Estimated extruded core density is 93% of theoretical. Only a slight amount of stringering of the PuO₂ particles was observed. The PuO₂ particle size used for this extrusion was -100 +150 mesh.

Four stainless steel-20% UO₂ extrusions were made to determine the effect of billet density on the extruded core density. One of the billets contained a cermet pellet pneumatically impacted to 97.5% of theoretical density. The other three billets contained cold-pressed, low density cermets.

Data from these extrusions are summarized in the following table:

BILLET DENSITY EFFECT ON EXTRUDED CORE DENSITY

	High Density Billet, P-1	Low Density Billets		
		PP-1	PB-1A	PB-1B
Billet Composition	← Stainless Steel - 20% UO ₂ →			
UO ₂ Particle Size, Mesh	-60 +200	← -60 +150 →		
Billet Density, % TD	97.5	82	76	76
Extruded Core Density, % TD	95	94	96	96
Pin Dia., inches (Avg)	0.191	0.174*	0.199	0.196
Clad Thickness, inches	0.009±0.001	0.012±0.003	0.013±0.002	0.012±0.003

*Smaller diameter due to "washout" in the extrusion die.

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Polished metallographic samples from the four extrusions show an excellent physical (and possibly metallurgical) bond between core and clad. One-half of the length of the fuel pin extruded from the high density billet and of a fuel pin from a low density billet (PP-1) was given a light finishing swage. In both cases swaging damaged the ceramic phase as evidenced by cracking and severe pull-out of the UO_2 in the polished specimens.

Density measurements and metallographic observations indicate that cermet pins extruded from low density billets are as good as pins extruded from high density billets. Low density billets have the advantage of not requiring a pneumatic impaction step in their fabrication.

High Density and Low Density Stainless Steel- PuO_2 Cermets. High and low density stainless steel-20% PuO_2 cermets being irradiated in the MTR have attained ~1.6% burnup of the plutonium. Two capsules, one containing high density and the other low density cermets, were discharged last month after about 2% burnup of the plutonium. Three capsules have been returned for postirradiation examination.

Low density (82% of theoretical) stainless steel-20% PuO_2 cermets heated to 900 C under vacuum for 1000 hours showed only a very slight decrease (<1.0%) in density. High density cermets tested at the same conditions for 409 hours decreased 2.18% in density. Apparently the porosity of the low density cermets accommodates any swelling which occurs.

UO_2 - PuO_2 Fast Fuel Development. UO_2 -20% PuO_2 physically mixed and impacted fuel material is being prepared for irradiation evaluation in EBR-II. Some of the material will be irradiated as a joint BNW-ANL program. Approximately 8 Kg of high density mixed oxide are being prepared by ball-milling the powders to obtain a homogeneous mixture of small particles followed by pneumatic impaction to obtain a high density feed material suitable for vibrational compaction.

The main problems encountered are stoichiometry control and excessive gas content of the high surface area powders. Various techniques, such as high temperature vacuum outgassing, are being employed to reduce the adsorbed gas content and at the same time control stoichiometry.

PuO_2 Microspheres. Impacted PuO_2 feed material for the production of microspheres has been fully characterized and shipped to Mound Laboratory. Mound will generate microspheres in their hooded plasma-torch. These will be evaluated for possible use in cermets by

measuring crush strength, fission gas retention, and radiation stability. The density of the particles is 95% TD as measured by mercury displacement. Gas content (1000 C) is 0.38 cc/g. Oxidation tests ($O/Pu = 1.997$), metallography, and x-ray analysis all indicate single phase material. Spectrographic analysis shows impurities to be at a satisfactory low level.

High Temperature DTA. The differential thermal analysis apparatus for use up to 1700 C was installed in a plutonium glovebox.

Plutonium Ceramics Metallography Facilities. The atmosphere supply for the plutonium metallographic research hood was changed from dry air to nitrogen to allow examination of boron-stainless steel impacted specimens now in progress. The atmosphere change required only a change in the main line source to the hood and an adjustment of the flow in each of the hood chambers. The interlock between the hood and metallograph is being altered to allow nitrogen purging.

3. Ceramic (Uranium) Fuel Research

Substoichiometric UO_2 Phase Equilibria. The relative partial molar enthalpy of uranium in UO_{2-x} between 1800 and 2400 C was estimated to be 20 kcal/mole. This relatively high value suggests formation of a UO_{2-x} compound rather than solubility of U in UO_2 .

UO_2 Melting Studies. UO_2 pellets containing small tungsten spheres (-100 +200 mesh) were irradiated with the major axis horizontal at a heat generation rate sufficient to cause melting to approximately one-half the fuel radius. The capsule was then rotated 180°, irradiated another five minutes, and withdrawn from the reactor. Destructive postirradiation examination showed that tungsten sank to the "new" bottom of the molten zone when the capsule was rotated. Although the results of this test are somewhat inconclusive, they indicate little change in the melt radius during two hours. The tungsten particles sintered into a rod early in the test and creep of the solid phase UO_2 near the liquid interface, and perturbation of the isotherm near the rod are apparent.

Detailed examination of this and a similar, previously irradiated capsule suggested that the molten UO_2 was significantly more viscous than previously observed in laboratory tests of molten UO_2 in tungsten crucibles, or of explosively melted UO_2 . Stoichiometry of various portions of the irradiated specimen will be determined.

Melting Points of Irradiated UO₂. Melting points of irradiated UO₂ specimens were remeasured using portions of UO₂ specimens used in similar measurements approximately four years ago. The specimens had been stored in air, in a high intensity gamma field. The measurements do not show the previously reported increase in melting point at low exposures but have relatively little change with increased neutron exposure to 4.1×10^{19} fissions/cm³. Changes in the UO₂ structure during the four-year storage are being investigated.

UN-20% PuN Irradiation. The examination of two irradiation capsules containing UN-20% PuN clad in 304L stainless steel is continuing. Determination of the nature of an apparent reaction zone between the clad and the fuel is being emphasized. Microhardness measurements across the cladding cross section, backed up by microchemical analyses, indicate that the reaction zone in capsule GEH-14-616 is not a nitride, oxide, or carbide.

Further studies of the cladding reaction zone will include: collection of microdrilled samples of the material for chemical analyses, autoradiography of the clad without the fuel, electron microscopy of the cladding, electron microbeam analysis across the cladding, and x-ray diffraction analysis of the reaction zone.

The third (final) capsule of this series (UN-20 wt% Pu/U) was discharged from the MTR during the month.

Pneumatically Impacted Cermet Pins. Pneumatic impaction was used to produce integrally-clad and capped, high density, stainless steel-UO₂ cermet fuel pins. Such pins, when produced to sufficiently close tolerance, could be welded end for end to produce any desired length fuel pins. An impaction mandrel designed to produce 19 pins per impaction was used to make the first set of pins. These pins are about 3-1/2 inches long and 0.295 ± 0.010 inch in diameter. The cermet density after impaction is >99% TD; prior to impaction the cermet density was 76% TD. Most of the pins were slightly bowed and flared at the ends; however, as a first attempt the results are encouraging. Metallographic examination of these pins is in progress.

Electron Microscopy. A 200 l/s ion pump was installed on Column 1 in an effort to reduce the ultimate pressure obtainable in the column to less than 1×10^{-6} Torr. Pumpdown time (to an operating pressure of 1×10^{-5} Torr) is somewhat longer with the ion pump because the column must be evacuated to about 5 microns with the mechanical pumps before valving in the ion pump. However, below this pressure diffusion pump speed falls off so that the ion pump is faster.

Preparations are under way to accurately determine the pressure in the sample chamber. Present measurements indicate this pressure is less than 5×10^{-6} Torr. There still remain about 20 neoprene o-rings and gaskets in the column which will be replaced with Viton-A seals, to provide a noticeable improvement in the obtainable vacuum. Addition of a molecular sieve trap to the rough pump has cut the roughing time in half.

4. Basic Swelling Studies

Irradiation Program. A general swelling capsule containing eight specimens of 2.88 at% enriched, high purity uranium in several heat treated conditions operated successfully in a reactor at a controlled temperature of 375 C. It was discharged after reaching goal exposure and is being held for postirradiation examination to obtain better insight into the role of higher burnup and burnup rate on swelling mechanisms.

The third capsule (P-4) in a series of controlled pressure-temperature capsules was charged in a reactor and is operating successfully at 1000 psi and 625 C. This capsule contains ten, naturally enriched specimens of high purity uranium, U + Fe-Al and U + Fe-Si with several metallurgical histories. Additionally, two controlled pressure-temperature swelling capsules which are scheduled to operate in a reactor at 500 psi and 450 C and 575 C, respectively, were bench-tested and final assembly is being made.

Postirradiation Examination. The second controlled pressure-temperature capsule (P-3), which operated at 1000 psi and 575 C to about 0.13 at% B.U., was opened in Radiometallurgy. The capsule contained six high purity uranium specimens and two U + Fe-Si specimens which had undergone several different heat treatments prior to irradiation. Postirradiation photomicrography and preliminary density measurements of each specimen are complete. The first pressurized capsule (P-2), which operated at 1000 psi and 450 C to 0.13 at% B.U., demonstrated the effectiveness of pressure in restraining the swelling of high purity and alloy specimens, and, similarly, preliminary results from the second pressurized capsule show that pressure inhibits swelling at the higher irradiation temperature, also. Optical and electron metallography is in progress to compare data from pressurized capsules with results from previous general swelling capsules.

5. Irradiation Damage to Reactor Metals

Alloy Selection

Several nickel-base alloys are being studied to determine the effects of irradiation and environment upon their mechanical properties. Specimens of Inconel 600, Inconel 625, Inconel 702, Inconel 718, Incoloy 800, Hastelloy N, and Hastelloy R-235 have been irradiated at 50 C, 280 C, 580 C, and 740 C. These specimens are currently being tested in tension at room temperature, 300 C, and 650 C. Data obtained this month substantiate previous data showing a marked decrease in ductility at the 650 C test temperature as compared to the room temperature properties.

Present and future fast-breeder reactor concepts utilize a liquid metal coolant. Components for a bench model of a liquid metal irradiation capsule to be used to evaluate the effects of irradiation and environment upon the mechanical properties of nickel-base and refractory metal alloys have been fabricated and are being assembled. Assembly of safety control circuits and a temperature control system has also been initiated.

In-Reactor Measurements of Mechanical Properties

The 650 C - 10,000 psi in-reactor creep test on annealed 304 stainless steel has accumulated 900 hours of test time and exhibits a rate of 4.4×10^{-6} /hr. The ex-reactor test at these conditions has a rate of 2.2×10^{-6} /hr. A new in-reactor test at 525 C - 30,000 psi on annealed 304 SS was started during this month. Neither the in-reactor nor the ex-reactor test has accumulated significant test time to determine creep rates.

Irradiation Effects in Structural Materials

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

Tensile tests have been conducted on BONUS transition joint specimens which were irradiated in the G-7 hot water loop to an exposure of approximately 2.0×10^{20} nvt (>1 Mev). These tests were performed at 290 C and at room temperature on specimens from both an explosively formed joint and a tandem extruded joint. All 12 tests resulted in a low ductility failure outside of the transition joint in the

Zircaloy-2 end of the specimen. All specimens previously tested in either the as-fabricated condition or after an exposure in the out-of-reactor loop also failed in the Zr-2 end of the specimen.

Metallographic examination of the as-fabricated materials revealed a fine precipitate equally distributed throughout the grain matrix and the grain boundary in the Inconel 600. This precipitant probably accounts for the low ductility exhibited by the Inconel. Additional metallographic studies are in progress on the irradiated specimens.

Irradiation Effects in Nickel-Base Alloys

The purpose of this program is to determine the effects of modified microstructures on the irradiation behavior of nickel-base alloys. Structural modifications made by preirradiation thermal and/or mechanical treatments are screened by tensile and stress-rupture tests and by metallographic studies of both control and irradiated material.

Irradiation of 24 Hastelloy X-280 specimens at 280 C to a goal exposure of approximately 2×10^{20} nvt is continuing during Cycle 69 of the ETR. These specimens have been given a variety of pre-irradiation treatments to increase the dispersion of matrix precipitation in order to reduce formation of embrittling grain boundary deposits during irradiation. Three more quadrants (18 specimens) have been sent to the ETR for irradiation under the above conditions commencing with Cycle 70. These contain Hastelloy X-280 specimens in the solution treated condition and Inconel X-750 specimens given five heat treatments which cover a range of aging conditions.

A metallographic precharacterization technique was used to allow positive identification of microcracks in ruptured specimens by electron microscopy. After a metallographic polish and chemical etch, a suitable area near the fracture surface of a specimen is photographed at 750 X. The specimen is then replicated and the same area, located optically on the replica, is again photographed at 750 X. The replica is then shadowed and electron micrographs are taken of the area at 3000 X. The origin and progress of microcracks are established by their relationship to shadows cast by precipitates in the grain boundaries and to other prominent structural features.

Development of a compact, high-temperature, stress-rupture apparatus with increased testing capability over available facilities has been started. The design is based on a recently-developed creep-rupture unit with adaptations for remote handling and automatic clocking of time-to-rupture.

Damage Mechanisms

The objective of this program is to determine how interactions between irradiation-induced defects and dislocations modify plastic deformation mechanisms in a metal. The role of interstitial impurities in alpha iron is currently being investigated.

Comparisons between experimental data and an activation energy-effective stress equation derived from the experimental temperature dependence of the effective stress are still in progress. The accuracy of the predictions of this new method continues to be better than that of the traditional method derived from an empirical dislocation velocity-stress equation. Latest achievements are prediction of the temperature dependence of the effective stress at a strain rate of 0.02 min^{-1} from data measured at 0.002 min^{-1} and calculations of effective stress from activation volume measurements which agree with those calculated from temperature change experiments.

Bulk samples of iron irradiated to 5×10^{18} nvt ($E > 1 \text{ Mev}$) were successfully sectioned, thinned and examined in the electron microscope. No spots similar to those observed in Mo at equivalent exposures were seen in the iron examined.

ATR Gas Loop Studies

Model Loop Studies. Using the newly installed water jacket, another series of conductivity tests was run on the foil insulation. The conductivity values were dependent on flow, similar to the previous run, but to a much lower degree. A conductivity of $0.475 \text{ BTU/hr-ft-F}$ was measured at a mean insulation temperature of 880 F (470 C) and a flow of 440 lb/hr . The temperature drop across the insulation was 1220 F (680 C). The previous run had shown a conductivity of $0.615 \text{ BTU/hr-ft-F}$ at the same flow and mean temperature and a temperature drop across the insulation of only 185 F (100 C). The decreased effect of flow with the lower skin temperatures, caused by the cooling water jacket, indicated that the heat leakage flow may have been through the annulus between the outside of the foil and the inside of the test section. Visual inspection indicated a definite flow pattern on the exterior of the foil insulation. The major portion of the leakage flow originated at the joint with a $1/16$ -inch gap, but some flow did come from the prototypical ATR joint. The joint with the gap was repaired by machining the foil to give the overlapping joint, and a flow dam was installed on the top section of the joint. The insulation has been installed, and another set of conductivity tests has been started.

Helium Purification System. The high pressure helium purification system is inoperative because of maintenance required on the compressor. Replacement parts are now on hand and the system will be working soon.

A small bypass purification system on the gas loop is working at loop pressure and utilizing pressure drop available from the gas bearing compressor. A catalytic copper oxide bed, part of the high pressure purification system, was connected to the bypass system to oxidize hydrogen. This is followed by activated charcoal and 13X molecular sieve at the temperature of liquid nitrogen.

During a typical run, the only loop gas impurity in an amount greater than 0.1 ppm was water vapor at 3.5 ppm. No special effort was made to achieve minimum levels of impurities during the runs.

An experimental low temperature adsorption bed for removal of neon is being designed. This would trap neon on charcoal at -340 F, achieved by maintaining a vacuum of six to eight inches of mercury on the liquid nitrogen dewar holding the adsorbent bed.

Model Loop Heater Design. Components of the second generation helium gas heater for the model gas loop are now being fabricated. The first stage of the new heater will use pure nickel (nickel 200) internal electrodes welded to the Nichrome (80 Ni-20 Cr) heating tubes. As no information was readily available on the welding of nickel to Nichrome, welding tests were made. Strong, ductile, TIG welds could be made using a filler rod of Inconel 82.

Exterior Insulation for the ATR Gas Loop Hot Piping. Testing of thermal conductivity has begun on Hastelloy-X felted wire being considered by the Architect Engineer for insulation on 1600 F (870 C) ATR Gas Loop piping. Precautions are being taken to guard against thermal losses and undesirable temperature gradients, while an independent check is being made on heat flux through the test piece.

ATR Loop Corrosion Studies. The inhibition of evaporation rates of superalloys by inert or reducing gas overpressures is being studied. Preliminary tests on Haynes Alloy 25 indicate that an established evaporation rate (temperature of 1120 C and pressure of 10^{-4} torr) can be effectively decreased by the addition of hydrogen. Hydrogen was chosen for initial evaluation because it reacts readily with any oxygen present. Oxygen contamination, because of its film forming proclivities, is a hazard to satisfactory determinations of the effect of gas pressure on suppression of metal composition. Hydrogen is also attractive because it is readily available in pure form. For

one test, the rate of evaporation was decreased by a factor of two upon the addition of 10 torr hydrogen. Another test gave a two and one-half fold decrease with the addition of 86 torr hydrogen. These tests were not completely reproducible. Additional tests are planned to verify the effect of inert gaseous overpressures on evaporation.

Oxidation of pure niobium in low oxygen partial pressures is being continued. The reaction tube has been modified slightly so that pressure measurements near the sample can be made. The following rates were observed from a 900 C test using the modified tube: 2.48×10^{-4} mg/cm² sec (0 to 130 min) and 1.80×10^{-4} mg/cm² sec (130 to 1400 min). The pressure maintained at the leak source was 12 μ oxygen. During the first 100 minutes of the test, the pressure measured at a point 100 cm removed from the sample was 7 μ , the pressure 10 cm from the sample was 1 μ . After 130 minutes, the pressure 100 cm from the sample rose to and remained constant at 10 to 11 μ ; the pressure near the sample rose to and remained essentially constant at 7 μ . Apparently, gas phase diffusion control obscures initial kinetics and the transition to the secondary rate. The rate after 130 minutes was similar to values presented in the literature at comparable temperatures and pressures.

Dynamic Materials Testing Apparatus. A "cleanup" bed consisting of zirconium turnings has been installed, and a preliminary test made. It was found that the heater capacity was insufficient to accommodate the full loop flow since the temperature would only go to about 280 C. On bypassing some of the helium, the heater capacity is sufficient to raise the temperature to 550 C where the zirconium is effective in removing oxygen. A test to measure the effectiveness of the "cleanup" bed and to measure the in-leakage of a cold loop is awaiting reassembly of the chromatograph.

The second run of 266 hours was completed on March 15, 1965, using a niobium sample. The sample showed considerable scaling, and the scaling was also noted by the increase of ΔP across the test section during the run. Oxygen concentration was below 0.5 ppm during the run, while hydrogen and carbon monoxide reached a maximum of 60 and 15, respectively. Residual oil vapor from a past diaphragm rupture was found in the check valves and undoubtedly contributes to hydrogen and carbon monoxide buildup.

6. Nuclear Graphite Studies

Radiolysis of Mixtures of Carbon Dioxide and Hydrogen. The effect of Co60 gamma radiation on mixtures of carbon monoxide and water vapor had previously been reported (HW-SA-3440). Now, the radiolysis of

carbon dioxide-hydrogen mixtures is being studied. The two gases are purified and then mixed in pyrex ampoules with break seals. After irradiation, the resulting gas mixture, analyzed by gas chromatography, contains carbon monoxide and methane. Water vapor is undoubtedly formed also, but equipment is not available to measure the small amounts of water vapor which are formed. The dose rate is determined by the decomposition of methane using $G(H_2) = 5.7$ molecules/100 ev.

At a temperature of 350 C, the yields for the CO_2-H_2 reaction are 3.0 molecules/100 ev for CO formation and 0.1 for CH_4 . The calculated yield for water formation is 3.2 molecules/100 ev.

Carbon Dioxide-Graphite Reaction. The determination of rate constants in the $C + CO_2$ reaction for TSX graphite have been completed. The final expression is

$$R = \frac{k_1 P_{CO_2}}{1 + k_2 P_{CO} + k_3 P_{CO_2}}$$

where

$$\begin{aligned} k_1 &= 8.33 \times 10^6 e^{-66.8 \times 10^3/RT} \text{ (hr}^{-1} \text{ mm}^{-1}) \\ k_2 &= 4.15 \times 10^{-7} e^{29.5 \times 10^3/RT} \text{ (mm}^{-1}) \\ k_3 &= 3.50 \times 10^{-3} \text{ (mm}^{-1}) \end{aligned}$$

Effect of Hydrogen on the Graphite-Water Vapor Reaction. The usual rate expression for the water-graphite reaction is:

$$R = \frac{k_1 P_{H_2O}}{1 + k_2 P_{H_2} + k_3 P_{H_2O}}$$

In the case of TSX graphite this equation is obeyed if the hydrogen level is above about 1000 ppm. At $P_{H_2} < 100$ ppm the rate is given by

$$R \approx k_1 P_{H_2O}^{1/2}$$

Studies in the intermediate range $100 \leq P_{H_2} \leq 1000$ ppm show a smooth transition. Possible mechanisms to explain these effects are being investigated.

In-Reactor Strain to Fracture. Preliminary results indicate that the in-reactor breaking strain in graphite is rate sensitive. Low-strain-rate samples developed higher strains before failure. No samples were

broken during low-strain-rate tests and four samples achieved strains of 0.36 to 0.41% at the test temperature, 625 C. Earlier results from high-strain-rate tests showed six of twelve samples broke in this strain range. Tests are continuing to verify this initial observation.

Annealing Graphite Irradiated at 30 C. A high-exposure (11,100 Mwd/AT) CSF graphite sample cut parallel to the extrusion direction was irradiated at approximately 30 C and then subjected to a series of annealing tests up to 1700 C. The irradiation treatment had caused the c spacing to increase about 15%, and length to decrease 1.23%. The bulk coefficient of thermal expansion (CTE) after annealing at 100 C was 50% greater than the unirradiated value, and the c-direction crystal CTE after a 200 C anneal was about 150% of the original value.

Annealing up to 1700 C restored the original properties of the sample except for a net expansion in the bulk length of 1%. This expansion has not been observed for CSF graphite irradiated to a lower exposure (5000 Mwd/AT) and consequently must be a function of the extent of damage.

The annealing data indicate that the fraction of crystallite dimensional changes transmitted to the bulk decreased significantly upon irradiation, regained the original value after annealing at 1200 C, and exceeded the original value at higher annealing temperatures. This could be explained if the process causing the net bulk expansion closed cracks or pores which take up the crystallite changes.

Several inflections appeared in the curves of c spacing and bulk length changes as a function of annealing temperature. More than half of the length changes occurred from 1100 to 1700 C. Inflections in both curves coincided at temperatures greater than 700 C indicating a close relationship between bulk length and c spacing. The large changes above 1100 C most likely are due to the decrease in damage from the increased mobility of vacancies.

High-Temperature Graphite Irradiations. The second capsule, GEH-13-11, of the high-temperature graphite irradiation series continues to operate satisfactorily. It has completed the first cycle of irradiation in the ETR with all thermocouples functioning. Maximum sample temperatures are being maintained between 1150 and 1175 C.

EGCR Graphite Irradiation. The tenth capsule, H-3-10, in the series of long-term irradiations of EGCR graphite was installed in the GETR. During the initial attempt at installation, the capsule would not seat properly because of a mix-orientation of both the core filler piece and

the grid adapter. These were properly oriented and the installation proceeded without further difficulty.

Revision of the neutron exposures for all samples contained in the nine previous capsules has now been completed. The revisions use the latest nickel and iron activation cross sections in the calculation of effective cross section for the neutron spectra characteristic of the various reactor loadings adjacent to the capsules.

For all H-3 capsules, the exposures increased. These increases varied from +0.7% to +31.2% with a mean value of +17.5%. In all cases the exposure based on the nickel was greater than that based on iron. The nickel exposure varied from +1.4% to +31.5% greater than the iron exposure with a mean value of +12.8%. The maximum neutron exposure is now estimated to be 1.6×10^{22} nvt.

Oxidation of Boronated Graphite. The oxidation rate has been measured for a sample containing 7 wt% boron as boron carbide particles. The sample was oxidized in a stream of helium containing about 1% O_2 and flowing at approximately $150 \text{ cm}^3/\text{min}$. The weight-loss rate increased from $0.5 \times 10^{-3} \text{ hr}^{-1}$ at 530 C to about 5×10^{-3} at 730 C. These rates were determined from a continuous measurement of the sample weight. They are actually net rates because the weight lost by graphite oxidation is partially compensated for by a weight increase from boron oxidation.

At the higher temperatures, the weight-loss rate and the concentration of CO_2 in the effluent gas decreased with increasing oxidation to the sample. This appears to be the result of a decrease in the graphite oxidation rate, probably due to increasing protection of the graphite by glassy B_2O_3 . Future measurements will employ separate samples at each temperature in order to determine the rate in the absence of this inhibiting effect.

Measurements on the various types of Fermi boronated graphites indicate that the "grey" (recrystallized) material has a significantly lower surface area. Pore-size distribution measurements are being performed on the "grey" and "black" samples containing 5% boron. They should help to explain the surface-area differences.

7. Boronated Graphite Studies

Exposure Correlations for Boronated-Graphite Irradiations. Irradiation studies have been conducted to evaluate boronated graphite as a fast-reactor shield material. Although in a fast-reactor shield there would be a relatively high fast flux and almost negligible thermal flux,

most tests have been conducted largely in a high thermal-to-fast flux ratio because of the lack of fast flux testing facilities. It is postulated that the radiation effects measured in a given fast flux would be less than those with the same fast neutron exposure but measured in a thermal-neutron spectrum.

In order to apply these data to fast reactor systems, it is necessary that the damage mechanisms be better understood. The assumption has been made that the persistent damage, such as bulk dimensional change, would be related to the number of carbon-atom displacements produced in the graphite. Displacements would be produced by carbon knock-on-atoms from fast neutron collisions or by energetic Li^7 and He^4 nuclei from the B^{10} reaction with thermal neutrons. By determining the relative effectiveness of each process the relative weighting for fast and thermal neutrons could be evaluated.

Although the bulk of the irradiation data was provided from the tests in the Hanford 2C irradiation facility with a ratio of thermal-to-fast neutrons of about 8.3, tests were also conducted in a shielded facility in the ETR which had a thermal-to-fast ratio of about 0.08. Data from the 2C irradiations were best correlated with ETR data if both were normalized on an exposure scale with corrections for dispersion of boron in the graphite according to the relationship:

$$D = \int_{1.8 \times 10^5 \text{ ev}}^{10^7 \text{ ev}} \Sigma_C(E) N(E) dE + \int_0^{10^7 \text{ ev}} A \Sigma_B N_B dE \quad \text{displacements cm}^{-3} \text{sec}^{-1}$$

where

Σ_C is the macroscopic elastic scattering cross section of carbon,
 N_E is the number of displacements produced by a neutron of energy E ,
 Σ_B is the macroscopic cross section for the $\text{B}^{10} (n, \alpha)$ reaction,
 N_B is the number of displacements produced by the $\text{B}^{10} (n, \alpha)$ reaction,
 A is a term which corrects for the dispersion of boron in the material.

The degree of dispersion of boron has a pronounced affect on the damaging characteristics of the specimens. A maximum of 2% of the boron could go into solid solution with carbon during manufacture if equilibrium were attained. The remainder would exist as B_4C precipitate particles, and $\text{B}^{10} (n, \alpha)$ reactions in these regions would produce little damage to the graphite since the displacement spikes would largely be contained in the particle.

Analysis of the irradiation data yield values for A of about 0.15 for the grey material and 0.05 for the black, indicating three times as

much "effective" boron concentration in the grey. Photomicrographs of these materials also indicate that boron is more highly dispersed in the grey material.

8. Metallic Fuel Development

Irradiation of Thorium-Uranium Fuel Elements. The irradiation of three tubular Zircaloy-2 clad thorium-2.5 wt% uranium-1.0 wt% zirconium fuel elements continued successfully in the ETR-P7 loop. An irradiation of three similar type fuel elements was started in the ETR-M3 loop.

<u>Loop</u>	<u>Core Temp., C</u>	<u>Surface Heat Flux BTU/hr-ft²</u>	<u>Integrated Exposure (Fissions/cm³ Mwd/T)</u>
P-7	425	5.1×10^5	3.8×10^{20} (11,000)
M-3	520	7.1×10^5	1.7×10^{19} (500)

No swelling beyond that expected from solid state fission products has been observed.

Irradiation of Metallic Uranium with Submicron Uranium Dispersions. The irradiation performance of fuel rods containing a submicron dispersion of uranium carbide in uranium is being evaluated.

Two NaK capsules containing Zr-2 clad uranium rods irradiated to approximately 0.3 at% burnup at a maximum uranium temperature of 500 C were opened in Radiometallurgy. Examination of the fuel rods after removal from the capsules showed all three uranium samples with the finer carbide had sustained brittle failures in the cladding even though little fuel swelling had occurred. The one uranium sample with the 2-5 micron size carbide particles shows some increases in diameter but the cladding remained intact.

Metallographic examination, density measurements, and burnup determination are planned for both fuels.

9. Advanced Reactor Concept Studies

Nitride Fuel Cycle Studies. Because of deficiencies reported last month in the 16-group YOM cross section set, a new cross section library has been developed for investigating the nitride fueled fast reactor concept. The new library incorporates 18 groups, with the lowest two groups spanning the resolved and unresolved resonances. Spectral weighting and basic data were obtained from HRG (Hanford Revised GAM) calculations.

Preliminary analysis of the fuel cycle costs for a carbide fueled 1000 Mwe fast breeder reactor design study by Combustion Engineering (CEND-200), and comparison with expected nitride cycle costs, confirms earlier conclusions that costs involved in the conversion to nitride may be critical in establishing the economic status of a nitride fuel cycle, and that considerable incentive exists to develop an economical method for nitride conversion, either by avoiding the use of a metallic intermediate or by performing plutonium reduction-to-metal at costs considerably below the \$1.50/gm AEC Handbook cost.

Comparisons of the conversion costs for carbide and nitride fuels illustrate the importance of this step in the cycle. In the comparisons if the cost of conversion from PuNH to PuN is taken as \$1.50/gm Pu (essentially due to the metal reduction step), and UN assumed at \$32/kg U (the estimated UC cost in the reference study), and if all other factors in the fuel cycle remained equal, the nitride conversion step would increase conversion and fabrication costs by 63% over the carbide costs. The reference carbide fuel cycle does not contain a step involving reduction of the plutonium to metal. It is doubtful that savings from other portions of the cycle, or increased Pu credit, could offset this penalty. Therefore, a major effort is planned in the nitride fuel cycle study to survey means of establishing an economical nitride conversion scheme.

A preliminary assessment of the goals of the fuel cycle study has been written and will be distributed as a BNCC document.

Integrated Nuclear Complex Studies. Three reports are in various stages of completion that consider the economics of an integrated nuclear complex versus isolated reactors with centralized service facilities.

The first of these, "The Effect of Exposure on Fuel Cycle Costs," comments on the possible savings at optimum exposure for an integrated system as compared to a single reactor facility. This document is being circulated in comment issue.

The second document, "Resource Utilization to Obtain Isotopes For Power Application," suggests that one of the possible uses for reactor-grade Pu is to furnish isotopes for producing alpha-emitters. The economics of Cm-242 and 244 production by this method have been calculated.

The third document considers a fuel cycle that can be completely closed at an integrated nuclear complex except for supplies of natural or depleted uranium and thorium. The cycle is based on fast reactor and

thermal reactor teaming in which the fast reactor produces "clean" U-233 to fuel the thermal reactor, and in which the thermal reactor produces Pu to fuel the fast reactor.

Plutonium and U-233 Fueling of a Fast Compact Reactor. All heat transfer and fluid flow calculations have been completed. Plotting of thermal hydraulic results is completed. Portions of the final report have been drafted.

A final nuclear criticality calculation of the reactor core models after coolant passage size zoning indicated slightly excessive reactivity for these cores, but the difference in size from the critical size is so small that it was unnecessary to revise the engineering models for the cores. Radial power profiles are now being compared. Since the SWISS-CHEESE computer code used for coolant passage zoning assumes power is proportional to fuel volume and modifies the power profile accordingly, there should not be a significant difference between the results from nuclear calculations and the assumptions used for engineering analysis.

The gas transport property computer program, MIX, was described in a rough draft of a document for publication. The final draft is being prepared. The core thermal hydraulic code, SWISS-CHEESE, is being documented in rough draft and is about 75% complete.

10. Critical Flow of Steam-Water Mixtures

The installation of equipment for the next series of high pressure critical flow experiments was completed. The most significant difference between the apparatus to be used in this new series of experiments and that used in the earlier experiments reported in HW-80535 is the method employed to measure pressures. The experimental method used to investigate the critical flow phenomenon requires the measurement of a large number of pressures. In the earlier experiments, a separate pressure transducer was required for each test section pressure tap. This transducer reduced the pressure signal to an electrical signal for the automatic data recording system. In the future experiments, the pressures will be sampled by means of a 48-position rotary scanning valve and routed to a single transducer. This method is a significant improvement over the older method because it reduces the amount of random deviations in the various recorded pressure signals. These random deviations resulted from small discrepancies between the calibrations of the different pressure transducers used. It is expected that the new method of pressure measurement will greatly enhance the accuracy and consistency of the data.

11. Fast Flux Test Facility Studies

Authorization was received from the USAEC to expend \$500,000 of FY-1965 funds for conceptual design of a "fast flux test facility" and to "determine its R&D requirements."

A Fast Flux Test Facility Design Section was established, effective March 15, 1965, in the Reactor and Materials Technology Department, the assigned "responsible department." The Manager of the FFTF Design Section (E. R. Astley) was appointed Research Program Leader, FFTF Program. An advisory body, the "FFTF Review Council," was also established.

The FY-1967 Construction Project Data Sheet for the FFTF was revised to \$75,000,000, and submitted to PNL management on this basis.

A scope for FFTF fuel examination facilities was prepared for comment. Principal bases include provision of underwater storage and examination facilities and provision of inert gas storage and examination for test fuels which are incompatible with water.

The hazards analysis group prepared a brief safety criteria for FFTF. One purpose of this action is to gain agreement for more basic safety questions as early as possible. The comment issue is being circulated.

12. Nuclear Rocket Fuels Studies

Studies conducted for AEC-SNPO on tungsten-UO₂ fuel element development and on fueled graphite problems are reported via a separate distribution list.

D. DIVISION OF RESEARCH - O5 PROGRAM

1. Radiation Effects on Metals

Experiments on thermally-activated deformation in unirradiated and irradiated polycrystalline molybdenum have nearly reached a conclusion, although a great deal of interpretative work remains to be done. These experiments were conducted over the temperature range 223 to 350 K, the lower temperature being dictated by the inherent brittleness of this material, and the upper temperature by the minor effect of thermal activation above 350 K. There seems to be little evidence to justify selection of the Peierls mechanism for the controlling process in the unirradiated material at the temperatures investigated. Rather, the results are more consistent with the motion of jogs in screw dislocations. Neutron irradiation results in a significant change in the flow

properties at low strains, but very little change at higher strains (e.g., >15%), at which the deformed substructure is thought to consist of a stable cellular network of dislocations. This observation concurs with the hypothesis that small point defect clusters interact strongly with glissile dislocations and are swept up by these moving dislocations. Evidence also exists that the dislocation multiplication rate in irradiated molybdenum annealed at 600 C is at least an order of magnitude greater than in the as-irradiated condition. This has been interpreted as the bowing out of loop segments from clusters which have collapsed to form small dislocation loops. Further correlation and interpretation of the data are expected to confirm these tentative conclusions.

X-ray diffraction measurements on rhenium foils irradiated to 7×10^{18} nvt ($E > 1$ Mev) have been extended to include foils annealed and cold-worked after irradiation. As reported earlier, irradiation results in a decrease in c_0 and an increase in a_0 , with a net unit cell volume increase of about 0.09%. After annealing at 800 C, the lattice parameters are identical with those of the unirradiated foils. One foil was cold-worked by bending after irradiation. This treatment increased the a_0 parameter slightly but did not change the c_0 parameter.

The irradiated foils were also examined by transmission electron microscopy. Defect structures due to irradiation were not observed in the microscope but when high magnification micrographs were examined, spot defects were found. These defects are 35 Å in diameter and the density of defects is approximately $4 \times 10^{16}/\text{cc}$. The nature of the spots has not been established. However, the x-ray diffraction results and the presumably low vacancy mobility due to the high melting point of rhenium suggest that the spots are interstitial clusters or irresolvable interstitial-type dislocation loops. Annealing the irradiated foils to temperatures as high as 400 C for two hours has caused no change in the size or density of the spots. Examination of specimens annealed at higher temperatures is currently in progress.

In order to study the effect of dislocations on the nucleation of defect clusters, samples of high purity molybdenum were pre-strained to the following amounts prior to irradiation: lower yield point, 2%, 5%, and 20%. Samples which were deformed >20% by rolling as well as annealed samples were also irradiated to 6×10^{18} nvt. Examination of micrographs made from the highly deformed foils shows fewer defect clusters than in the annealed foil irradiated to similar doses. Further study is in progress.

Since the interpretation of electron transmission intensity photographs through thin foils requires a knowledge of the foil thickness, investigations of methods for determining the foil thickness have

been conducted. The dependence of transmitted intensity upon foil thickness and the deviation of the electron beam from the Bragg diffraction condition has served as the basis for numerical and experimental evaluations. Diffraction conditions were varied in annealed and irradiated foils of molybdenum until the intensity of the diffracted beam from one reciprocal lattice vector was significantly greater than the intensity from the other reciprocal lattice vectors. Various diffraction conditions were obtained and the deviations from the Bragg condition were determined by the separation of the Kikuchi lines from the diffraction spot. A paper describing this method for determining the diffraction condition has been prepared. The methods of determining foil thickness from transmitted electron intensities are (1) counting the thickness contours in wedge-shaped foils, (2) measurement of fringe separation in the diffraction plane (Kossel-Mollenstedt fringes) when a diffuse incident beam is used, (3) measurement of fringe separation in a plane containing the center of curvature if the foil is bent, and (4) measurement of the separation of intensity maxima as a function of deviation from the Bragg condition. The last method has not been successful for the foils tested thus far since the measurements of the intensity, the increments of foil rotation, and the separation between the Kikuchi lines and diffracted beam cannot be measured or controlled with sufficiently high precision.

The nature of the physical changes in irradiated molybdenum which result in the low-temperature (<200 C) length changes, stored energy releases, and resistivity changes are being investigated by x-ray methods. A foil containing <10 ppm carbon irradiated to 1×10^{19} nvt ($E > 1$ Mev) showed no measurable lattice parameter change on prolonged annealing at 200 C. However, a foil with 100-200 ppm carbon showed a small decrease of about 0.006% from 3.1483A to 3.1481A after four hours at 200 C. Changes of this magnitude are of the same order as the experimental error of the measurement, so this result must be considered tentative until verified. Since the lattice parameter change at this temperature is negligible, it may be concluded that vacancies, which have a proportionately small effect on lattice parameter, must be the defect species concerned in the low temperature annealing process, and that the process probably consists of vacancy migration to sinks such as interstitial clusters.

A second approach to the study of the point-defect movements in molybdenum involves the precision measurement of x-ray intensity diffracted from a single crystal face. Faces parallel to (110) and (100) were cut on annealed and annealed-irradiated single crystals. Electropolishing removed the cold-worked material from the surfaces. X-ray intensities were measured by the symmetrical Bragg method and were converted to

an absolute scale by calibrating with a standard aluminum crystal. These experiments indicated that the annealed crystal surfaces were highly perfect. Irradiation and postirradiation annealing at 200 C did not change the measured intensities appreciably. This result is unexpected, in view of the high density of spot defects observed with the electron microscope. These experiments are being repeated with samples having faces cut and polished prior to irradiation, to investigate the possibility that the surface perfection is a result of the polishing treatment.

A note entitled "Effect of Neutron Irradiation on Rhenium" has been submitted for publication in the Philosophical Magazine. An article entitled "Combined Effect of Carbon and Neutron Irradiation on Molybdenum" has been submitted for publication in Acta Metallurgica.

2. Plutonium Physical Metallurgy

The development of texture in alpha plutonium samples by means of stressed phase transformation has continued. It has previously been reported that the [010] texture developed by carrying out the beta to alpha transformation under a compressive load is apparently shifted 90 degrees by a subsequent cycle into the beta region. A tensile stress during the beta to alpha transformation favors this latter texture in which the [010] is perpendicular to the direction of applied stress.

It appears from these data that the <010> direction is the direction of major change in the dimensions of the specific volume involved in the beta-alpha transformations. During the contraction inherent in the beta to alpha transformation this <010> tends to line up parallel to the compressive stress which favors the transformation while in the case of a tensile stress which restrains the transformation this direction is perpendicular to the applied load.

With these facts in mind, it appears that alpha to beta to alpha cycles with tensile stresses applied during the alpha to beta portion and a compressive load during the beta to alpha portion of the cycling would enhance the resulting texture. An apparatus which permits cycling under such conditions has been constructed and an initial test has been made. The results thus far obtained agree with the predicted texture. With the exception of a thin circumferential shell of near random grains, the sample exhibited almost perfect texture. In the transverse section only the (020) reflections appear in the diffraction pattern if the sample surface is maintained essentially parallel with the diffractometer axis. The extent to which the reflecting portion varies from this coincidence with the

diffractometer geometry can be accounted for on the basis of inaccuracies in cutting the specimen. It has been found possible to bring several strongly diffracting planes individually into a reflecting position by altering the goniometer angles. In this respect the sample behaves much as a single crystal over surface areas of the order of square centimeters. The volumes of these pseudo-single crystals can as yet only be inferred. An attempt is under way to plot these diffracting plane positions on a pole figure and check their interangular relationships with those calculated from the known crystal structure and unit cell dimensions.

It is interesting to note that although these transformations were permitted to proceed near the equilibrium temperatures, there is very little evidence of microcracks as indicated both by the microstructure and density determinations.

The x-ray diffraction study of these textured samples has revealed several apparent anomalies. For example, the second and fourth order reflections of (010) appear to vary in relative intensity, depending upon the area of sample being irradiated. The relative intensity of the (040) reflection has been observed to vary from a value of 40% to less than 10% of the (020) intensity. The slit systems and other diffractometer parameters were not altered. Since it is, in some cases, necessary to depend upon the (040) reflection as a means of identifying the (020), since the latter is almost coincidental with the (211), this variation in relative intensity results in confusion in the interpretation of the pattern in some cases. Some feature of the alpha structure, as yet unknown, must be responsible for the observed anomalies.

The beta to alpha kinetics of electrefined plutonium received from the Los Alamos Scientific Laboratories have been established. The transformation rates are slightly higher in this metal than in metal received in 1963.

The beta to alpha transformation of plutonium has been classified as a martensitic type transformation. This conclusion is based on the following general observations: (1) Both uniaxial tensile stresses and uniaxial compressive stresses raise the temperature for the beginning of the beta to alpha transformation at a fixed cooling rate. (2) Stresses applied during either the alpha to beta or the beta to alpha transformations produce pronounced crystallographic texturing and microstructural shape texturing. (3) The anisotropic alpha to beta and beta to alpha transformations result in a 90-degree rotation of the crystallographic and microstructural textures and imply that a specific crystallographic orientation relationship exists between the

parent and the product phases, a characteristic to be expected of martensitic transformations. (4) The kinetics data are consistent with thermally activated isothermal martensitic transformations.

E. CUSTOMER WORK

1. Radiometallurgy Laboratory

Examinations. The results of routine examinations and measurements are or will be reported as part of the sponsoring research and development programs. During the reporting period, examination and testing included the following:

Photomicrography - - - - -	34
Photomacrography - - - - -	112
Photomosaics - - - - -	8
Autoradiography - - - - -	29
Replication - - - - -	1
Fission gas - - - - -	2
Hydrogen analysis - - - - -	43
Density - - - - -	13
NaK capsules - - - - -	3
Decladding - - - - -	3
Dissolution - - - - -	13
Microhardness - - - - -	14
Macrohardness - - - - -	309
Tensile Testing (Rm temp) - -	64
Tensile testing (Hi temp) - -	50
X-ray - - - - -	12

Project CAH-136, Service Addition - 327 Building. The low construction bid submitted by George Grant Company was accepted. Minor Construction forces started radiation zone work which must be completed before the lump-sum contractor can start construction.

"E" Cell Metallographic Facilities. The "E" cell metallographic facilities were completed and placed into operation. New rough-grinding, fine-grinding, and polishing equipment was installed to provide faster specimen processing.

Zeolite Ion-Exchange Capsule Test Equipment. Design and fabrication of the capsule positioning device for remote welding was completed. Satisfactory welds were made in a test mockup.

Stereo Zoom Viewer. Modification of the stereo zoom viewer positioner is in progress so that the unit can be used as a measuring microscope as well as a magnified viewer.

Molding of Spline Seals. The prototype spline seals which were molded for IPD Plant Engineering performed satisfactorily during testing.

Remote Metallograph. Consulting assistance was provided Technical Shops on preparation of a bid for fabrication of a remote-controlled metallograph.

Remote Disassembly and Sampling. A report entitled "Disassembly and Sampling of Irradiated Materials in the United States" was completed for presentation at the International Symposium on Working Methods in High Activity Hot Laboratories.

2. Metallography Laboratories

Routine metallography work 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.

The microstructural detail of a thorium, 2-1/2% uranium, 10% zirconium alloy clad in niobium was revealed considerably better than has been observed with previously known chemical etches. The alloy, in fact, resisted all attempts at etching until a solution normally used for pure niobium metal was tried. The etch, consisting of 3 parts lactic acid, 1 part nitric acid, and 1 part hydrofluoric acid was applied by swabbing.

During the report month 525 samples were processed, 238 electron micrographs and a total of 1222 optical micrographs and macrographs taken, 2427 negatives printed, and 8636 prints processed.

3. High Temperature Lattice Test Reactor

Environmental Tests on HTLTR Materials. The high temperature reaction of water with graphite - 5% B₄C are being investigated in a series of tests in which this control rod material is exposed to nitrogen-water mixtures at 1100 C. In a test currently under way nitrogen containing 5000 ppm water is being passed through the test chamber. The weight change of the B₄C - graphite material will be determined for this and other water contents for various times to predict reaction rates for HTLTR conditions.

Thermocouples which were sheathed in molybdenum, Inconel 600, and Hastelloy B were examined metallographically after exposure during the first run of the 314 mockup. Corrosion effects on the Hastelloy B were severe, moderate on the Inconel 600, and nonexistent on the molybdenum.

The second heatup of the mockup has started and the core temperature has reached 260 C. The system was alternately evacuated and then filled with nitrogen three times at room temperature. A total of 350 ml of water was removed during the room temperature evacuations which lasted 24 hours. A heated evacuation at 230 C, lasting approximately 15-1/2 hours, resulted in the removal of 500 ml of water. During the recirculating portion of the fill cycles, O₂, CO, CO₂, H₂O, and N₂ concentrations of 0 to 0.2%, 0%, 0.2 to 0.3% and 99.8 to 99.9%, respectively, were observed.

The gas cleanup system is now in service. Concentration of water vapor volume in the recirculating gas entering the mockup, was reduced from 2400 ppm to 800 ppm. Water content in the cleanup system exit gas is down to 38 ppm.

A problem with induction heating in the shell near the exit bus bar has been encountered since electrically insulating the bar from the shell. Installation of a cooling coil in this vicinity has been completed as an interim solution to the problem.

Twelve scrams of both the horizontal control rod and vertical safety rod were completed during the present temperature rise. No malfunctions occurred.

Critical Mass Physics Support Work. Fabrication of the Mark-II safety rod for the variable volume slab tank is 85% complete, and the assembly is 35% complete.

Installation of the concentrator and condenser for testing in 314 Building is complete. The concentrator volume has been calibrated and preliminary boil-off studies have been made. These studies indicate a non-reflux condensate rate of eight to ten liters per hour. The limiting boil-off rate will be established by carry-over tests now in progress.

4. EBWR Fuel Elements

EBWR Plutonium Recycle Fuel. On March 8, 1965, 715 Zircaloy clad UO₂-1.5 PuO₂ fuel rods were shipped to Argonne and will complete the plutonium fuel required for the EBWR core in support of the Plutonium Recycle Demonstration Experiment. Another 150 spare fuel rods are being fabricated and are scheduled for delivery to Argonne in April.

Thermal Cycling of Aluminum Core EBWR Rods. Several EBWR rods have been fabricated for ANL with AlPu cores, and in order to obtain information on dimensional changes which might occur in service, thermal

cycling studies are being performed. The rods are heated in an autoclave to 370 C and 600 psig in one hour, held for three hours and cooled to 90 C and 0 psig over an eight-hour period. Measurements of rod diameter and lengths are made every two cycles. In general, rod lengthening was obtained initially reaching a maximum of 0.039 inch after six cycles; subsequent cycling produced rod shortening up to 0.052 inch after 24 cycles. Anomalous results were obtained on one rod which exhibited an abnormally close fit on loading. This particular rod has shown very little growth or shrinkage after 24 cycles. All rods show only minor (0.001-0.002 inch) diameter changes.

Stress Studies of Cladding During Vibratory Compaction. Strain gauge measurements were made of stresses encountered in EBWR Zircaloy-2 tubing during fuel loading on the M-B electrodynamic shaker unit. Stress measurements were limited to the area of tubing immediately above the end cap clamp assembly. Comparisons were obtained between a normal clamp arrangement, a normal clamp mounted on a reinforced resonance plate, and a modified tapered end cap holder on a reinforced resonance plate.

During three loading runs cladding stresses from excited bending modes, under an applied load of 20 g's were similar, with maximum stress values ranging from 11,000-15,000 psi, the higher values being associated with the modified tapered clamp. Reinforcement of the resonance plate appeared to increase the number of excited bending modes and also to increase stress in the cladding. With increased applied shaking load cladding stresses increased appreciably up to 25,000 psi for an applied load of 40 g's and a normal clamp arrangement. With the other clamp arrangements values from 28,000-55,000 psi were measured at this applied load. Loading and packing of the fuel powder appeared to intensify cladding stresses slightly when vibrated at constant load.

Irradiation Testing of EBWR Prototypic Fuel Rods. Seven capsules (one with prototypic fission gas plenum) are being irradiated in the MTR and 25 have been discharged. Maximum exposures achieved are 4.5×10^{20} fissions/cm³ (16,000 Mwd/ton of fuel) and 4.8×10^{20} fissions/cm³ (17,000 Mwd/ton of fuel), respectively. The Zircaloy-clad, UO₂-1.5% PuO₂ specimens (1.08 cm OD) continue to perform satisfactorily.

The 21-rod bundle (5x5 array with corner rods missing), Fuel Element No. 6501, with full-size, replaceable EBWR fuel rods successfully completed one reactor cycle of irradiation in the rupture loop of the PRTR. The bundle was subsequently charged in a PRTR core position to

continue the irradiation experiment. The bundle acquired an exposure of 10.6 Mwd or 551 Mwd/ton of fuel or 0.16×10^{20} fissions/cm³ in the rupture loop and currently has an accumulated exposure of 16.5 Mwd or 860 Mwd/ton of fuel or 0.25×10^{20} fissions/cm³.

The twelve, three-rod clusters of full-size (148 cm long) EBWR rods under irradiation continue to indicate satisfactory behavior.

Nineteen rods (two contain high-exposure plutonium - 20% and 27% Pu-240) are being evaluated for density and plutonium distribution and a portion will be used for planned rod replacement in the 21-rod, PRTR irradiation bundle.

Saxton Fuel Element Fabrication. Eighty-four Saxton rods were fabricated varying in density from 86%-88% TD. The rods are now being etched and autoclaved in preparation for shipment in early April.

5. Other Customer Work

Carbon-Sm₂O₃ and Carbon-Gd₂O₃. Service work on potential reactor poison materials was conducted for the Irradiation Processing Department, General Electric Co. Pellets of carbon-5% Sm₂O₃ and carbon-0.5% Gd₂O₃ were prepared by cold pressing. These pellets were heated for eight hours at 1000 C. Chemical analyses which will be conducted by General Electric Company will be used to reveal the formation of samarium carbide and gadolinium carbide which are detrimental to in-reactor use because of their high reactivity with water.

Westinghouse UO₂. Pneumatic impaction of 11 containers of UO₂ for Westinghouse was completed. This material was canned by Westinghouse and will be decanned by them. The only service performed here was the pneumatic impaction.

Manager,
Reactor & Materials Technology

FWA:kb

PHYSICS AND INSTRUMENTS DEPARTMENTMONTHLY REPORTMARCH 1965FISSIONABLE MATERIALS - O2 PROGRAMREACTORCoprodut Experiments for N-Reactor in the Exponential Pile

Experiments to determine material buckling B_m^2 and extrapolation distances in the coprodut lattice have been completed in the N-mockup exponential pile. Seven different experiments were performed and analyzed. The final report is being typed.

Coprodut and Control Rod Experiments for N-Reactor in the PCTR

Perturbation experiments are planned for the PCTR to determine the relative worth of a ceramic to a metal target, and of a standard B_4C control rod to an amorphous B^{10} control rod. A metal target having the isotopic composition of the standard production model has been received from N-Fuels for the experiment. A standard B_4C control rod sample and a new enriched boron control rod sample are also on hand for the experiment. Preparations for the experiment are nearly completed so that it will be ready to go when the reactor is available.

Experimental Study on Pu-Al-Thorium Lattices

Planning is under way for a PCTR experiment to measure values of k_{∞} for both wet and dry lattices, moderated with graphite and consisting of alternate cells of Pu-Al fuel and thorium target. Required fuel lengths have been specified; the graphite core has been designed; and plans for the experiment are under way.

NPR Coprodut Studies

In last month's progress report it was reported that diffusion theory underestimates the (PCTR) measured k_{∞} of the NPR coprodut lattice by approximately 25 mk. This error was tentatively attributed to too soft a spectrum in the highly absorbing target material which, in turn, would overestimate target absorption rates. This conjecture has been substantiated by comparing diffusion theory calculations with transport calculations. Quantitatively, the ratio of target to fuel thermal absorption rates in diffusion

theory is 20% greater than in transport theory. Correcting for this systematic error yields a predicted k_{∞} for the cell that is only 2 mk higher than measured in the PCTR. Further, these comparisons illustrate the suspected inadequacy of diffusion theory for this highly absorbing cell having discrete absorbing annuli.

In passing, it has been noted that the transport code THERMOS and double-P3 code FLEX yield results in this comparison that are in essential agreement.

Development of Calculation Methods for Tube-in-Tube Geometry

A fairly detailed comparison of diffusion and transport calculations for tube-in-tube geometries is being carried out. While it is recognized that transport calculations are in general preferable to diffusion calculations for discrete fuel geometries, the greater ease of diffusion calculations often makes that approach more desirable. Under certain conditions very good results are indeed achievable with the simpler diffusion approximation.

Comparisons have been made for a tube-in-tube, NPR geometry using the diffusion code HFN and the S-XIII, S_n transport code. k_{∞} 's of 1.05056 and 1.05846 were obtained, respectively, from S-XIII and HFN for a difference of only 7.9 mk. The thermal flux depressions obtained from diffusion theory for the inner and outer fuel regions were, respectively, 7% and 14% less than those obtained from transport theory.

The k_{∞} values agree well because most of the absorptions take place in the fuel region and discrepancies in the spatial flux distribution are, therefore, minimized.

Preparation for PCTR Driver-Target Supercell Experiments

A series of PCTR experiments is being planned to investigate the characteristics of supercells consisting of Pu-Al drivers and ThO₂ targets. In support of these experiments, a series of two-dimensional calculations has been completed to determine the variation of k_{∞} as a function of Pu concentration in the Pu-Al driver. In all calculations a driver-target ratio of unity was assumed. The results of these calculations will be used in the specification of Pu-Al driver elements for the PCTR experiment.

Survey Calculations for Driver-Target Configurations for Production Reactors

A series of survey calculations is under way to determine optimum driver-target arrangements for possible use in production reactors. At present, only Pu-Al drivers and ThO₂ targets are being considered in the calculations.

Instrumentation

A second complete channel for the N Reactor subcritical neutron monitoring system is being readied for installation in accordance with shutdown schedules.

A prototype irradiated fuel cooling age monitor employing scintillation detector and solid state circuitry was delivered on schedule to C Reactor. Initial tests are in progress at the cooling basin and general operation appears to be satisfactory.

System Studies

A N Reactor conversion system study which simultaneously employed both analog computers has considered the effects of primary and secondary coolant system variables on turbine trip-offs with and without reactor scrams. Runs were made for both the five cell, 110 psia steam header pressure case and the six cell, 150 psia steam header case. The effects of varying condensate temperatures, the number of on-line dump condensers, main header pressure setpoints, and controller settings were determined.

SEPARATIONS

Criticality Experiments with PuO₂-Polystyrene Assemblies

Criticality studies were continued with PuO₂-polystyrene compacts using the Remote Split-Table Machine. The fuel compacts contained ~ 8 wt% Pu²⁴⁰ and had a total plutonium concentration of 1.03 g/cc with an H/Pu²³⁹ atomic ratio of ~ 16 . The experiments provide data for evaluating the effect of Pu²⁴⁰ content on criticality.

The fuel compacts were covered with thin coats of rubberized plastic to control contamination. The evaluation of the effect of this coating on the critical mass was made by measuring the critical sizes of reflected and bare arrays with various thicknesses of the coating material placed between the cubes. Extrapolation to zero thickness will then give the critical size of a clean system; i.e., one that contains no plastic coating material.

Several foil exposures were made in the fuel assembly. These exposures were directed at obtaining spectral sensitive measurements in the plutonium-oxide core. Initial exposures were made with dysprosium, lutetium, and plutonium-239 foils. Plans are being made for the exposure of other foil materials to better define the neutron energy spectrum.

Pulse Neutron Source and Reactor Noise Experiments

Pulse neutron source measurements were made on a bare assembly of PuO_2 -polystyrene compacts containing about 8 wt% Pu^{240} . The base dimensions of the assembly were 14.29 x 14.23 inches. The average plutonium concentration in the assembly was 1.03 g/cc and the H/Pu^{239} atomic ratio was 16. The value of β/λ for the assembly was 1357 sec^{-1} which is about 25% less than the value observed (1763 sec^{-1}) on a similar assembly where the Pu^{240} content of the plutonium was 2.2 wt%. This corresponds to an increase in the neutron lifetime from 1.2 to 1.6 μsec as the Pu^{240} content is increased from 2.2 to 8.08 wt% and is in quantitative agreement with previous observations made on systems where a small amount of the Pu^{239} was replaced with Pu^{240} .

Noise recordings were also made on this assembly; however, final analysis of the data has been deferred pending the results of a calibration technique currently being attempted on the reactor noise analyzer.

Subcritical Experiments with Enriched N-Fuels for Nuclear Safety Guidance

A series of neutron multiplication measurements was started with 1.96 wt% U^{235} enriched tubular fuel elements (2.345 in. o.d.; 1.364 in. i.d.) in light water. These measurements are being carried out in the TTR room of the 305-B Building. They will provide requested data for nuclear safety guidance in processing and storing of enriched N-Reactor fuels. The critical masses will be estimated from inverse neutron multiplication curves and buckling values determined by measurements at two different heights with the same fuel spacing.

Fuel columns of 27 and 40 inch lengths will be loaded into hexagonal lattices. The fuel elements are zirconium clad. The spacing for the first lattice is 3.1 in. ($\text{H}_2\text{O}/\text{U}$ volume ratio of 2.22). Other templates for lattice spacings of 2.8 and 3.4 have been fabricated to cover the range of maximum buckling and minimum critical mass.

Calculation of Critical Mass for Reflected Pu^{238} Metal

Pu^{238} is being made at Savannah River from irradiation of Np^{237} shipped from Hanford. There has been some concern about the criticality of this isotope because of the high fast fission cross section and the relatively large amounts under consideration for use in radioisotopic heat sources. A Monte Carlo calculation using the SMC Monte Carlo computer code gave a value of $2.66 \pm 0.27 \text{ kg}$ for Pu^{238} metal (density 19.6 g/cc) in spherical geometry, when surrounded by a 10 cm stainless steel reflector. The unreflected critical mass was previously calculated to be $5.24 \pm 0.35 \text{ kg}$. The error limits given represent the statistical uncertainties in the Monte

Carlo calculation but do not reflect uncertainty in cross sections which are undoubtedly large for Pu^{238} .

The Pu^{238} fast fission cross sections were obtained from the measurements of Butler and Sjoblom at Argonne. The Pu^{238} capture, elastic and inelastic cross sections were assumed to be defined satisfactorily by the Pu^{240} cross sections in the RBU basic library. Anisotropic elastic scattering was determined from the first six terms of a Legendre series.

Critical Mass Laboratory Instrumentation

Calibration of the noise analyzer at the Critical Mass Laboratory was completed. Replacement of the output smoothing circuits with ones of longer time constants necessitated the recalibration of this instrument. The calibration was accomplished with a sine wave oscillator rather than a random noise generator. Each of the 40 bandpass filters was calibrated at its center frequency with equal gain. Using the oscillator and treating the bandpass filters in this manner make the calibration easier and more accurate than can be accomplished with a random noise generator. When calibrated in this manner, a pre-whitened spectrum is produced. This type of spectrum attenuates the first 15 low frequency filters off scale. These filters are then calibrated on a -10 db per decade curve to bring them on scale with the others.

Consulting Services on Nuclear Safety - Criticality Hazards

1. Nuclear Safety in Battelle-Northwest

The following nuclear safety specifications were reviewed:

- a. Temporary Specification No. 2 for Metallurgy Development revising limits for PuO_2 -polystyrene block fabrication.
- b. Temporary Specification No. 8 for Metallurgy Development covering Model 600 part processing.
- c. Temporary Specification No. 8A for Reactor Lattice Physics covering the loading of fuel cans with 20 wt% Pu-Al alloy discs.
- d. Temporary Specification No. 9 for Plutonium Fuels Engineering covering fabrication of Saxton fuel rods.
- e. Specification B-12 for Experimental Reactors covering 18-23 wt% Pu-Al alloy discs.

- f. Specification C-19 for Critical Mass Physics covering 1.96 wt% enriched N-Reactor fuel.
- g. Specification J-4 for Metallurgy Development revising limits for handling metallographic samples.

2. Nuclear Safety of Off-Site Shipment of Fissile Material

Two shipments were approved: 510 g PuO_2 to Mound Laboratory (Class I), and 810 EBWR fuel rods (1.5 wt% $\text{PuO}_2\text{-UO}_2$) to Argonne National Laboratory (Class III).

3. Lecture on Nuclear Physics and Nuclear Safety

A lecture entitled "Nuclear Physics and Nuclear Safety" was given for the Health Physics Certification Refresher Course on March 19, 1965.

Separations Instrumentation and System Studies

Tests in process at 234-5 Building are evaluating modifications recently made to an instrumentation system developed to measure Am^{241} in chemical columns.

METALLURGY - Nondestructive Testing

N Fuels Testing

The surface contamination tester developed to monitor uranium contamination on finished N fuels, has been installed, checked out, and is undergoing a production evaluation by N Fuels, Quality Control. Prior to delivering the unit, each of the six test heads was individually calibrated against known microgram planchett standards. Other instrument modifications have been made which permit contamination tests to be made on inner and outer fuels simultaneously.

Ultrasonic transducers have been ordered to evaluate a through-transmission cap-to-clad bond test. This method is expected to reject cap defects of the type that caused the recent fuel failure. It also detects unbond in the end cap braze.

Preliminary tests to detect clad-to-core unbonds have given encouraging results. Further production line tests are planned.

The beveled portion of 200 special fuel elements of the chevron end cap design were successfully tested in spite of abnormal contour difficulties.

A special crystal manipulator has been designed for this test. Fabrication of this fixture is scheduled for completion early in April.

TIG braze closure test, designed to insure melting of the braze wire, is being fabricated on schedule. An order has been placed for the special transducers required for this project. A prototype crystal mounting and manipulator is under fabrication. Delivery is expected April 5.

There is evidence that irradiated N fuels could fail by splitting of the outer clad as a result of circumferential stresses concentrated at longitudinal striae in the cladding. An eddy current tester is being developed to detect these striae.

Mechanical portions of the remotely operated irradiated fuel test station (for use in 105-N) is being assembled at 189-F. Parts from the inactive 100-K tester are being utilized.

N Reactor Department - Test Engineering Support

Work on detecting fuel spacer rattle in the 189-D NPR-PCE loop was reactivated. A 60 cps Fuel Cap Tester was modified to operate at 1500 cps and to provide a filtered output suitable for a strip chart or oscilloscope readout. Vibration amplitude, frequency, and impact velocity are to be quantitatively recorded during initial tests.

In conjunction with the periodic steam generator inspections, NRD has requested the development of a system to detect defects in that portion of the tubing which is rolled into the tube sheets. A developed ultrasonic system consisting of two transducer assemblies, one for the axially and one for circumferentially oriented defects is presently being used. Both transducers are mounted on a lead screw which translates and rotates the transducers (in a self-contained water chamber) into the first six inches of the tube.

Work continued on the engineering study of a tester to measure the enrichment of fuel elements as they are charged into N Reactor. The instrument will employ a detector for each of the three positions of the charging machine, and a fourth detector is planned for the monotube loading machine. The electronic console is to be positioned in an adjoining room. Laboratory tests show that a 1/8 inch change in detector-to-fuel spacing, about what value is expected in practice, causes a 3% change in detector counting rate. Special techniques are being considered to minimize the effect of this variable.

AlSi Fuels Testing

Work continued on the dual bond test being developed to simultaneously monitor the interior and exterior bond quality of AlSi fuels. Fabrication of a laboratory prototype, containing approximately 50% of the necessary electronics, is now complete. Laboratory checkout has made all circuitry operable. Design of the remaining electronics has been initiated.

Preliminary measurements of the emission spectra of the AlSi end cap welding arc suggest that equipment less sophisticated than originally envisioned may reliably reveal pin hole formation. Laboratory analysis of spectra centered at 6560 Å(H) and 5875 Å He readily revealed: (1) A small drop of water on the end cap, (2) a small bead of oil on the end cap, (3) a speck of paraffin on the end cap, (4) heating of the fuel element, and (5) spongy welds. These experiments were performed in the laboratory using a Simplex welder and a Beckman Spectrophotometer which was connected to a 4-ft. fiber light pipe.

The experimental apparatus has been moved to the 306 pilot plant where it will be used to study the arc spectrum of fuel elements that are being specially prepared to have a high defect probability.

IPD - Test Engineering Support

The observation of severe hydriding in two K Reactor Zircaloy-2 process tubes has created need for a rapid, accurate, nondestructive method of detecting the presence of hydriding in the installed tubes at K Reactor. A preliminary study utilizing a developmental 100 kc eddy current tester has indicated that this test should be conducted at approximately 3 megacycles. This problem is currently receiving intensive investigation on several candidate methods to determine optimum inspection procedures.

An investigation to determine the feasibility of using ultrasonics to determine percent of bonding of ribs welded inside Zircaloy-2 process tubes was started. The ribs are .075 in. x .100 in. welded inside a .040 in. thick Zircaloy process tube. Two ribs are welded 90° apart down the length of the tube. Testing is necessary to assure integrity of the weld, since a slight misalignment during welding can cause incomplete bonds.

A pulse echo test using a probe which traverses the inside of the tube to assure proper transducer-to-rib alignment appears the most promising, as was revealed by tests made to a section of Zircaloy-2 sample machined to simulate a bond standard in steps of 11, 33, 50, 67, 84 and 100% bonding. High sensitivity broadband transducers, necessary for resolving the received echoes, have been ordered.

NEUTRON CROSS SECTION PROGRAM - 02/04 PROGRAMTriple-Axis Spectrometer

The use of copper crystal monochromators for the analyzing spectrometer is being studied. Available copper crystals show reflection curves which are much more symmetric than those of the beryllium crystal now in use. The use of crystals with more symmetric reflection characteristics should decrease the systematic errors now encountered. Methods of cutting thin copper slabs are being investigated in the hope of producing a wider effective mosaic from a composite of several slabs of crystal.

Time-of-Flight Spectroscopy for Slow Neutrons

The resolution elements for the Rotating-Crystal Spectrometer were calculated for a range of rotor speeds, incident and down-scattered neutron energies, scattering angles, different beam profiles, and crystal planes. A manuscript describing the Rotating-Crystal Spectrometer was partially completed.

A number of repairs and modifications were made on the TOFA and the transistorized counting circuits.

Elastic scattering distributions from zirconium, vanadium, and zirconium hydride were obtained using the stationary crystal method.

The calculation of the differential inelastic scattering cross section of zirconium hydride by means of the computer programs LEAP, ADDELT, and SCS has been started. It has been determined that LEAP is unable to completely calculate the scattering from the acoustical modes when they are assigned only a one percent weight. Work is in progress to correct this deficiency of the program.

Fast-Neutron Cross Sections

The revision of the data reduction program BIG NED for an improved error analysis has continued. Work has also progressed on an "overlaid" version of the program LIL NED which can read from the Hanford binary library tape. Work has also progressed on the program TAPEWORM which prepares, updates, and copies the binary library tape.

Preparations have been made for a series of total cross section measurements during April. The deflection plate assembly for the rf oscillator has been reinstalled and tested on the Van de Graaff. Several components of the time-of-flight electronic system have been revised.

REACTOR DEVELOPMENT - 04 PROGRAMPLUTONIUM RECYCLE PROGRAMPRTR Gamma Scan Facility

Calibration of the PRTR Gamma Scan Facility was resumed this past month after completion of work modifying the collimator head and aperture. The redesigned collimator head includes a shutter and the aperture can now be closed to obtain background readings. The head has also been modified to enable better positioning. The aperture has been increased to 1/4" diameter throughout the collimator.

As a result of these modifications, the count rate has increased and data can be obtained in one-third less time than needed before. The shutter completely shields the source and the background count is the background of the basin (~ 450 counts/min). An average count to background ratio is 1/120. The electronic systems are constant, and negligible drift was experienced over a three-day operating period. All difficulties experienced due to positioning have not been eliminated, however, and the data taken show that the magnitude of error in count rate when positioning the collimator is $\sim 6\%$. Further modifications to correct this difficulty are being considered.

HPD Core in PRTR

Previous calculations on the new high power density core in the PRTR have utilized a solid rod equivalent for the 19-rod cluster. In order to assess the importance of this approximation, a comparative study was made using a concentric ring model. It was found that the calculated thermal η_f is insensitive to the model used.

Preparations for the PRCF experiment using the new short fuel (scheduled for July) have begun. Special fuel elements for flux measurements have been ordered and a letter has been written giving a brief description of the tests, and an itemization of special equipment that will be required. Calculations pertinent to these measurements have been started.

Analysis of Pu-Al Criticals

A typical Pu-Al critical (5 w/o, 1.0" L.S.) has been studied in detail to determine the sensitivity of the calculated effective multiplication for the "just-critical" loading to small changes in the various cross sections used as input in an HFN calculation. The calculation employed the standard 4-group energy mesh and two spatial regions (core and reflector). The

change in k_{eff} resulting from a 10% increase in each input cross section was calculated. The results show that, in addition to the obvious importance of the thermal absorption and fission cross sections, the fast group parameters (diffusion coefficient and inelastic scattering cross section) are also quite important. For example, a 10% increase in the fast diffusion coefficient in the core region results in a 1.3% decrease in the calculated value of k_{eff} . These results permit a quantitative definition of those areas in which more analysis of basic data is required.

Reactor Physics Code Development

1. LISTIV

A FORTRAN IV subroutine, LISTIV, was written to replace the FORTRAN II subroutine, INPUT/LISTIN/UNIT. The objective of LISTIV is to make LISTIN-type input possible with two classes of programs: (1) Those FORTRAN II codes which are being converted to FORTRAN IV without extensive revision, and (2) new "quick-and-dirty" applications, in which programming time can be cut and data preparation simplified because of the familiarity of the input format. BNWL-41, a descriptive document for LISTIV, was issued.

2. Decay of Resonance Radiation in Gases

As one of a series of studies initiated to evaluate the capabilities of current analysis methods in transport phenomena, Program S is being used to study the imprisonment of resonance radiation in low density gases. A Doppler-broadened resonance line is assumed (natural broadening is unimportant in the low opacity systems considered). Preliminary results indicate reasonable agreement with experiment. The emitted light shows the proper self-reversal, and the imprisonment time increases approximately linearly with the line-center opacity. The extension of this study to include larger opacity systems is in progress.

3. Gamma Ray Transport

To initiate studies of high energy (0.1 MeV - 1.0 MeV) photon transport, we are in the process of writing a code to compute Klein-Nishina cross sections (in this energy range Compton scattering is the primary scattering process). Practical uses of high energy photon transport are found in the field of nuclear shielding.

4. Comparison of THERMOS with Program S-XIII

Work has begun to compare the results of Program S-XIII and THERMOS

when both programs use the same cross sections and source as input. While Program S-XIII computes more useful information and is potentially more accurate, its running time was typically three to four times longer than THERMOS for the cases studied.

Pu-Al-D₂O Experiment in the PRCF

The moderator coefficient of reactivity was determined between 98" and 105.5" in a Pu-Al core moderated with D₂O. The total change in k_{eff} due to raising the moderator over this range is 5.5 mk or about twice what it was worth in a two-zone core of Pu-Al and UO₂ fuel elements. A control rod was calibrated from 3.75" to 6", and had a worth of about 1.36 mk for the movement. This is about 1-1/2 times its worth in the two-zone core; hence, its total worth is expected to be about 2.8 mk. From kinetics experiments and subcritical multiplication data, the worth of a safety rod was determined to be about 46 ± 2 mk. The worth of a 19-rod cluster has not been determined very accurately, but is estimated to be about 45 mk. Flux data from bare and Cd covered BF₃ counters have been obtained.

Three small BF₃ ion chambers for measuring reactor noise and kinetics are being constructed. Other equipment is also being assembled and tested for reactor noise and kinetics experiments in the Pu-Al-D₂O core of the PRCF. A break frequency is expected for the Pu-Al loading which is lower than those found in previous measurements. This requires lowering the frequency response of the recording system to 0.01 cycles/sec.

For kinetics experiments using a digital voltmeter, an accurate electronic timer is required. An existing timer is being modified to replace three pieces of equipment which were used previously. The use of this timer eliminates previously required adjustments. It will determine more accurately the time at which the readings are obtained. The timer also provides a signal to trigger an oscilloscope.

The spectral indices T and r are being measured at several positions. The first of two irradiations employing lutetium foils has been completed. Foils were placed radially across the central fuel cluster and in the moderator between clusters. The cadmium covered and bare foils will be reversed in the second irradiation.

Irradiated Fuel Samples

The fabrication of five irradiated fuel samples at various stages of exposure has been completed. The samples were cut from one rod of a Pu-Al cluster which has been irradiated in the PRTR for 102 MWd. The samples are 4"

long and have been cut, along with samples for chemical and isotopic analysis from various positions along the rod. The samples were weighed and then clad in Zircaloy. They were again weighed after cladding to give the weight of the Zircaloy. The samples are stored awaiting measurement of their reactivity in the PRCF. The samples for chemical and isotopic analysis are also in storage awaiting chemical and isotopic analysis. The analysis will be used to more accurately determine the exposure and content of the samples.

The control unit of the pile oscillator which will be used in measuring the reactivity of these samples is being fabricated. This is the last component needed to complete the oscillator.

PuO₂UO₂ Graphite Lattice Studies

The paper summarizing the results of the 8-3/8" lattice experiment containing 0.9 w/o L_xPuO₂ has been accepted for presentation at the June meeting of the American Nuclear Society. Lattice parameter measurements on this same fuel in a 6-1/2" lattice are now being performed in the PCTR.

Calculations to help specify the PuO₂ concentration for the 29.6% Pu-240 fuel have continued. THERMOS was used to generate ηf values for single 0.5" diameter rods in graphite at moderator-fuel volume ratios of 17, 27, 73, and 150. Percent concentrations, PuO₂UO₂, of 1, 2, 3, and 10 were considered. THERMOS was also used to calculate ηf values for 19-rod clusters of 0.5" diameter rods, the core of which contained 2.0 w/o PuO₂ in depleted UO₂. The probabilities of escaping U-238 and Pu-240 resonance capture were calculated from Hellstrand's and Nichol's data, respectively.

Instrumentation and System Studies

Design and development work was continued on refinements to the scanning instrumentation to be incorporated in the PRTR fuel rupture monitoring system. The addition of the scanner, with improved signal to noise ratio characteristics over the present approach, should materially enhance system operation. Reed relays will form the heart of the new scanner. The chart recorders ordered for incorporation in the system are expected to be delivered soon.

General design considerations are being reviewed for a new radiation monitoring system for PRTR exhaust air measurements. An improved and considerably more reliable and stable system is desired for this problem.

The equations for a PRTR steam pressurizer were developed and assembled and a MIDAS program was written. The mathematical model will be complete

with determination of equation constants and function generator curves. The purpose of the simulation is to investigate the characteristics and the adequacy of the steam pressurizer for the PRTR.

Simulation studies have begun regarding hazards of operating the PRTR at proposed increases in power level.

Isotopic Analysis of PRTR Samples

Isotopic analyses were performed on 37 burnup samples of $\text{PuO}_2\text{-UO}_2$ PRTR-irradiated fuel elements: 17 uranium and 7 plutonium samples from fuel element number 5185, 9 uranium samples from fuel element number 5187, and 3 uranium and 1 plutonium samples from fuel element number 5213. The last 13 samples were duplicates or backup samples from previously analyzed fuel elements.

EBWR PROGRAM

Approach-to-Critical Loading Calculations

Calculations using multi-thermal energy groups in HFN have been performed for the series of EBWR-fueled approach-to-critical lattices. The Δk_{eff} 's between one and five thermal groups varies from 0.2% for the largest core (0.55" l.s.) to 3% for the smallest core (0.30" l.s.). Further study is under way to determine the effects on k_{eff} when using varying numbers of groups for the 0.80" lattice.

PHOENIX FUEL PROGRAM

$\text{U}^{235}\text{-Al}$ Polyethylene Experiments in the PCTR

In the course of analyzing the $\text{U}^{235}\text{-Al}$ polyethylene system, two interesting aspects of the experimental thin plate, tubular can geometries have been observed.

The first area of interest concerns the relatively large void fraction and its effect on the multiplication. While the reactivity of an infinite homogeneous system is dependent only on the relative concentrations of the materials present, the same is not true for an infinite heterogeneous system. In the heterogeneous system spatial flux depressions do depend on the absolute value of regional absorber concentrations. Proper account of this must be taken in the mathematical model. In the specific experiment now being analyzed the total effect of treating all voids as null regions--rather than smearing all voids--resulted in a reduction of the calculated k_{eff} from 1.0344 to 1.0120.

Another interesting phenomenon in the analysis concerns the angular import-

ance of neutrons incident on a thin plate. Transport calculations using from 3 to 31 double angles (and smeared voids) have been carried out and the results are tabulated below:

<u>Number of Double Angles</u>	<u>Calculated Reactivity</u>
3	1.05889
5	1.04505
7	1.03986
13	1.03444
16	1.03341
31	1.03173

Neutrons which just graze the surface and thus have a long track length within the fuel disk are quite important and a large number of angles is required. All of the transport calculations tabulated had the angles evenly spaced. Current work is aimed at producing the best answer with the least number of angles to be used. Both slab and cylindrical geometry can be subject to this "geometrical resonance" effect if the thickness of the region and the mean free path in the region combine to give narrow solid angles of high importance.

Pu-Be Studies

Nordheim's method for extending the calculation of the effect of grain size on resonance absorption in BeO-PuO₂ systems to considerably greater concentrations of absorber grains in the moderating medium is now operative. The previous method was limited to roughly 10 v/o of absorber grains in the moderator. The ZUT code is used in the new method with a modified Dancoff factor which accounts for increasing grain concentration. Although originally of interest in BeO-PuO₂ systems, these calculational methods could also be of use for mixed oxide fuels.

Resonance parameter cards were prepared for use with ZUT. Parameters from the HRG library were used for Th-232, U-235, U-238, Pu-239, and Pu-240. Unfortunately, only multi-level data are available for the Pu-241 resonances.

Work on the analysis of the Livermore BeO-U²³⁵ critical experiments continued. Input data for RBU Monte Carlo calculations have been prepared, but the necessary computer time has not been made available yet.

Calculations for possible PCTR experiments using Pu-Al discs and beryllium metal discs have been initiated.

Pu-Al Polyethylene Experiment in the PCTR

Buffer cans containing U-Al disks and borated polyethylene disks were added

to the PCTR core to flatten and harden the flux. For the same purpose, J-slugs and driver rings were put into the graphite surrounding the stack of fuel cans. These changes reduced the spatial flux variation to less than $\pm 2\%$ over the 6" test region.

Void substitution and boron sensitivity measurements showed that about 30% too much boron was present in the original loading. The core has been removed, and the fuel cans are now being reloaded with less boron.

Pu-Value in H_x-Pu Burners

A brief study of fuel cycle costs in H_x-Pu burners was carried out. Pu values were determined for some typical PWR's in the range of 50 to 200 MW. It would appear that Pu fissile/U-235 value ratios in excess of unity are obtainable for an interesting range of fuel fabrication costs.

FAST REACTOR STUDIES

FFTF - Nuclear Design Status

During the last reporting period, two informal reports covering various neutronic aspects of the FFTF were completed and distributed. The first report deals primarily with neutron statics, i.e., critical size and reflector control worth. The second report covers a preliminary investigation of the kinetics properties of the FFTF.

Cross Section Compilations

In order to perform meaningful and rapid fast reactor neutronic calculations, complete microscopic cross section sets stored in a convenient form must be available. Both the 16-group YOM set and the new 26-group Russian set are now being recorded and prepared for rapid retrieval and use. A comparison of the YOM, Russian, and Hansen-Roach compilations will be made as soon as the data retrieval apparatus is completed.

HIGH TEMPERATURE REACTOR PHYSICS PROGRAM

Mockup Tests

The second run of the HTLTR mockup has been started. Prototype control and safety rod elements as well as modified heater elements are being tested. Initially the system will be heated to 700°C, then later to 1000°C. As the system reached 200°C on the initial heating up, the gas analysis system showed contaminant levels of 2500 ppm water, 200 ppm oxygen, and a small amount of CO₂ and no detectable CO. For the purposes of

this run a small gas cleanup system has been installed in the system to help in reducing these contaminant concentrations, as rapidly as possible, to levels more comparable with what is at present considered a tolerable level in the reactor, about 300 ppm total.

Material Tests

Work has started on setting up a system for testing materials in 1100°C nitrogen carrying various concentrations of water vapor. The first material to be tested will be the B₄C-graphite matrix material that is planned for use in the control rods, safety blades, and chopper collimator of the HTLTR. Calculations based on existing data on the B₄C-graphite reactions with water indicate that in the HTLTR the rate of these reactions would be tolerable. The purpose of the above tests is to reduce our uncertainties regarding the dependence of the reactions on time and water vapor pressure.

Reactor Calculations

Reactor calculations which provide data for safeguards analysis and fuel specifications are continuing. The reactor calculations have now been refined to consider discrete fuel elements in x-y geometry and to yield details of the neutron flux through a rectangular, transverse cross section of the reactor. Cell calculations were made to predict the temperature coefficient compensation that can be obtained by placing Gd₂O₃ in the reactor. Sensitivity of the calculated multiplication constant to different choices of cell boundaries was also checked.

Design Review

The design review has continued and comments have been submitted on the oscillator assemblies, chopper, and flux wire manipulator assemblies. Due to budgetary considerations many of the suggested changes have not been made to the design. Thus these changes will probably have to be incorporated after acceptance of the reactor. In addition, the first review of the detector specifications and heat-up, cool-down, and vacuum system acceptance tests has been completed and comments offered.

Investigation of the high temperature properties of B₄C, Gd₂O₃, and various other substances to be used for experimental investigations has been initiated.

Fuel Element Development

Work has started on developing methods of welding T.D.-nickel and on vibrapacking of UO₂ powder in metal and graphite cans. Sample welds of flat

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20 mil thick T.D.-nickel strips will be tested for tensile strength and corrosion resistance and will be examined metallographically. Methods of packing UO_2 are to be studied from the standpoint of obtaining a consistent, constant, and reproducible density of the fuel.

HTLTR Driver Fuel Design Calculations

Evaluation of driver fuel requirements from a reactor multiplication and flux pattern standpoint is continuing. Emphasis this past month has been on computing the effects of various configurations of 5.0 w/o driver elements.

FUEL CYCLE ANALYSIS

Fuel Utilization Studies

Several revisions have been made to the VESTA code to improve its usefulness. An alternative short output form has been added so that output from many cases can be examined with a minimum of effort. Another revision to the code enables the use of a variable time period. Heretofore, the future economy has been hypothesized as a series of five-year-step functions. This period length can now be changed. Calculations to determine the effect of the size of the step functions are being made. The code is also being revised so that the rate of introduction of breeder reactors into the economy can be calculated when no plutonium availability limitations on initial inventories are to be applied. The breeders can be fueled with U^{235} .

If plutonium availability limits the rate of breeder construction, plutonium should be more valuable than if its availability does not limit the construction rate. At the other extreme, if there is more plutonium available than both thermal and breeder reactors can efficiently use, the relative value of plutonium should be small. Data on optimum operation of thermal reactors under such conditions are being generated and will be used in VESTA to calculate the effect on future uranium utilization.

Linear Programming Studies

The evaluation of the conservation aspects of the future nuclear power industry must include analyses based on the assumption that the relevant decisions will be primarily based on economic rather than conservation factors. A useful technique for simulating a situation which has economic optima (best decisions) and physical restraints (conservation) is linear programming. Consequently, as part of our evaluations of the conservation problems of the nuclear power industry, we are preparing the linear pro-

gramming tools which will be needed. Machine codes, such as LP/90, are already available for solving linear programming problems. As a first step in using this code and learning all its capabilities, we have prepared a simple problem which consists of one reactor having a requirement for power and plutonium production and a choice of operating methods. The optimization that must be performed is to make the choice of operating methods which gives, in turn, minimum total cost or maximum profit. This problem has been solved by hand calculations and on the 7090, and checks have been obtained. It will now be used to explore the various features of the LP/90 code and to learn the use of an output analyzer routine which translates the problem solutions into forms suitable for report generation.

Density Optimization

Indications of an optimum density, less than full density, for plutonium enrichment of natural uranium in a tight packed water lattice have been obtained. Bred plutonium was recycled in a BWR initially fueled with uranium and the optimizer code was used to determine the optimum fuel density. A case recycling the bred plutonium once yields an optimum density of about 70% of theoretical for the plutonium enriched fuel instead of the slightly over 90% of theoretical density stipulated by the manufacturer for uranium fuel.

Plutonium Value Equation

Plutonium values resulting from a recent series of recycle computations in four reactor types (BWR, PWR, D₂O, and SGR) have been fitted by an equation using two reactor parameters and the Pu²⁴² fraction in the discharge fuel. Initially four models, based on previous experience, were chosen for examination. The four models investigated are listed in Table I along with the results of the least squares fit performed by the GEORGE code. Model 3 was chosen as the best fit of the data based on multiple correlation and various data as developed by the GEORGE code.

In these equations, SNF, SDPV, and % Pu²⁴² refer to the cell total macroscopic nonfuel cross section (flux weighted) per unit fuel volume, the cell averaged slowing power volume ratio (flux weighted), and the % Pu²⁴² in the discharged fuel, respectively.

SNF appears in the equation on the basis that in a reactor with a large SNF, plutonium should improve the thermal utilization significantly over that obtained with uranium. Thus this parameter would be expected to affect the base plutonium value but should have little effect on the Pu²⁴² concentration.

TABLE I
FUEL VALUE EQUATIONS

<u>Model</u>	<u>Coefficients</u>			
	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
1) $PuV^* = A + B \frac{SNF}{SDPV} + \frac{C}{SDPV} (\% Pu^{242})$	7.87	38.28	-.418	-
2) $PuV = A + BSNF + \frac{C}{SDPV} (\% Pu^{242})$	7.99	17.96	-.410	-
3) $PuV = A + B \frac{SNF}{SDPV} + (C + \frac{D}{SDPV})(\% Pu^{242})$	8.16	33.92	-.0944	-.256
4) $PuV = A + BSNF + (C + \frac{D}{SDPV})(\% Pu^{242})$	8.28	16.26	-.125	-.196

* Plutonium Value in \$/Gram Total Plutonium = "PuV"

B-20

ENC-8.5

SDPV, however, can be expected to affect both the base plutonium value and the variation with Pu^{242} since it has a strong effect on the spectrum and, therefore, the cross sections of all the plutonium isotopes.

Previous investigations along these same lines have shown that the Pu^{242} has a strong negative effect on the batch plutonium value. The values were calculated in the PUVE code using this assumption so that the resulting values are linear functions of the Pu^{242} fraction.

Table II shows the plutonium values for the four reactor types and the values of SNF, SDPV, and % Pu^{242} corresponding to these. Note that $1/\text{SDPV}$ is used since a large SDPV corresponds to a thermal (soft) spectrum and a smaller SDPV (near 1) corresponds to a harder, or less thermal, spectrum. Apparently, the increase in cross section with smaller SDPV's offsets any negative effects that may be incurred by Pu^{239} , Pu^{240} , and Pu^{241} due to spectral hardening. The effect on Pu^{242} is reversed and it exhibits a negative effect with spectrum hardening.

The Relative Value of Bred Fuels as Enrichment in Gas-Cooled Reactors

Plutonium and U^{233} relative bred fuel values have been calculated for pressurized water reactors using both thorium and natural uranium as fertile fuels. To complete this work it is necessary to assume that U^{233} can be isolated at zero cost from a mixture of U^{233} and U^{238} . This resulted in value ratios (value U^{233} /value plutonium) of 1.20 for the natural uranium systems, and a slightly higher ratio of 1.22 for systems using thorium as a fertile fuel.

The same type of calculation has been completed for a gas-cooled reactor design similar to the HTGR, described in GA-4706. The average graphite moderator temperature was 1200°K with 3/8-inch-diameter fuel channels. The coolant system used helium and the specific power of the fuel was 50 MW/M.T. Figure 1 shows the fuel cost as a function of assigned bred fuel credit for slightly enriched uranium, plutonium enrichment of natural uranium, and U^{233} enrichment of natural uranium--each with optimized lattice spacing. The U^{233} -natural case used special graphite coated fuel particles whereas the other systems used UO_2 and PuO_2 fuels. The additional cost incurred with fuel particles must be added to the system fuel cost which would detract from the U^{233} value.

The intersection of the plutonium-in-natural uranium curves with the slightly enriched uranium curve is termed "the indifference value for plutonium." To compare U^{233} with plutonium, the system fuel cost must be the same; thus, the value ratio becomes the ratio of assigned prices which yield the same fuel cost. For example, a fuel cost for comparison might

TABLE II

PLUTONIUM VALUES AND STATISTICAL FIT PARAMETERS

Reactor	Cycle Number	Actual Pu Value	Fitted Error*	% Pu ²⁴²	SNF	SNF/SDPV	1/SDPV
SGR	1	9.87	.54	4.54	.1270	.06454	.507
SGR	2	8.08	.22	11.06	.1270	.06454	.507
SGR	3	6.88	-.06	15.20	.1270	.06454	.507
SGR	4	5.79	-.57	17.79	.1270	.06454	.507
SGR	5	5.35	-.65	19.39	.1270	.06454	.507
BWR	1	8.42	-.10	4.26	.0724	.04040	.558
BWR	2	7.11	.06	10.45	.0724	.04040	.558
BWR	3	6.22	.18	14.69	.0724	.04040	.558
BWR	4	5.63	.24	17.42	.0724	.04040	.558
BWR	5	5.25	.28	19.20	.0724	.04040	.558
PWR	1	8.06	.36	4.65	.0344	.02412	.701
PWR	2	6.34	.17	10.24	.0344	.02412	.701
PWR	3	5.39	.05	13.26	.0344	.02412	.701
PWR	4	4.76	-.09	15.04	.0344	.02412	.701
PWR	5	4.42	-.15	16.09	.0344	.02412	.701
D ₂ O	1	6.54	-.73	5.52	.0134	.00515	.384
D ₂ O	2	5.39	-.30	13.71	.0134	.00515	.384
D ₂ O	3	4.47	-.15	19.24	.0134	.00515	.384
D ₂ O	4	4.08	.19	23.03	.0134	.00515	.384
D ₂ O	5	3.84	.48	25.78	.0134	.00515	.384

*Results using Model 3

BRED FUEL INDIFFERENCE VALUES FOR GAS COOLED REACTORS USING THORIUM

FIGURE 1

be made at 1.0 mill where the assigned plutonium price is \$10.90/gm, and the assigned U^{233} price is \$14.50 per gram. Thus, the value ratio is $(14.50/10.90) = 1.34$. Value ratios determined at other fuel costs result in the same ratio. This ratio is higher than for the PWR and the difference is probably due to an increase in the alpha (α_c/α_f) for Pu^{239} which results from the higher neutron temperature in gas-cooled reactors ($\alpha_{PWR}^{239} = 0.54$; $\alpha_{HTGR}^{239} = 0.59$). High temperatures have very little effect on U^{233} fuels.

Figure 2 shows the same enrichment materials but using thorium as a fertile fuel. The value ratio determined from these curves was 1.47. The reason for a higher value is that the U^{233} -thorium system has no fission taking place at an alpha of 0.59 whereas the U^{233} -natural uranium has about one-half of the fission in bred plutonium.

MONTE CARLO Calculations

Last month two MONTE CARLO calculations were performed to check the code against experiments. The results were not satisfactory, so the closer packed lattice (water-to-metal ratio of 1.0) was studied in greater detail this month. The result reported last month was extended to a one-hour calculation, and seven variations were also computed. The results are presented on Table III along with the experimental results of Kouts and Sher (BNL-486). All calculations are for identical concentrations, cross sections, and geometry with variations in importance weighting and particle selection. Since RBU MONTE CARLO uses a pseudo random number routine, calculations are reproducible and two identical problems will yield identical results. Any minor change such as omitting the first random particle or using the first particle twice will cause a completely independent solution to the problem. RBU MONTE CARLO also provides importance weighting in space (zone weights) and in energy (group weights) to permit more detailed study of pertinent parts of the problem. If a particle representing one neutron enters a zone or group having a weight of 5.0 from a zone or group with a weight of 1.0, it will be treated as five particles representing 0.2 neutrons each. If a particle representing 0.2 neutrons attempts to enter a zone or group having a weight of 1.0 from a zone or group with a weight of 5.0, it is subject to a random selection with probability of survival of 0.2; and, if it survives, it will be treated as one particle representing 1.0 neutrons. Thus, variations in importance weighting in two otherwise identical problems will also produce independent solutions. The variations in the results in Table III raise some questions regarding the validity of importance weighting, so some of these calculations will be continued for a longer time. The low value of the fast effect indicates (and inspection confirms) that the high energy cross section, and/or nu for

BRED FUEL INDIFFERENCE VALUES FOR GAS-COOLED REACTORS USING NATURAL URANIUM

FIGURE 2

B-25

BNMC-8 3

TABLE IIIMONTE CARLO CALCULATIONS FOR WATER-TO-METAL RATIO OF 1.0

<u>Run Characteristics*</u>	<u>Fast Effect</u>	<u>Resonance Escape</u>	<u>Thermal Utilization</u>	<u>k Infinity</u>
Experiment (BNL-486)	1.099	.72 ± .027	.946	1.119
Broad group and zone weights	1.061	.720	.943	1.032
Broad group and zone weights	1.061	.723	.944	1.041
Broad group and fast weights	1.079	.708	.944	1.023
Narrow resonance, fast, and zone weights	1.072	.725	.942	1.053
Narrow resonance and zone weights	1.062	.714	.947	1.022
Zone weights only	1.055	.706	.942	.998
Uniform weights	1.081	.705	.942	1.016
Uniform weights	1.084	.702	.942	1.013

* All zones and groups are weighted 1.0 except as noted.

Broad group weights: .007 to 4 eV 5.
4 eV to 79 eV 20.
79 eV to 961 eV 5.

Fast group weights: above 0.5 MeV 5.

Narrow resonance weights:
5.7 to 6.5 eV 20.
6.5 to 7.3 eV 400.
20 to 23 eV 200.
33 to 37 eV 100.
37 to 42 eV 20.
61 to 70 eV 50.
101 to 115 eV 25.

Zone weights: Fuel 5.

U^{238} is low in the RBU library.

Code Development

The resonance integral input for MELEAGER-ALTHAEA was further subdivided to provide an improved calculation of the fast effect. The lower integral provides for epithermal reactions with neutrons with energy from 3.68kT to 0.68256 eV. The upper integral provides for reactions with neutrons with energy from 0.68256 eV to 200,000 eV, while the fast integral provides for reactions with neutrons above 200,000 eV. The fast flux is obtained from an input value of epsilon from JASON which is based on the fast effect in U^{238} only. Reaction rates of the other isotopes may then be calculated using the fast flux and the fast integrals.

(n,2n) Cross Section Investigation

A literature search for all available experimental (n,2n) cross sections was made so that the (n,2n) fast effect can be included in the MELEAGER burnup code for all fissionable and fertile isotopes currently being examined as well as D₂O and beryllium.

Because of the lack of experimental data, it will be necessary to use theoretically calculated values for most of the desired cross sections. The recent work of Pearlstein⁽¹⁾ is being used, and an empirical method for quickly extending his results to those isotopes not included has been developed. The detailed results will be reported in the future.

Space Dependent Kinetics

A paper, Space Dependence of the Reactor Noise Spectral Density Function, based upon earlier work of the author, J. R. Sheff, was completed for presentation at the Neutron Dynamics and Control Symposium, April 5-7, Tucson, Arizona.

(1) S. Pearlstein, Analysis of (n,2n) Cross Sections for Nuclei of Mass A > 30, BNL-897, 1964.

NEUTRON FLUX MONITOR PROGRAM

The two regenerating neutron flux detectors under irradiation at K Reactor continue to provide data, although it is now intermittent. Attempts to obtain valid lifetime measurements are made difficult by progressively degraded voltage saturation characteristics. Two more chambers designed to eliminate this difficulty are planned for early fabrication.

Another series of cable irradiation tests suggest promising methods of reducing cable background noise currents which have so far prevented successful demonstration of the boron-11 neutron flux detector. The use of twin-wire (twin-axial) essentially eliminated the transient signals associated with the buildup and decay of various isotopes and significantly reduced the magnitude of the net current between the leads. It now appears that unwanted "background" currents can be reduced to about 5×10^{-10} A, an acceptable value for the boron-11 detector. In addition, an irradiation test was performed using two cables which contained MgO and Al_2O_3 insulation in a titanium tube. Results of this test were consistent with earlier experiments in that in both cases excessive background currents were generated by aluminum containing materials. The superiority of MgO insulation in a titanium tube was clearly demonstrated. Based on these results, development of a new detector design employing parallel iron plates was accelerated. The assembly will be encased in a quartz container and the boron-11 will be placed within the iron detector assembly. The twin cable balancing technique will be employed to reduce background currents.

Microwave Detectors

In preparation for conducting "cross-coupling" microwave experiments at KW Reactor, two experimental glow-discharge (gas) tubes of small diameter were fabricated. These will be used to determine optimum mode of operation. Difficulties experienced with pressure changes in the wax-sealed tubes led to consideration of another technique employing extensive out-gassing and an epoxy sealant. Units fabricated with the latter method are being tested in the vacuum loop, and if successful, an in-core test at KW Reactor will be conducted when the facility is next available.

NONDESTRUCTIVE TESTING RESEARCH PROGRAM

Electromagnetic Testing

The development of the tubing cross section display device for use with the multiparameter tester is nearing completion. Further adjustments

have been made on the multiparameter tester, and now good separation between artificial flaws on the outer and inner walls of the tubing is obtained. However, an undesired variation in the displayed flaw depth is caused by the variation in hole diameter of the artificial flaws. This effect on displayed flaw depth has been confirmed by measurements on both flat plates and a tubular section. This condition is believed to be due mainly to the operation of the multiparameter tester in a range beyond its linearity limitations. In addition, some of this undesired effect may be due to holes of different diameters producing similar test signals for two different test frequencies.

Fundamental Ultrasonic Studies

As discussed in the previous monthly report, experimental attenuation data were fitted to attenuation curves having a smaller gradient at higher frequencies. This was being done to determine the influence of lowering high frequency attenuation when making pulse shape calculations. Good representations of the experimental attenuation data were obtained with calculated curves having the desired high frequency behavior.

The calculated pulses, which had previously been compared to the measured results, were recalculated using the new attenuation curves. Some improvements were obtained in the shapes and amplitudes of the calculated signals. One calculated pulse amplitude agreed exactly with the measured amplitude.

The computer program which describes the wave propagation model is being formalized and rewritten in FORTRAN-IV. Studies are also being conducted to determine the net effects of phase velocity changes due to attenuation.

Heat Transfer Testing

An improved version of the sinusoidal thermal wave transducer which holds the stainless steel foil in tension has been fabricated. The foil was held reasonably flat over a $\pm 20^{\circ}\text{C}$ temperature range in initial tests. Thermal expansion had caused buckling of the foil in a previous model of the transducer. Although performance of the new experimental transducer was satisfactory over this 40°C temperature range, an even larger temperature range is desirable. Nilvar foil which undergoes negligible thermal expansion over a 100°C temperature range has been ordered.

First attempts to use a transducer for measuring the thermal surface impedance of a steel sample were not successful. Phase shifts expected on the basis of the theory were not detected. A resistance-capacitance

analog simulator was used to check the theory, and the expected phase shifts were found to be correct. Measurements and theoretical analyses are under way to determine the nature of all parameters of the transducer in an attempt to compensate for this loss of phase shift.

Quantum Physics Devices

The piezoelectric semiconductor, CdS, has been used to produce a diffusion layer ultrasonic transducer. A 10 x 10 x 1 mm, single crystal of CdS having optically polished faces oriented perpendicular to the optic axis and doped with indium to produce an n-type semiconductor having a bulk resistivity of 0.1-1.0 ohm-cm, was coated on one face with copper by vacuum plating. The copper was diffused into the crystal by heating it in a helium atmosphere in the diffusion furnace at 400°C for 30 minutes.

The resulting transducer had a fundamental frequency of 25 Mc corresponding to an effective diffusion layer approximately 0.015 cm thick. The response curve of this transducer shows a band width of approximately 70 percent of the fundamental frequency. The response curve is irregular with the irregularities being separated by a frequency which corresponds to the half wavelength frequency of the crystal as a whole (approximately 1.8 Mc). The observed band width and response curve are typical of this type of device. Since there is no acoustical mis-match at the diffusion layer boundary, the device is not mechanically resonant at its fundamental frequency. Because of the acoustical match at the diffusion layer boundary, this device may prove particularly useful as a "spike" pulse device since it does not tend to "ring" as conventional mechanically resonant devices do.

Electrostatic Transducer

Pulse-echo techniques using the electrostatic transducer were investigated, and excellent results were obtained. In the pulse-echo technique only one transducer is used to generate and detect the ultrasound. To enable the electrostatic transducer to both generate and receive ultrasound, it is necessary to have a bias voltage across the dielectric at all times except during the transmit pulse. A circuit was developed which satisfied the above conditions, and the practicality of using the electrostatic transducer with pulse-echo techniques was shown. The efficiency of the pulse-echo transducer appeared to be as good or better than the efficiency of the two transducer method.

A high temperature holder was evaluated by using the pulse-echo technique

and isolating the holder from the associated electronics with 3-ft. lengths of unshielded, high temperature wire. Noise was greatly reduced as compared with the two transducer method. This experiment indicates that the device is now able to function in a furnace with the controls being remote. Pulse shape and velocity changes were observed in a brass rod as the rod was heated to 500°C by heating the electrode.

The effect of transmit pulse shape on the transducer response was also investigated. The rise time of the transmit pulse was held constant while the decay time was varied from a slow exponential decay to a rapid decay with accompanying oscillations. These oscillations which appeared in the transmit pulse were accurately reproduced in the receive pulse; thus it is indicated that the electrostatic transducer may be driven with a continuous sinusoidal wave. It was also noted that the frequency of oscillation could be varied by changing the resonant frequencies of the transmit circuitry. Tests will be performed to determine the frequency range of this transducer.

Nondestructive Testing of Isotope Heat Sources

Mechanical, ultrasonic, and electronic equipment for the automated test system for use in inspecting the wall integrity of isotope heat sources is being assembled. The automated mechanical system has been fabricated and is currently undergoing evaluation tests. With this system, complete ultrasonic scanning of the isotope heat source of various sizes and shapes may be obtained either automatically or manually.

A versatile transistorized test instrument is being developed which will extract the ultrasonic information from the detected ultrasonic pulses and provide a permanent record of the data. At present, circuits will be provided to measure wall thickness, cold worked areas of the cladding, and possible defects oriented parallel to the heat source surface. Circuits which will use ultrasonic shear waves to detect defects positioned in any orientation will be added at a later date.

Plutonium Properties Measurement

The ultrastable eddy current instrument is being modified for use in eddy current testing of plutonium. This instrument has been used with success to detect hydrogen in zirconium and the onset of fatigue in certain metals. Main emphasis at present is to raise the test frequency from 200 Kc to 3 Mc. Hastelloy-C, having a conductivity approximately equal to Pu, is being conveniently used for laboratory development of the eddy current instrumentation.

A versatile mechanical system is being designed which will permit evaluation of all the proposed ultrasonic and eddy current tests as applied to the inspection of plutonium.

DETECTION OF COOLANT IMPURITIES AND TEMPERATURE MEASUREMENT PROGRAM

Theoretical investigations of using infrared techniques for measuring the moisture content of gas coolants demonstrated the definite usefulness of the approach and an appropriation request was submitted for the procurement of an infrared analyzer. In addition, a small laboratory test loop was designed for use with the analyzer.

Design work was continued on a microwave cavity to be employed in studies of frequency variations with moisture content in gases, using closely controlled moisture levels.

BIOLOGY AND MEDICINE - 06 PROGRAM

Atmospheric Physics

Feasibility of using xenon-133 as an atmospheric tracer for routine field operation has been demonstrated. The initial test of the system made in January was partially successful. The activity in the samples collected then were significantly greater than the measurement background, but icing at the sample inlet reduced the flow throughout their operation. The varying flow rate which was not continuously monitored made interpretation of the data impossible.

A second test was completed on the night of February 28 in a stable meteorological regime. Zinc sulfide and xenon were released simultaneously near the ground for a period of 30 minutes. Twelve samplers that had been modified to eliminate icing were positioned on arcs 200 and 1600 meters downwind. Significant samples were obtained at all positions within the plume and the flow rate remained constant throughout the operation. In addition, the time required for startup was reduced two hours from that of the first test. This success indicates that with minor modification, dual tracer field tests with 24 xenon samplers could be conducted routinely.

Data collected in these experiments are to be analyzed to evaluate magnitude and rate of deposition. At the present time, no satisfactory technique exists for measuring deposition from the atmosphere on heterogeneous surfaces. Direct comparisons of exposures of the relatively inert gas, xenon, and other tracers are a new approach that eliminates many of the complexities of direct surface measurements.

Washout coefficients for gaseous iodine have been calculated from Best's rain spectra and the Nusselt diffusion numbers measured by Kinzer and Gunn. These are very close to those previously calculated by Chamberlain for the same spectra and somewhat different Nusselt numbers, which are in current use.

The calculations were then made using the rain spectra measured at Sunny-side, Hanford, and in India, and these washout coefficients were much smaller. Best's fitted spectra are unrealistic in the small drop region, while the Washington spectra may have a minimum number of small drops because of evaporation. The spectral difference shows up more in gaseous scavenging than in the particulate case, because small droplets are effective for scavenging gases, but not for particulates. The calculations show that the small-drop region of the rain spectrum is critical in gaseous scavenging, and currently accepted washout coefficients for iodine are erroneous.

Successive 20-minute stack samples have shown the REDOX plume to vary substantially, and it is now necessary to discard washout tests in which the stack samples are not coincident with the snowfalls. This leaves five measured coefficients in snowfalls, two of them in agglomerated spatial dendritic snow, and three in columnar snow. Of the latter, two of the coefficients, though measured in different storms, show the same coefficient for the same snowfall rate. The third shows the coefficient to increase linearly with snowfall rate. The coefficients for the spatial dendrites, however, are much lower than for the columns in the same snowfall rates. This is attributed to the smaller fall velocity and longer time in the plume of the columnar-type of crystal.

Radiological Physics

An approximate relationship was discovered between the radiation doses on the two sides of a plane boundary between two different media, one of which contains the radioactivity. If the average ranges in the two media and the activity density of the radiation are known, it is possible to solve for the two dose rates by using this new relationship. The predictions of this theory are in good agreement with experimental results.

The use of exoelectron emission, a relatively unknown phenomenon related to thermoluminescence, is being studied. It appears that it will have applications in dosimetry studies. Experiments have been planned to study these possible applications.

A vacuum manifold has been built which allows experiments with the helium ion separator to be carried on while the Van de Graaff accelerator is used

for other experiments.

A "thermal shorting switch" was developed for the fast calorimeter system. It consists of a copper bar which can be rotated to make a thermal connection between the two cells of the calorimeter after it has been opened or otherwise thermally disturbed. With this switch the time required to come to equilibrium is 3 to 4 hours rather than the 20 to 30 hours required without the switch.

In order to get the maximum X-ray dose from the pulsed X-ray machine more knowledge is required about the range of impedances it will accept. Experiments to determine this range are being made.

The problem of bubbles forming in the molding of our tissue equivalent plastic has been overcome by using a suggestion of Dr. Shonka of St. Procopius College to heat the mold from one end to another rather than heating each part evenly. This causes the bubbles to move along with the heat. A tissue equivalent liner for a proportional counter was constructed and a spherical proportional counter with a shaped electrical field is now under construction with this material.

The technetium metabolism study with the University of Washington is still continuing. Nine subjects were given Tc^{95m} , 6 by injection and 3 by ingestion. The retention and excretion patterns are in reasonable agreement with the values used by the ICRP, with 95% of the initial dose being excreted with a 2-day half-life and 1 or 2% being excreted with a half-life of more than 20 days. Whole body counts are being used to determine these half-lives, and they will be continued at 2-week intervals for another month.

One of the subjects in the Tc^{95m} experiment was counted on the plutonium counter at our laboratory. The Tc 17 keV X-ray was easily detected and this demonstrated that this isotope can be used in an inhalation experiment to calibrate for plutonium in the lung.

A method of making thin layers of metal salts for use as Ross filters has been developed. By dissolving the salt in an aqueous solution and then adding gelatine a thin homogeneous film can be made which makes an excellent filter. Ross filters contain elements which have a "K jump" in absorption coefficient just above and below the X-ray of interest, and they provide increased resolving power for the X-ray. Experiments are being made to determine the effectiveness of these filters in determining the 32 keV X-ray emitted from in vivo Cs^{137} .

Instrumentation and System Studies

The detection and transmitting portion of the blood pressure measurement channel was completed for use in the animal physiological function and radionuclide uptake telemetry system. The temperature, respiration rate, and thyroid uptake measurement channels are also completed in bench-model form. The various signal recovery circuits for the receiving end of the telemetry system were designed and are being fabricated. Blood pressure circuitry includes a selective bandpass filter, a trigger circuit to produce a square wave output at the sub-carrier frequency, and converter to provide an analog signal to be read out with a rate circuit and a recorder. Except for probable transmitter changes and simplifications, the complete system, now in bench-model form, is ready for translation into the final, miniaturized form.

Excellent developmental progress was achieved on the experimental instrument that will measure raindrop charge and velocity. An aluminum cylinder (12-in. long and 6-in. diameter) was prepared with internal provisions for mounting various inductors concentric to the cylinder. The inductors are brass tubes and are insulated from the cylinder. The electrometer, using field effect transistors, is connected to the inductors. A charged raindrop passing through the inductors induces a measurable charge on the electrometer, whose output is fed to an operational amplifier. This arrangement has been used in the laboratory to measure a charge on distilled water drops of about 10^{-11} coulombs. Further tests indicated that the minimum detectable charge per drop was about 2×10^{-14} coulombs, as limited by 60 Hz noise. Reduction of the noise would enhance the sensitivity by a factor of 10. The experimental instrument was determined to have a linear response over at least three decades.

Fabrication of a dual-cylinder ionization chamber was partly completed for use in a rodent whole body counting system. Plans were established for testing the detector as soon as it is completed.

The experimental fish trough monitor, to be used in measuring dose rates to fish, was calibrated and delivered to the trough location at the 100-KE Aquatic Biology Facility. The scintillation detector and solid state circuitry instrument measures dose rates from 0.1 mR/hr to 1 R/hr with an accuracy of about $\pm 20\%$ over the gamma energy range of interest. Operation appears to be fully successful. Further work on the system will include the addition of more lead shielding to reduce the high background levels being encountered.

Successful laboratory operation of an experimental bench-model algae radio-

nuclide uptake monitor is being achieved through the use of background subtraction circuits to improve the signal to noise ratio. Samples of algae are now being obtained for detailed tests, and if these are successful, the detectors and circuitry will be completed in final form.

To improve light transmission between scintillation detectors and multiplier phototubes, several silicone resins are being investigated. One of particular interest is Dow-Corning "Sylgard 184." It is optically clear and cures at room temperature without air bubbles. It demonstrates exceptional adhesion to glass. Several "models" are being prepared to permit comparison with similar-size Lucite light pipes, as normally used in scintillation work.

Development of low-noise, charge-sensitive, solid state preamplifiers continued. Regulated power supplies were completed to supply power to the developed circuits and fabrication was started on the complete amplifier. Insulated gate field effect transistors are being used at the input. The amplifier will be directly useful for experimental work with both solid state diode detectors and scintillation detectors.

The variance and covariance analyses for the Meteorology wind turbulence program were completed and the preliminary analog program for the cross correlation analysis was completed. The problem board has yet to be patched. Further analysis work is required when Atmospheric Physics personnel provide additional data.

Modifications were made to the biological system radioisotope model for standard man and analyses were made to determine the effects of Tl^{132} and I^{132} in the various portions of the body when these parent-daughter isotopes were injected into the body and fed through the stomach. The modifications included changing the recorder inputs and connecting the effects of the liver to the small intestine. When the other organs simulation was changed to liver, the effect on the small intestine was not added. Next to be analyzed is I^{131} injection in the parent portion of the model. Modifications will be made to the feeding circuits so that three non-periodic feedings per day can be given.

WASHINGTON DESIGNATED PROGRAM

Isotopic Analysis Program

Isotopic analyses were provided on program samples received during the month.

Studies of the operation and performance of the newly installed ion source

and ceramic vacuum-lock sample changer continued. The sample changer appears to operate satisfactorily, except for the mechanism for making electrical contact to the sample filaments. Although several air leaks were found to exist in the source parts, a pressure of approximately 1×10^{-7} torr can be maintained in the source region and 6×10^{-8} torr in the analyzer.

The ion optical properties of the source are not clearly understood at present. The ion transmission is estimated to be about an order of magnitude below the expected 75 to 100%.

EXPERIMENTAL REACTOR PHYSICS FACILITIES

PRCF

Experimentation continued on the Pu-Al D₂O reactor system. Following reactivity adjustment to a critical core at a moderator level of 103.25 inches, the flux instruments were calibrated relative to reactor power. Moderator sensitivity and worth measurements were obtained from 99.25 to 106.25 inches during the reactivity adjustment. Preliminary safety mechanism worths were obtained from the differential galvanometer and log N recorder techniques during rod drop test. A safety rod and control rod were found to be worth 51 mk and 2.86 mk, respectively. The last 62% of the control rod was calibrated and found to be worth 1.35 mk.

Subcritical horizontal and vertical flux traverses were completed using bare and cadmium covered BF₃ chambers. The experiment was conducted under Special Condition No. 14 because of the significant reactivity effect of the cadmium cover.

Two new linear chambers were installed in the core increasing the flux sensitivity including the galvanometer by a factor of 9.

Project CAH-153, H₂O Irradiated Fuel Handling Capability, is essentially complete. All hardware and special equipment with exception of the cask has been received. Acceptance testing will be conducted when the cask is received. Work on Project CAH-119 is continuing. PRCF heat pump and ventilation system installation are in progress.

PCTR

Operation of the PCTR continued routinely during the month. The first part of the Phoenix fuel experiment was completed. The experiment was interrupted to make a change in the number of borated polyethylene disks used in the Pu-Al fuel assemblies. The 19 element cluster PuO₂-UO₂ fuel elements

in a 6-1/2 inch pitch graphite lattice was started during the month.

TTR

The TTR was operated during the month on a one-night-a-week basis for the University of Washington Graduate Center.

Development Items

A dual clock is being modified to control the printout of the digital voltmeter and trigger the oscilloscope. Increased timing accuracy and control simplification will result.

A counting module for reactor noise analysis experiments was designed and built. The module consists of two channels, 7 decades per channel, and each channel indicates the noise level by counting the pulses from an analog to digital converter.

A PCTR graphite core was designed for the Pu-Al, thoria experiment. Pu-Al and thoria elements will be in alternate channels.

The demonstration pile modulator was completed. Its use will result in improved reactor kinetics data. Remote control now allows the rotation speed to be changed from the reactor control room.

COMPUTER FACILITIES

Four rolls of punched paper tape were pre-edited on the G.E. 412 computer and several tape punch errors were found and corrected. According to PRTR personnel, the time required for subsequent routine processing by EDPO was cut nearly in half. These tapes represented nearly a full month's PRTR data.

Analog computer utilization was as follows:

<u>EASE 1132</u>	<u>EASE 2133</u>	
230	212	Hours Up Time
0	16	Hours Scheduled Down Time
<u>2</u>	<u>4</u>	Hours Unscheduled Down Time
232	232	Hours Total

The up time on both computers includes 80 hours of swing shift usage.

Problems considered during the month were:

1. Meteorology Study.
2. Biological Distribution of Radioisotopes.
3. N Reactor Power Conversion Study.
4. PRTR Hazards Study.
5. ANN Optimization.
6. Nuclear Decay Chain Optimization.

An example nuclear decay-chain problem was used to demonstrate the capability of the iterative analog computer for NRD and IPD Reactor Physics groups. The problem formulated by the physicists was to determine the optimum reactor flux, temperature, and epithermal ratio for optimum recovery of certain radioisotope decay products over a 15-year period. The computer simulation pointed out the need for additional model development by the reactor physicists.

A simplified model of a reactor was set up on the 1132 analog computer for demonstration purposes for the Northwest Simulation Council meeting.

The winter quarter of the Electronic Computer Applications class was completed. Certificates were prepared to issue to those who completed the course.

CUSTOMER WORK

Weather Forecasting and Meteorological Services

Consultation was provided to 1) N-Reactor Electrical Systems on the frequency of severe cold waves, and 2) Radiation Monitoring on the measurement of water vapor in a small air stream.

A summary of the discharge and temperature of the Columbia River at Priest Rapids for 1964 and a forecast of these conditions for 1965 was distributed. The peak flow rate is expected to be about the same as last year and the river temperatures during July-September are expected to be considerably higher than last year.

Weather Summary

<u>Type of Forecast</u>	<u>Number Made</u>	<u>% Reliability</u>
8-Hour Production	93	87.7
24-Hour General	62	86.0
Special	115	86.1

March temperatures averaged below normal with a warm first half followed by very unseasonably cold temperatures during most of the second half. Precipitation was very light with the only measurable amount occurring as snow on the 26th. There were strong NE winds on the 11th and 12th, but speeds for the month as a whole averaged a little below normal.

Mass Spectrometry

Isotopic analyses were provided on 11 uranium samples in support of Purex U^{233} production studies and one uranium sample for Metallurgy Development Operation (U^{235} content of scrap U for safety reasons).

A series of 33 analyses on uranium isotopic standard samples was made to study the performance and accuracy of the heavy element mass spectrometer. A bias of about + 0.8% was detected in the measurement of U^{235}/U^{238} ratio and the precision of individual analyses is $\pm 0.4\%$ at the 95% confidence level for isotopic ratios greater than 0.01.

The over-all operation of the mass spectrometer "B" reached a first in one respect. Limits of measurement of trace amounts of U^{233} in 4 uranium samples were placed at ± 0.3 parts per million at the 95% confidence limit. The samples were analyzed as a service to CPD.

Instrumentation and System Studies

The initial investigation of methods of measuring PuO_2 grain sizes in PuO_2-UO_2 fuel element samples was essentially completed. Results suggested the use of an electron "microprobe" instrument in which a fine beam of electrons is impinged on the specimen. The characteristic X-rays are then detected and with a suitable scanner, a two-dimensional "picture" is obtained of distribution of the grains.

Fabrication and initial testing were completed on a scintillation detector and solid state circuitry gamma monitor that is to be used on moving ground vehicles for general survey work by Environmental Monitoring of the Radiation Protection Department. The instrument is now undergoing general field testing.

Design work was initiated on modified gamma scintillation well-logging probe and on a core-sample radionuclide measurement instrument for Geochemical and Geophysical Research of Chemical Effluent Technology.

Evaluation tests were made of dry reed relays for switching low-level signals for the Containment Systems Experiments. Test results show the reed relays to be too noisy for this application. Several types of mercury-

wetted contact relay will next be evaluated. Detailed circuit diagrams were completed of the computer interfaces for the multiplexer drive logic and multiple multi-channel analyzer. The multiplexer drive logic selects input signals, provides a buffer register for the multiplexer, and generates timing signals for an analog-to-digital converter. The analyzer interface permits transfer of data from four multi-channel pulse-height analyzers into the computer.

The 6144-channel analyzer paper tape punch was found to be faulty and was repaired.

The radio-telemetry network central station was modified to accept an extra data channel from the remote stations. This increases to eight the number of data points transmitted at each call-in.

The Containment Systems Experiment program is being further supported with development of a method of detecting crack propagation in stressed vessels. Considerable effort was made to obtain information on suitable sound detection transducers. Various types are being ordered for testing and evaluation. Those under consideration are piezoelectric crystal accelerometers, crystal microphones, piezoelectric ultrasonic transducers, moving coil stereo cartridges, and lithium-sulfate ultrasonic transducers.

Work continued on the fabrication of a data processor for use with the micropositioner readout portion of the creep data logger. The new data processor was designed to replace an older system which uses mechanical relays. Mercury-wetted relays and a simplified wiring layout in the new unit are expected to improve the reliability of the system. Fabrication is 90% complete.

The fabrication of a watt meter transducer assembly to be integrated with the creep system instrumentation at the KW facility is 80% complete. These transducers are for the purpose of measuring the power dissipated in each of the capsule heaters.

The Minneapolis-Honeywell data logger at KW has operated continuously since 1-18-65. During that time, it has correctly logged the data presented to it by several N Reactor tests.

Analysis of the vibrating capacitor stabilized current-to-frequency converter being developed for use with HTLTR instrumentation has shown that capacitor drift and thermal offset voltages are serious as long as the source impedance is high. If the source resistance is decreased or if shunt capacitance is increased, the converter amplifier demonstrates instability. A new mechanical chopper, which has a low thermal drift

and low offset voltage, is being obtained to replace the vibrating capacitor. Considerable operational improvement is expected. A bridged "T" feedback amplifier was developed for use as the chopper amplifier; it has a gain of about 1200 and a variable bandwidth at the frequencies of interest.

Optics

A formal report which describes equipment developed to traverse and measure distortion in reactor process tubes has been completed except for figures and final typing. Traverses have been run at C and B Areas with the new equipment, and a test calibration run was completed on a tube outside the reactor.

Optical lens systems are being automatically designed with a computer program which computes from existing requirements, such as glass material, aperture, diameter, etc., on optimum design for any given situation. The program is currently being used in evaluating candidate optical systems for the Purex crane periscopes.

Assistance to Chemical Research Operation is being provided the design and acquisition of optical components for a spectrophotometer. To date, two nondarkening lenses have been fabricated for this spectrophotometer. A periscope to view alignment of the fuel element on the annulus gauge bed is being fabricated for the 100-N fuel examination facility. The lens system will be evaluated and designed by the lens program.

During the four-week period covered by this report, the following work was performed:

1. Three Redox crane periscope heads were repaired.
2. Two cylindrical mirrors were aluminized.
3. One set of glass bearings was fabricated.
4. Two Purex high power nondarkening lenses were fabricated.
5. Two shutters were repaired.
6. One camera back was fabricated.
7. One metallograph was packaged for shipment back to the factory.
8. Twelve glass boxes were fabricated.
9. One borescope was repaired for PRTR.
10. Two nondarkening lenses were fabricated.
11. One lens mount for a film projector was fabricated.
12. Several mirrors were aluminized.
13. One light pipe was fabricated.
14. One special quartz cylinder was fabricated.

Physical Testing

An inspection program of major proportions has been completed on the ten steam generators at N Reactor. Tests included eddy current, hydrostatic, ultrasonic and fluorescent penetrant on the radioactive units. Data were provided for tube removal and confirmation of the general condition of the steam generators after 15 months of operation.

Ribbed zirconium tubes for Advance Fuels development were ultrasonically tested for laminations in the rib material and for rib to tube bonding. None of the tubes passed.

Hanford Specification 8070, "Ultrasonic Inspection of Welded and Seamless Tubing", was revised to include provisions for testing large grained tubing.

Fluorescent penetrant tests of N fuels end cap continues with a total of 6101 tested this month. One-hundred and thirty were rejected.

The first twelve inches of 700 EBWR fuel elements were ultrasonically inspected to determine if the vibration compaction method of loading had created cracks in the cladding near the hold-down clamp area. Some defective areas were found.

All quarter-inch diameter by .035 inch wall stainless steel tubing in Central Stores was eddy current tested to select the best possible noise free material for experimental PRTR fuel elements.

Work started on standardizing the 1004 eddy current tester for checking the graphite thermal shield tubing in the KE and KW Reactors. New probes and standards are being fabricated.

N Reactor turbine impeller blades were magnetic and fluorescent penetrant tested for cracking. The cracks occur on most blades. Further tests are being done in the field.

Some highlights of radiographic services performed during the past month were insuring weld integrity of PRTR rod end caps, and inspecting cermet fuel pin extrusions for the Fast Reactor Fuel Fabrication program and Experimental Boiling Water Reactor program. Saxton fuel rods were also radiographed to determine location and alignment of internal components. Five construction projects were serviced including the D-14 concentrator, TA-3 dissolver, 200 East Area hot trench piping, and PRTR corrosion loop.

Test Engineering Support

Work is continuing on the design of a nondestructive testing station for the waste solidification pots. Design is complete on the ultrasonic signal processing device which will connect the ultrasonic system to a G.E. 412 computer. The device will be made up of commercial, transistorized digital logic modules. The logic will supply the gating and conversion necessary to change the time-echo signal to a digital signal. Pulses generated in the logic system will drive the ultrasonic pulses. A four-transistor video amplifier with a gain of 1000 and a bandwidth of 20 Mc, was built and will be tested with the logic system in the near future.

Signals will be processed and transmitted over telephone lines to the 412 computer, thus providing a versatile readout. This will have the advantage of providing a more versatile readout while saving a typewriter and extra digital logic modules.

A new liquid level probe is being fabricated for installation in a solidified waste storage pot. This probe will permit electrical entrance through the pot head connector, but must be manually connected. If successful, the probe will be redesigned for remote operation.

A study is continuing to determine if eddy current methods can be used to accurately measure the thickness of plutonium deposited on a metallic substrate. An analysis of the problem has been made and a prototype instrument assembled. Materials for use as standards to substitute for plutonium and beryllium were obtained.

Fabrication of two thin wall tubing testers for Physical Testing has been completed with the exception of the amplifier installation. The scheduled delivery date for these commercial amplifiers is April 15.

INSTRUMENT EVALUATION

The eight hand and shoe contamination monitors, of which six are for alpha-beta-gamma measurements and two for alpha-only measurements, were received from the offsite fabricator and evaluation testing was initiated.

A number of minor changes required became immediately apparent and these are being incorporated on one instrument. The number of errors in wiring appears to be small and the principal difficulty is lack of power supply decoupling to the various circuit boards. One instrument is being modified at this time, and once it is performing properly, the other seven units will be changed accordingly.

Evaluation tests were initiated on a Tracerlab Corporation gross high-level alpha air monitor of the line-operated type. The evaluation is being conducted at the request of the Biology Department.

Instrument maintenance training classes were completed regarding the combined alpha-beta-gamma hand and shoe monitors received from an offsite fabricator several months ago. These units are now all in service in various Hanford locations and the general performance appears to be satisfactory.

A developed "palm size" alpha portable instrument, which employs a silicon diode surface barrier alpha detector and all solid state circuitry, was employed at 100-KE to monitor for alpha surface contamination on plastic bags containing irradiated thorium oxide. Excellent operation was achieved; the instrument could detect 500 dis/min of Pu^{239} in a 15 Rad/hr field (principally beta).

Circuit design was continued on a solid state field effect transistor circuit to be used with a 600 cm^3 ion chamber to replace the standard Hanford CP electrometer radiation survey instrument.

RS Paul

Manager

PHYSICS AND INSTRUMENTS DEPARTMENT

RS Paul:mcs

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CHEMISTRY DEPARTMENT
RESEARCH AND ENGINEERING

FISSIONABLE MATERIALS - O2 PROGRAM

IRRADIATION PROCESSES

Overheated N-Reactor Fuel Study

A target element irradiated to a gas volume ratio of 1.5 was heated inductively in a partially shielded quartz furnace to observe the rupture of the can and flow of the molten core. Colored movies recorded that near its melting point a slow flow of the core occurred and the flow increased in volume as the result of a second breach. About 13% of the core metal flowed before heating was stopped near 950 C. Penetration of the can was at each of two weld spots of the downstream supports. Since any internal pressure was relieved through the initial breach, the second penetration must be the result of diffusion or reaction between the core and can.

Preliminary tests with unirradiated target elements showed that the alloy melted and flowed early in the heating cycle. This suggested that heating by direct coupling to the element increased the rate of the destructive reactions and might have influenced what at least appeared to be an early rupture at a temperature lower than the 800 C indicated for the tests using radiant heating.

Two unirradiated elements, fabricated to duplicate the irradiated targets tested, were heated in the same manner used for earlier "cold" tests as well as the irradiated targets. The conditions and severity of destruction indicated that anodized or oxidized interior surfaces retarded reaction rates and in some cases prevented flow in the unirradiated elements. A third element with a bright etched rather than a dry blasted core surface did not react much differently than the above two elements.

Concentration of N-Reactor Decontamination Solutions

A short study of the physical properties of three simulated N-Reactor decontamination waste solutions during evaporation at reduced pressure was completed. Boiling point, quantity of solids, foaming tendency and fluidity were observed as the solutions were concentrated by factors up to 25 by evaporation at about five inches of mercury pressure.

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SEPARATIONS PROCESSES

Disposal to Ground

Low but detectable concentrations of Sr-90 have been noted in recent ground water samples from two of the three wells monitoring the Redox S-7 process condensate crib. The maximum concentration detected, 6×10^{-8} μc Sr-90/cc is about a factor of two below the recommended ground water limit of 1×10^{-7} μc /cc. The increasing trend in concentration resulted in a recommendation to the Chemical Processing Department that the replacement crib, constructed about a year ago, be activated.

Low-level analyses indicated the possible presence of radioactive I-129 at trace concentrations in the ground water beneath the Purex process condensate crib (A-10). The fission yield of this long-lived (1.6×10^7 years half-life) nuclide is quite low; however, the MPC_w (168 hr) is also relatively low, 4×10^{-6} μc /cc. A likely source of any I-129 that might not be released to the silver reactors during fuel dissolution is the Te-129 which has not completely decayed at this point in the process. Additional ground water and several process waste samples have been submitted for I-129 analysis.

Plutonium Aerosol Studies

The last of six plutonium rods was ignited at about 600 C in air flowing at 20 cm/sec. The resultant oxide particle size distribution was similar to the range of distributions obtained for earlier ignitions. In the series, air velocities between 3.3 and 50 cm/sec were investigated for ignition temperatures of 400 and 600 C. On the basis of sieve data, the resultant oxides are coarser than previously reported in the literature. On a weight basis, approximately 7% of the oxide is reported in the literature to be less than 74 microns in diameter; in the present study, only 2.2 to 3% of the oxide was found in this range. Corresponding numbers for particles less than 44 microns in diameter are approximately 3% and 0.8 to 2%, respectively.

The document, "A Review of Research in Plutonium Releases During Overheating and Fires," (HW-83668) was issued.

Thoria Dissolution Studies

The investigation of the excessive abrasion which can be caused by thoria particles suspended in a liquid has been virtually completed. Previous information had demonstrated that the potential existed for a severe "honing" action upon the dissolver floor by the jet streams from agitating devices (spargers or air lift circulators). The study

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has shown that the velocity of the impinging stream is by far the most important and controlling variable in regard to this mechanism. The results have also shown that previous recommendations to limit the air velocity from the air lift circulators in the Purex annular dissolvers to less than 50 ft/sec are consistent with minimizing this effect. An additional long-term run will be made in an effort to clarify the effect at relatively low velocities.

Pilot plant mock-up dissolver modifications were completed and a co-dissolution (simultaneous jacket and core) run made. The solvent for the dissolution was 12M HNO_3 -(0.55 to 0.83)M ANN-(0.076 to 0.114)M HF-0.003M $\text{Hg}(\text{NO}_3)_2$. The dissolver was operated with periodic nitric acid (12.2M) additions and reflux was routed to a hold tank (i.e., similar to the Purex 6-ton dissolution). About 50 percent of the charge was dissolved after boiling for three hours. A material balance after eight hours of dissolving indicated that 77% of the charge had dissolved. The system was held overnight at about 160 F. This permitted some thoria dissolution and introduced an unknown factor into the data after about 3 - 4 hours of dissolution.

Modifications are underway to include a centrifuge in the dissolver system.

Laboratory work is completed for a study to define the reaction rate of C-64 aluminum alloy with nitric acid as a function of acid concentration, nickel nitrate concentration, aluminum concentration, mercuric nitrate concentration, hydrogen fluoride concentration, and aluminum area to solution volume ratio.

Polonium-210 Recovery and Bismuth Recycle

Development studies on the processing of bismuth for polonium-210 recovery continued along two routes. The first involves dissolution of irradiated bismuth followed by solvent extraction recovery of Po-210 employing dibutyl carbitol and pyrometallurgical reduction of bismuth nitrate to the metal for target element refabrication. Exploratory pilot plant flowsheet and equipment development and evaluation of the solvent extraction process have been largely completed.

Additional studies were conducted on the conversion of bismuth nitrate to bismuth metal for recycle. Spray calcination continued to receive attention as a means of conversion of the nitrate to Bi_2O_3 . A run in the 14-inch diameter calciner employing 2.5M bismuth nitrate at 7 gph feed rate was apparently successful; product analytical results are pending. Other studies showed that either a protective slag cover or an inert atmosphere will be required in the reduction of Bi_2O_3 to metal

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with carbon. The high corrosiveness of protective slags indicates emphasis should be placed on the inert atmosphere approach.

The recovery of polonium-210 via volatilization from molten bismuth is also under study as a potential means for close-coupling Po-210 recovery to a reactor loop operating with a circulating molten bismuth target. This process, if successfully developed, may offer a minimum cost route to Po-210 production. In pursuing this potential, molten bismuth was successfully sprayed in a two-fluid nozzle with strong indications that a particle size of less than 50 to 100 μ can be readily achieved. This small particle size should promote rapid (in seconds) release of Po-210. Since this portion of the proposed process appears feasible, scouting studies during the next month will evaluate techniques of condensing or otherwise trapping the volatilized bismuth vapor, inert gas and polonium product. If this latter aspect appears feasible, integrated process development will be undertaken.

Recovery of Polonium from Irradiated Bismuth by Solvent Extraction

General feasibility of a solvent extraction process, using dibutyl carbitol as solvent, for separating polonium from bismuth has been shown in cold semiworks studies by G.L. Richardson. Preparations for miniature mixer settler runs to test the flowsheet on feed stock prepared from irradiated bismuth are completed. A sample of aged, irradiated bismuth was dissolved. The solution will be used for runs at one to 10% full polonium level.

Uranium has been suggested as a stand-in for polonium in further cold semiworks tests of the flowsheet. The kinetics of extraction, scrubbing and stripping of uranium (U-233) under the proposed flowsheet conditions were studied. Under all conditions studied, equilibrium uranium distribution ratios were attained in about one minute of vigorous contact. Similar batch contact studies with polonium in place of uranium will be made to permit comparison of the distribution kinetics of uranium and polonium.

Polonium Chemistry

The rate of vaporization of polonium from bismuth oxide into a flow of inert gas is slow. The possibility of an increased vaporization rate during reduction of the oxide metal was tested at 1000°K using 8 w/o H₂ in argon as a carrier gas. Although complete bismuth reduction occurred, only about 19% of the polonium was removed. This amount was about 50% greater than polonium removed from bismuth oxide using pure argon as the carrier.

SECRET

Formation of aggregates of bismuth oxide particles during polonium vaporization experiments at 1000°K and the decrease in volatility after removal of approximately 10% of polonium were thought to be related processes. Visual and microscopic examination, density and surface area determinations of samples of bismuth oxide treated in the same procedure as in the volatility studies did not correlate with the time the sample was held at 1000°K, although there were indications of a change in the properties of the material heated at 1000°K compared to unheated material or to material heated at 900°K.

Thermal Decomposition of Plutonium(IV) Oxalate

Thermobalance studies of the decomposition of plutonium(IV) oxalate have continued. Recent results may be summarized as follows:

1. The quantity of carbon left in the oxide product was reduced significantly by increasing the time at the calcination temperature from 30 to 90 minutes and by increasing the temperature from 350 to 400 C.
2. The use of a CO₂ atmosphere during the calcination as a means of reducing the carbon content of the oxide was found inadvisable. The CO₂ stabilized one of the intermediate compounds formed in the calcination reaction and kept the oxalate decomposition from going to completion until a temperature of about 500 C was reached. These results show again the need for a continuous air flow through the calciner if complete calcination and a product low in carbon are to be achieved.
3. An oxalate "aging" effect has been found. Plutonium(IV) oxalate which has been stored for 6 or 7 days since precipitation undergoes complete and rapid decomposition to PuO₂ at the relatively low temperature of about 270 C (under thermobalance conditions; small sample size, etc.). By contrast, temperatures in excess of 375 C appear necessary for rapid calcination of freshly precipitated oxalate. The mechanism of the "aging" process is not understood; however, it may be due to radiation damage effected by the plutonium alpha radiation.

WASTE MANAGEMENT AND FISSION PRODUCT RECOVERY

Zeolite Storage of Strontium

The storage of Sr-90 (and Cs-137) on zeolites, and the pressurization phenomena which result from radiolytic decomposition of residual water, has been the subject of extensive earlier hot-cell studies. These resulted in the tentative conclusion that equilibrium pressures will

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not be extreme and that the proposed storage system is reasonably safe. The conclusions were clouded, however, with the suspicion that the equipment used may have leaked. To eliminate any ambiguity concerning the behavior of this system, a new test using an all-welded capsule has been undertaken.

Loading was initiated later in the report period and is continuing with a feed containing 0.035M Sr-90 at pH 5. This is being passed through the column at room temperature at a flow rate of 1.0 to 1.4 column volumes per hour. No breakthrough of strontium was detectable on the in-cell gamma spectrometer after 20 column volumes of feed had been loaded, and the column now contains 1.7 milliequivalents of strontium per gram of Linde 4A zeolite. The loading will be continued to breakthrough. Following loading, washing and drying, the capsule will be transferred to the Radiometallurgy Laboratory where the inlet and outlet tubes will be welded shut and the capsule helium leak tested. The internal pressure will be monitored for a prolonged period during which the capsule will be held at a temperature simulating that of a full-size storage container.

Waste Packaging

The heat transfer characteristics of an internally finned waste container (Sr-90 loaded inorganic zeolite) were analyzed. The container holds a 6.4 cubic foot, 27-inch high bed of inorganic zeolite. Sixteen equally spaced internal fins are used to improve heat transfer to the container wall; eight of these extend from container wall to container axis; eight extend only halfway to the axis. Internal and surface temperatures were estimated for a container of this design loaded with one megacurie of Sr-90 (plus 10% additional heat from Sr-89) and cooled by free convection and radiation to 100 F ambient surroundings. Estimated surface temperature was 525 F and the maximum temperature drop through the bed was 850 F. These correspond to a maximum internal temperature of 1375 F. Without fins, the loading would have to be limited to about 0.167 megacurie in order to keep the internal temperature from exceeding 1375 F.

Semiworks Support

The process described last month for removal of manganese from rare earth concentrates, has now been successfully applied in the Strontium Semiworks and the product promethium concentrate is being readied for shipment. A manganese decontamination factor of 80 was obtained, which is remarkably good considering that no centrifuge was employed.

A report, "A Process to Remove Zirconium from Purified Strontium-90," L.A. Bray, BNWC-24, February 5, 1965, was issued summarizing the work

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done (and reported in earlier monthly progress reports) on the removal of stable daughter zirconium from stored Sr-90 concentrates.

Yttrium-D2EHPA Extraction Behavior

The continuing study of the D2EHPA solvent extraction behavior of various ions has been extended to include yttrium. Data on yttrium is of interest because it extracts readily and has always been difficult to strip from the solvent in plant operations. Its equilibrium distribution into 0.4M D2EHPA, 0.2M TBP-NPH was measured as a function of added complexant and acidity (from pH 6 to 16M HNO₃). Nitric acid was found to be a very poor stripping agent as the minimum E_a^0 was 0.2 at 6M HNO₃. Hydroxyacetic acid, tartaric acid and HEDTA were also ineffective. DTPA and EDTA gave attractively low E_a^0 values of 0.01 at about pH 4; however, EDTA tended to precipitate at pH values below 3.3 or above 4.8. DTPA appears to be the most satisfactory choice at this time.

Clean U-233 Studies

A series of runs are in progress in the spray calciner to scout effect of feed composition and operating conditions on the quality of the thorium produced by spray calcination, both with respect to suitability for cold pressing and as a feed for the Sol Gel process. Feed variations being tested include thorium nitrate concentration, addition of sucrose (to produce a "thermal spike"), and the use of slurry feeds produced by addition of oxalate, hydroxide, carbonate or peroxide.

The 67% theoretical density (TD) cold pressed target elements described last month have been welded shut and are ready for charging to the reactor. Irradiation will be for eight to ten weeks.

Cerium-Rare Earth Partitioning

Work was continued on the cerium-rare earth separation flowsheet described last month. In this flowsheet, a D2EHPA extract of mixed rare earths is contacted with a strip solution containing HNO₃, K₂S₂O₈ and AgNO₃. The cerium is oxidized to Ce(IV) which, with the aid of a scrub stream, is retained in the solvent while the remaining rare earths are stripped.

The pilot plant pulse column runs this month demonstrated that high cerium decontamination factors (greater than 300) could be obtained with the feed point five feet above the bottom of the 16-foot tall cartridge. A scrub stream of clean solvent was introduced at the bottom of the cartridge at a flow rate approximately equal to the strip flow plus 25% of the solvent feed flow. The column temperature was 50 C. Eliminating the scrub stream in one short run caused the cerium DF in the aqueous

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product to decrease to less than 12. A small amount of unidentified precipitate was found in aqueous grab samples after a short period of standing. The color varied from nearly white to yellow, apparently as a function of the amount of cerium present.

Additional work is planned to adapt this flowsheet for use in the Waste Management Program in B-Plant.

Solvent Extraction Recovery of Np and Pu from Purex Waste Sludge

Studies toward developing a batch solvent extraction process for recovering neptunium and plutonium from feed solutions (PAS) obtained by HNO_3 leaching of Purex process stored waste sludge were continued. Results with synthetic PAS solution show extraction of Pu(IV) into 0.04M D2EHPA-diluent is very rapid. Minimum contact time may be desirable to minimize extraction of zirconium and iron. More than 99% of the plutonium is stripped from the solvent during 30 to 60 minutes contact with an equal volume of 0.1 to 0.25M oxalic acid at 25 or 50 C. Extraction of Pu(IV) reduction of Np(V) to Np(IV) by hydrazine and ferrous sulfamate and subsequent extraction of Np(IV) were not impaired by making the PAS 0.02 to 0.05M in citric acid, tartaric acid or EDTA. Previous studies indicate these complexants may be of value in reducing zirconium extraction. It is estimated that PAS will be from 0.01 to 0.02M in uranium. In batch contacts with synthetic PAS containing 0.02M uranium, essentially all of the uranium was extracted along with Np and Pu by an equal volume of 0.04M D2EHPA-diluent. However, only 10-15% of the uranium was stripped by oxalic acid.

EQUIPMENT AND MATERIALS

Ion Exchange Column Instrumentation

Instrumentation is under development for application to remotely operated chromatographic columns used for promethium and americium purification. Laboratory tests of neutron absorption techniques indicate that these methods may be useful in detecting the presence of high cross section impurities such as europium, gadolinium, and samarium. Thus, a neutron absorptiometer may aid in identifying the various bands of isotopes as they progress through the multicolumn system. An absorption cell, plus an in-line alpha cell for americium detection are being readied for installation as prototypes on Chemical Processing Department prototype columns in the 222-S Building.

In related tests, a magnetic flowmeter and solid state converter system produced flow control within the required accuracy over the range 15 ml to 4 liters per minute. A ceramic capacitance probe for resin level

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measurement does not appear feasible based on testing to date. Its use for liquid level detection is under study; current emphasis is on defining a suitable coating to improve setting characteristics to reduce the response time.

Computer Control Studies

Chemical process control systems normally use three-mode pneumatic or electronic controllers. A process control computer can effectively substitute for several score of such controllers. The computer is programmed to solve numerically the equations which describe the controller action, given the setpoint (from optimization schemes possibly) and input data from the process. The result of the computation is an output signal to a final control device, such as a valve or pressure regulator. The GE-412 computer has been programmed to do these computations and the system has been demonstrated to perform at least as well as a standard three-mode controller when controlling pulsing frequency.

When a computer is used in this fashion it is possible to effect very rapid and smooth startups of chemical processes using a technique known as "bang-bang" control. This technique, in the case of a second order process (one adequately described by a second order differential equation), results in a sequence such as the following: (a) valve full open for 60 msec, (b) valve full closed for 200 msec, and (c) valve to final position for steady state conditions. This type of control has also been incorporated into the GE-412 and tested using the column pulsing frequency system. Steady state conditions were achieved in about 1.5 seconds from cold start as compared with about 15 minutes when the system is started by advancing the set point on a controller from zero to the steady state value.

Three programs were written and debugged to enable telemetered signals to be accepted and processed by the computer. The programming system provides for 20 signals to be monitored, a running average to be calculated, the number of signals comprising the average to be tallied and a typewritten log to be generated whenever a tally reaches any specified number. Potential applications of telephone line telemetry to the study of various chemical processes are now being investigated.

Effect of Chromium and Iron on Corrosivity of Thoria Dissolvent

Stainless steel corrosion products (chromium and iron) increase greatly the corrosivity of thoria dissolvent to 304-L stainless steel. During a 100-hour exposure of 304-L to boiling thoria dissolvent (12.5M HNO_3 -0.01M HF, 0.10M $\text{Al}(\text{NO}_3)_3$) containing 0.01M chromium(III), the corrosion

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rate was 62 mils/mo; in a similar test with 0.05M Fe(III) present instead of chromium, the rate was 9 mils/mo. Thoria was dissolved to 1M $\text{Th}(\text{NO}_3)_4$ in a high-acid thoria dissolvent (15.2M HNO_3 -0.01M HF -0.1M $\text{Al}(\text{NO}_3)_3$). While thoria was dissolving (24 hours) the corrosion rate of 304-L was about one mil/mo. Exposure of 304-L in the solution at boiling was continued 110 hours after thoria dissolution was complete. During the last 24 hours, the corrosion rate was 75 mils/mo. Chromium concentration in the solution at the end of the test was 0.006M, based on weight loss of the test sample.

Protection of Cable Junctions

Three resistance temperature detectors were connected to the approved type cable and the junction was potted for protection. The first junction was cast in Ren RP 3260-A epoxy (Ren Plastics, Inc.) with an overlay of the same material. The second was cast in the same epoxy with an overlay of RTV 60 silicon rubber (General Electric). The third was cast in the RTV 60 rubber with an overlay of the same material. All three junctions were tested in water in accordance with HWS-8278, Rev. 1, paragraph 8.4.5 except that the resistance to ground was tested after each step. The first junction failed after 7 cycles, the third after two cycles, and the second withstood the complete test.

REACTOR DEVELOPMENT - 04 PROGRAMPLUTONIUM RECYCLE PROGRAMElectrochemistry in Molten Salts

The laboratory program of the past few months has been devoted to development of improved equipment and to studies of melt preparation problems prior to work on the electrochemistry of some of the fission products of importance to Salt Cycle development. Apparatus has now been built to enable conversion of a commercially available potentiostat to either cyclic or conventional chronopotentiometry. Apparatus for the double pulse method for determining kinetic parameters of electrode reactions has also been completed.

In work on melt preparation it has become obvious that, for LiCl-KCl melts at least, recrystallizing techniques for purifying the salts prior to fusion are not as good as electrochemical purification of the molten salt. The chief difficulty with the latter procedure is that when excessive amounts of impurities are present in the melt, and are electro-deposited in the clean-up step, they tend to spall off the cathode and remain in the melt.

Salt Cycle Process Engineering Development

In the High Level Radiochemistry Facility, processing was continued with a mixture of UO_2 -0.5% PuO_2 fuel elements irradiated to 2000 and 5000 Mwd/ton in the Plutonium Recycle Test Reactor. After additional plutonium enrichment was added in the salt bath, parts of the codeposited product from two electrolyses were mixed and refabricated by Remote Processes and Equipment Development personnel into nine fuel pins containing about 4.7 kg of UO_2 - PuO_2 . The pins were then incorporated into a 19-rod cluster and are now being irradiated in the PRTR.

An analysis of the fuel is given below:

<u>Mesh</u>	<u>Wt %</u>	<u>Wt % Pu</u>
- 4 +10	55.0	0.87
- 20 +65	25.1	11.0 14.1 0.87 3.25
-100	19.8	5.6 14.2 0.87 4.49

The average weight percent PuO_2 in the fuel is estimated at 1.96%.
The fuel came from two electrolysis runs with an average analysis:

<u>Run</u>	<u>Wt %</u>	<u>Wt % Pu</u>	<u>ppm Cl</u>	<u>O/U Ratio</u>	<u>ppm "oil"</u>
H 30	71.6	0.87	41	2.0056	<50
H 31	28.4	3.87	120	2.0063*	<30
Total	100	1.73**	63**	2.0058**	<46**

*After hydrogen reduction; value was about 2.05 before reduction.

**Average

RADIOACTIVE RESIDUE PROCESSING DEVELOPMENT

Coupled Spray Calciner-Melter Solidification

Four runs were made with simulated Nuclear Fuels Services' first cycle waste (111 gal/ton U) containing added phosphoric acid in concentrations ranging from 1.15 molar to 1.73 molar ($M^+/P = 3.0$ to 2.0) as feeds. The spray calciner furnaces were operated at 700 C for these runs and the melt temperatures ranged from 1060 C to 1125 C. Feed rates were restricted to three gallons per hr. by the capacity of the melter. This rate produced 0.6 cubic feet of melt per day.

Phosphoric acid concentrations of 1.15 molar and 1.28 molar ($M^+/P = 3.0$ and 2.7) were the most successful of the four concentrations tested. For these two feeds, calciner wall deposition was almost non-existent; pressure drop across the filters was relatively high (22 inches of water).

Increasing the phosphoric acid concentration to 1.51 molar ($M^+/P = 2.3$) and subsequently to 1.73 molar ($M^+/P = 2.0$) brought about a marked increase in calciner wall deposition. Powder built up around the entrance to the melter and eventually plugged it. The higher phosphoric acid concentration feed plugged the calciner filters in two hours of operation.

The melt produced from these feeds is a purple-black crystalline material with a density of 3.3 grams/cc.

One hundred and thirteen pounds of calcine for laboratory glass and corrosion studies were produced in the spray calciner from a simulated NFS first cycle waste (73.6 gal/ton U) with no chemical additives. Liquid feed rate for this 16-hour run was six gallons per hour at a calciner wall temperature of 700 C. Calciner wall deposition was small and filter performance was satisfactory. Inspections after this run revealed a crack in one of the filters. The crack occurred adjacent to a weld

connecting the porous stainless steel filter to a solid stainless steel retaining collar. Failure is believed to be due to metal fatigue sustained from vibrations caused by the calciner's pneumatic hammer. The filter was replaced.

Spray Calciner-Melter Sulfate Recycle

In one mode of operating a spray calciner-melter combination for solidifying wastes, the off-gases from both devices are condensed and returned to the feed concentrator. If sulfate is present in the waste and volatilized from the melter it will be recycled to the spray calciner feed. To determine steady state conditions it is necessary to know how sulfate volatilization varies with concentration of sulfate in the feed.

Simulated Purex 1W wastes containing additional sulfate to simulate recycled sulfate were prepared. Phosphoric acid was added to give a M^+/P ratio of 2.3. The wastes were batch calcined, melted and held at 950 C for three hours. Sulfate retained in the melt was determined. The ratio of sulfate remaining in the melt to initial sulfate decreased from 0.11 to 0.022 as the sulfate concentration in the waste was increased from 0.137 to 0.5M. Under these conditions, sulfate retention was more nearly a constant amount per unit weight of melt than a constant fraction of the sulfate in the feed.

Waste Solidification Engineering Prototype

Design verification tests continued with two calcination runs, a 12-inch pot test using simulated TBP-25 waste as feed and an 8-inch "spindle" pot test using NFS first cycle waste as feed. The spindle pot has a hollow center (spindle) and represents a concept for a pot with centerline thermocouples which are not a permanent part of the pot.

Eleven hundred liters of TBP-25 waste were processed in 84 hours during the 12-inch run, netting 83.5 kg of calcine with the pot 95% filled. Seven hundred liters of NFS waste were calcined in 66 hours during the 8-inch run, netting 65 kg of calcine. Thermocouple-type liquid level probes provided excellent pot level control during both runs. Performance in other respects was, in general, good.

WSEP Instrumentation

On-line computer processing of two WSEP runs was accomplished during the month. Additional flow meter signals were incorporated into the program to improve material balance calculations.

Additional instrumentation work included viscosity-temperature measurements of 7 glass samples and development of a pneumatic pot pressure

device. An analog computing device was placed in service as a reflux computer on the evaporator. The device multiplies a flow signal and a ΔT with a calibrated accuracy of 0.5 percent.

Systematic Glass Studies

Completion of the measurements on drip temperature and solubility of the sulfate free, 10% fission product series has shown that the sulfate markedly affects these properties only at high metal to phosphate ratios (M/P) where a substantial fraction of the sulfate is retained after melting at 1000 C. The 1200 C melt (sulfate bearing) closely parallels the sulfate-free in its properties. The effect on solubility is not sufficient to be a deciding factor as to whether or not a sulfate-free system should be produced.

Some additional measurements are suggested to understand the influence of crystallization on drip temperature. It appears that the high drip temperatures previously reported as occurring at M/P 1.5 - 1.6 are drastically reduced by devitrifying the glass.

Two samples of melts from simulated projected NFS waste have been prepared at M/P ratios of 2.3 and 1.0. These have relatively high drip temperatures and lower solubilities in accord with the higher iron content.

The 75% Fission Product series is underway. Melting temperatures are markedly higher than the 10% series, thus indicating the existence of an upper limit to the fission product concentrations which can be handled in the phosphate system at less than 900 C.

Waste Solidification Condensate Treatment

Scouting studies are in progress to evaluate methods for removing sulfate from the Waste Solidification Engineering Prototype glass melter off-gas condensate. Design flowsheets predict this condensate waste will contain 1-4M HNO_3 , 1-3M H_2SO_4 , 0.05-0.2M H_3PO_4 and up to 5% of the feed ruthenium (i.e., activity about 1 curie/gallon). Present effort is directed toward evaluating practical ways of removing the sulfate by crystallization techniques, estimating the activity levels of the salt cakes formed, and determining how to dispose of the salt cake.

Calcite (0.124-0.246 mm crystals) was used to precipitate calcium sulfate from simulated phosphate glass melt off-gas condensate (3M HNO_3 , 2.5M H_2SO_4 , 0.1M H_3PO_4). The gypsum precipitated by this method forms a filterable porous deposit at the calcite granule site. Ninety-six percent of the sulfate was removed from the condensate with a stoichiometric

addition of calcite. Five percent of the Cs-134 and 60% of the Sr-85 remained with the precipitated gypsum after washing with dilute nitric acid.

Crystallization of sodium sulfate by addition of a stoichiometric amount of sodium hydroxide removed 63 to 71% of the sulfate from the waste at 0 C. "Spiked" solutions of the simulated waste showed 4, 5, 7, 7 and 9% of Cs-134, Ru-106, Sr-85, Zr-95 and Ce-144, respectively, in the condensate to be retained on the vacuum-filtered sodium sulfate crystals. Crystals washed with saturated sodium sulfate solution showed less than 1.5% of the Cs-134 and Sr-85 remained on the crystals.

CONTAINMENT SYSTEMS EXPERIMENT

Major Facilities and Equipment

Chicago Bridge and Iron Company completed the three-part containment vessel during the week of March 22. Pressure testing, as required by Section VIII of the ASME Code, should begin the week of March 29.

In preparing for the pressure test, an engineering appraisal of structural damage to T-Plant and danger to personnel was made in the event of vessel failure. Safeguard procedures were established to minimize risks assumed. These were reviewed with the Battelle Safeguards Council. Physics and Instrument Department personnel will be testing an acoustic method for detecting crack propagation during the pressure test.

Struthers-Wells Corporation is on schedule with the design of the reactor simulator vessel. Preliminary drawings were provided for information and comment.

Mathematical Models

The alteration of the blowdown portion of the computer program to include the effect of vessel sensible heat and to indicate temperature gradients in the vessel walls was refined further to take into account variations in the boiling heat-transfer coefficient with pressure and temperature difference. The effects of the pressure-temperature state at saturation upon nucleate boiling, transition film boiling, and the maximum and minimum heat fluxes were evaluated using the theoretical equations of Zuber and others. Empirical correlations relating the heat-transfer coefficient to temperature difference and pressure were obtained and included in the program. The program has not been run to date. Results of the calculations of heat transfer coefficients indicate that at high pressures maximum coefficients are considerably above that assumed in the initial run of the program. Temperature gradients may therefore be somewhat more severe than previously reported.

Instrumentation

Specifications were completed for the multi-point temperature monitoring system for the outer containment vessel. This system will utilize precision platinum resistance temperature detectors and double resistance bridges to read out temperatures with an accuracy of ± 0.1 F and a repeatability of ± 0.03 F. Recent information indicates the need to read out temperatures well above the specified calibration range of 50 - 100 F. Investigation of this problem revealed that the input circuit boards for the data logging computer peripherals can be field modified to accommodate the increased millivolt range.

A study was made to compare a multiple pin connector system with the use of individually sealed signal leads. It was concluded that rigorous moisture penetration tests must be passed by the multiple pin connector to permit their use.

SHIPPING HAZARDS

The initial tests will be concerned with fission product release and transport behavior from simulated phosphate-glass waste material at high temperatures. Design and construction of experimental apparatus is continuing. Stainless steel crucibles have been fabricated for use at temperatures up to 1,200 C. A quartz furnace is being fabricated and induction coil configurations have been developed. A kilogram of phosphate glass representing an expected waste from power reactor fuel reprocessing was ground to minus 60 mesh to provide a uniform source for preparation of individual samples of "spiked" glass.

OFF-GAS DISPOSAL TO GROUND

Consideration of homogeneous anisotropy for flow of fluids (cylindrical coordinates with vertical z-axis) shows that (1) the flow rate is determined by the value of the horizontal permeability; (2) the travel times are determined by the vertical permeability, and (3) the anisotropic section can be transformed into an equivalent isotropic section by multiplying the actual radial distance by $K_v/K_h^{1/2}$. The permeability of the equivalent isotropic section is equal to K_h .

For isothermal flow of a compressible gas with given relative geometry, there is an optimum depth of injection where the flow rate becomes a maximum. This optimum depth varies with injection pressure and with temperature. The travel time increases with increasing depth.

A new plotter program was written for the off-gas research program which, in addition to plotting equipotentials and streamlines on the same plot, permits the use of these data to plot various flow pattern developments considering soil anisotropy. This capability eliminates several machine passes for each anisotropic condition.

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BIOLOGY AND MEDICINE - 06 PROGRAM

TERRESTRIAL ECOLOGY - EARTH SCIENCES

Hydrology and Geology

The STREAM and GENORO computer programs are being used with actual field data to obtain additional knowledge on the fitting and calculating of permeabilities from multi-term polynomials. Fitting trials of 28-, 36- and 45-term polynomials were tried with the decision that 36 terms provided the best fit in this case. Streamlines and associated permeabilities are now being evaluated.

A relatively shallow (30 feet) test hole was core-driven immediately next to the 699-11-45 well to obtain sediment samples for soil moisture and density determinations. These data will be correlated with neutron probe (moisture) readings in the adjacent cased well to obtain an accurate calibration curve for the instrument.

Drilling of a water supply well by private operators about 4 miles west of the Yakima Barricade in the upper Cold Creek Valley artesian well basin is providing abundant data not otherwise obtainable on the basalt series. The well has penetrated more than 500 feet of the basalt series. Samples of cuttings and data from field mapping indicate that the well has penetrated the four Priest Rapids basalt flows, two Rosa flows, the Squaw Creek diatomite bed equivalent, and is now in the Frenchman Springs flows where artesian water is expected. The area is a major source for natural recharge of confined and unconfined ground waters that move eastward beneath the Hanford project. Data are expected to help explain differences in head noted in different aquifers beneath Hanford.

Soil Chemistry

Studies of anion-soil reactions were continued in an effort to further characterize simultaneous transfer of solutes and solvent through soil material. Diffusion coefficients of phosphate and dichromate were determined at various soil moisture contents. The diffusivity of the dichromate ion ranged from 0.88×10^{-8} to 3.9×10^{-8} cm²/sec at volumetric moisture contents of 0.17 and 0.24. Diffusivity of the phosphate ion ranged from 0.47×10^{-8} to 0.90×10^{-8} cm²/sec at the same moisture contents. Attempts to fit dichromate and phosphate breakthrough curves to chromatographic equations were not successful.

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ATMOSPHERIC RADIOACTIVITY AND FALLOUT

Fallout Studies

Eight radionuclides were measured by direct multidimensional gamma ray spectrometry on an ashed sample of Alaskan caribou liver obtained by Wayne Hanson of the Radioecology Section. These nuclides and their concentrations in disintegrations per minute per kilogram (of liver) were: Na-22, 783; Mn-54, 320; Co-60, 85; Zn-65, 87; Ag-110m, 461; Cs-134, 36; Cs-137, 23,000; and Th-232, 10. By comparison with measured air and vegetation concentrations the route of these radionuclides from fallout to man can be studied.

Two samples of euphausiids, a shrimp-like crustacean, obtained from the Pacific Ocean were counted by multidimensional gamma ray spectrometry. Concentrations of seven radionuclides, Mn-54, Zn-65, Co-60, Ag-110m, Th-228, Cs-134 and Cs-137 were measured. In one sample the concentration of Cs-137 was about 100 times greater than the other sample and contained an even greater relative concentration of Cs-134. This indicates that the origin of some of the radionuclides in this sample was the Columbia River.

Meteorology Support Studies

A second field experiment was conducted with Atmospheric Physics personnel on February 28 utilizing Xe-133 and ZnS particles as dual tracers for atmospheric dispersion studies. Six sampling stations were placed on a 200 meter arc, six samplers on a 1600 meter arc, and a single sampler upwind to obtain a background sample. All phases of the experiment proceeded well with 1.5 curies of Xe-133 being emitted along with 760 grams of ZnS.

RADIOLOGICAL AND HEALTH CHEMISTRY

Radiation Chemistry

A series of Food and Drug Administration approved food colors were tested as radiation damage inhibitors in 0.2% agarose-99.8% water jellies. The food colors were incorporated into the jelly before solidification to give an $8.00 \times 10^{-3}M$ solution. Damage was measured by the dose of Co-60 radiation required to cause liquefaction. Erioglaucine was the most effective protective agent but FDC Blue No. 2, an indigo dye, was found to be nearly as good. This finding may introduce a new class of protective agents to our study.

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Multidimensional Gamma Ray Spectrometry

Our second multidimensional gamma ray spectrometer achieved its first operation on March 17. This spectrometer appears to be better than the first system, as the large plastic anticoincidence shield is a more efficient Compton canceller and background reducer and the thin aluminum cladding of the primary detectors allows measurement of gamma rays and X-rays of lower energy than the stainless steel clad of the first spectrometer.

A computer program was written to allow machine computation of data from the multidimensional spectrometer. This program is not one of great sophistication but simply solves a series of simultaneous linear equations heretofore done by hand.

An important application of multidimensional gamma ray spectrometry when combined with neutron activation analysis was shown to be in the study of certain trace elements in the blood. Measurements were made after neutron irradiation and four weeks delay to allow for decay of the shorter lived radionuclides, Na-24 and Br-82. The trace element concentrations of Sc, Fe, Co, Zn, Ag, Sb and Cs were determined by direct multidimensional gamma ray spectrometry measurements of Sc-46, Fe-59, Co-60, Zn-65, Ag-110m, Sb-124 and Cs-134. With 0.1 ml samples of blood serum and an irradiation time of one week at a neutron flux of 5×10^{13} n/cm²/sec levels as low as a few $\mu\text{g/g}$ (for Sc) could be measured. The values for Sc, Ag and Sb in serum obtained in this study appear to be the first ever reported.

RADIOISOTOPES AS PARTICLES AND VOLATILES

Turbulent Deposition

Turbulent deposition theories were reviewed to determine possible areas of improvement in the theoretical understanding of particle deposition. Areas of improvement include better estimates of particle eddy diffusion coefficients, particle approach velocities to the tube wall, and particle concentration at the edge of the boundary layer. Based on this review a model approach is planned which will incorporate improvements in these areas.

Additional experiments were carried out on the turbulent deposition of particles in vertical 0.5 inch diameter tubes. These measurements generally fit into the pattern reported previously, although departures at both extremes of the correlating parameter, ϕ_{st} , range has appeared. At very low values of ϕ_{st} , the deposition ratio, K/V , is higher than would be indicated by a straight line extrapolation of the bulk of the

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data. For example, 1.8 micron particles were deposited from a stream flowing at 0.7 cfm with a K/V value of 2.6×10^{-6} . This value is several orders of magnitude greater than the extrapolated value. This measurable amount of deposition at the small ϕ_{st} value (2.4) is probably the result of mass transfer due to the finite value of the particle diffusivity.

Attempts to extend the measurements into regions of high ϕ_{st} values were unsuccessful due to particle re-entrainment at high flow rates. For example, using 8 micron diameter particles and gas flows of 3.8, 4.25 and 4.6 cfm, the observed values of K/V were 2.2×10^{-5} , 1.26×10^{-5} and 0.55×10^{-5} , respectively. These values are roughly two orders of magnitude lower than would be expected for no re-entrainment, indicating that only about 1% of the particles impinging on the wall adhered.

ISOTOPES DEVELOPMENT - O8 PROGRAM

PROMETHIUM ISOTOPIC FUEL DEVELOPMENT

Prototype Heat-Source Development Equipment

Following a decision earlier in the month to remove the dry chemistry operations from the hot cell, necessary design changes were made and fabrication has been initiated on two lightly-shielded glove boxes, which will house the precipitation, calcination, hydrofluorination, metal reduction and capsule loading operations. Capsule decontamination and welding will be performed initially in existing facilities of another component. These changes will greatly increase operational reliability by eliminating the danger of cross contamination with impurities present in incoming promethium crude (cerium, europium and other fission products). Operational capability will also be improved through the potential for simultaneous (rather than sequential) operation of the purification and conversion equipment.

Delivery of most of the in-cell equipment and material items is on schedule. Most of the large fabricated steel items (trays, tanks, equipment racks, 8-inch loading column, valves, etc.) have been delivered and are being assembled in the mock-up area for checkout and partial piping while awaiting movement into the cell. Delivery of the in-cell gamma spectrometer is expected this week. Design of the in-cell optical gamma spectrophotometer is complete and fabrication is well along. (The experimental in-cell optical spectrophotometer, if successful, will greatly simplify control of the ion-exchange process and should be applicable to a full size plant.) Meanwhile, removal of all of the waste calcination equipment from the cell has been completed, the cell decontaminated to permit

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personnel entry, and the cell door removed to the shop for installation of a dry-waste transfer mechanism. Determining factor on when in-cell installation of the promethium equipment can begin is the time required to decontaminate, dismantle, overhaul and repair the failed General Mills manipulator. Current estimate is two months.

With decision to separate the purification and conversion functions, the conversion chemistry and metal reduction boxes (which had been designed some time ago when it was planned to do the entire operation in shielded glove boxes) were redesigned slightly for installation in another location. Fabrication should be under way early in April. The 30-ton press is under construction. The induction heating equipment for the vacuum reduction furnace has been received and an order has been placed (with three weeks delivery promised) for the associated vacuum equipment. The precipitator and reduction furnace are also under construction.

Compatibility Studies

The core-clad compatibility program (described last month) continued; however, explosion of several of the sealed Vycor containers (of Nd_2O_3 with test specimens of various metals and alloys) necessitated premature removal of all of the samples from the furnace. The explosion was traced to probable reaction between a steel shim and a zirconium compound contained in the walls of the mullite crucibles. The Nd_2O_3 has been recovered and purified for re-use in capsules which will not contain steel shims.

In higher temperature work, specimens of Ta, Ta-10W, W, W-25, Re, Mo and Pt have been prepared in graphite crucibles for heating with neodymium or samarium oxides in a vacuum induction furnace belonging to Graphite Research and Development.

Re-entry Burnup

Several interesting and informative comparisons of the burnup behavior of promethium (metal) and strontium fluoride (SrF_2) were run on the Hanford BISOAR (Burnup of Isotopic Sources on Atmospheric Re-entry) computer program. The outstanding feature of this program is the simultaneous solution of the trajectory as it is perturbed because of the decreasing source radius. Thus ablation results in more rapid deceleration of the source remaining because of its reduced ballistic coefficient. This rapid decrease in velocity in turn results in less ablation than that calculated on the basis of a constant ballistic coefficient and decreases the probability of complete burnup. The behavior of two otherwise similar heat sources, one of Pm and the other of SrF_2 , were

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computed for 2-in. x 6-in. Rene-41 clad cylinders released at 7600 m/s and -1° at 96,000 meters. Because of the low thermal conductivity of SrF_2 , the center temperature was relatively unaffected by the surface heating, and burnup was incomplete. The results with promethium were quite different. Because of Pm metal's relatively high thermal conductivity, the temperature at the center of the capsule begins to rise soon after the surface temperature does, and melting of the promethium is complete by the time the cladding is completely ablated. The promethium is thus released completely in the molten state at relatively high speed, which should result in dispersion to quite small particles and essentially and "instantaneous" burnup (which does not occur with SrF_2).

In another calculation, the ability of a Rene-41 clad promethium capsule to maintain its integrity under conditions of a launch pad missile fuel fire-ball was computed. It was concluded that the temperature would not exceed the melting point of the cladding. Similar results were obtained with several other typical materials, such as Haynes-25 and Inconel.

Pneumatic Impaction Development

Installation and calibration of strain gages for measuring punch pressure on the 1220-F Dynapak machine continued during the month. Readout of strain gages on an oscilloscope is satisfactory but further adjustment is required on the oscillograph. The strain gages on the frame continue to show promise as a method of measuring punch pressure during impact.

Since the bolts were torqued, a total of 500 impactions have been made without failure of any equipment that would require remote maintenance in a hot cell.

Strontium oxide and strontium fluoride have been produced with the multi-purpose precipitation-filtration equipment. Both products have been analyzed by X-ray diffraction and appear suitable for feed to the pneumatic impaction process as was the cerium oxide product reported last month. An X-ray diffraction analysis of the latter indicated no detectable impurities.

The cerium fluoride product reported last month was found to be unsuitable due to the presence of several percent CeO_2 in the unsintered product and about 50% CeO_2 in the sintered product.

Q M Platt for
Manager
Chemistry Department

MT Walling:AMP:cf

BIOLOGY DEPARTMENT

A. ORGANIZATION AND PERSONNEL

C. R. Watson, Research Scientist, joined the Experimental Animal Farm Section as a new employee on March 22.

B. TECHNICAL ACTIVITIES

FISSIONABLE MATERIALS - 02 PROGRAM

Columnaris

No columnaris organisms were isolated from approximately 300 fish sampled during the past month at Bonneville Dam, Patterson Ferry, Hanford slough and Wenatchee. The percent of fish with agglutinating titres remained about the same as in previous months. There appeared to be a slight decrease in titre values but it is as yet impossible to determine whether this is a trend or merely a modest variance for this particular sampling period.

Reactor Effluent Monitoring

Monitoring of salmon in 0, 2, and 4% reactor effluent at three different temperatures continues at 100-KE. The lots in coldest water completed hatching in early March. Egg mortality averaged 5.5%, with a range of 3.8-7.6% for all lots. These values are well within normal limits. To date, there is no correlation between mortalities, temperatures, and effluent concentrations. Total mortalities for all stages averaged 10.8% with a range of 6.9-15.0% for all lots.

Toxicity of Alkaline Potassium Permanganate Cleaner

Toxicity of a spent reactor tube decontaminant containing alkaline KMnO_4 and sulfamic acid was determined on chinook salmon fingerlings. No mortality occurred at 0.75% test solution, but at 1.25% (1.84 ppm KMnO_4 , pH 8.8), more than 80% of the fish died within 24 hours. Fish tested in 2.95 ppm chemically pure KMnO_4 (pH 7.7) suffered a 10% mortality at 24 hours. Data in the literature indicate 12 ppm as the LD_{50} for 24 hours for Gambusia, which illustrates the importance of species difference and testing on site.

BIOLOGY AND MEDICINE - 06 PROGRAMPhosphorus

The estimate of 11-12 days for the effective half-life of P^{32} for our stock of trout and Columbia River whitefish, over a period of 1-28 days postingestion, indicates little or no species difference. At 28 days postingestion the liver and GI tract of whitefish showed the highest concentration of P^{32} .

Calcium - strontium

Urinary-fecal excretion ratios for Ca^{45} and Sr^{85} were determined at daily intervals following intraperitoneal injection. Data are presently available for only the first four days and indicate an unchanging ratio for both Sr^{85} and Ca^{45} subsequent to the first day. These results are somewhat at variance with earlier data but cannot be properly evaluated until the experiment is completed.

Strontium

A technique was developed, using thin layer chromatography, for fractionating lipids extracted from tibia and femur marrow of rats. Rats to be used in this study are now receiving 0.02-0.5 μCi Sr^{90} /100 g, twice weekly, intraperitoneally.

Two swine which had received 125 μCi Sr^{90} /day for about three years were killed because of a rapidly deteriorating physical condition. Peripheral blood values and findings at necropsy were highly suggestive of leukemia. One of the animals showed severe icterus prior to death. At necropsy, the hepatic lymph nodes appeared enlarged and were occluding the cystic and common bile ducts. In addition to the four confirmed cases of leukemia in this group, there are now four cases of probable leukemia awaiting histopathological confirmation. It is of particular interest that there is a close genetic relationship among six of these eight cases.

Dr. Jack Alexander has been retained as a consultant in radiology and is assisting us in developing proper techniques for early detection of bone damage.

Iodine

Six ewes fed 3 mCi I^{131} six years ago and three control ewes were sacrificed after being fed 5 μCi I^{131} /day for 30 days, during which period I^{131} excretion data was obtained. Fecal I^{131} excretion was variable but not greatly different between the two groups. The urinary excretion of I^{131} was about a factor of three greater in the group previously fed 3 mCi. Average tissue I^{131} concentrations were about equal in the two groups with the exception of the adrenal gland which contained up to eight times more I^{131} in the control ewes. Stable iodine analyses of various tissues are being obtained for comparison of specific activity.

Cesium

The pregnant ewes fed Cs^{137} daily since December have body burdens about nine times the average daily intake. This value is slightly higher for March than for February or January, but only about one-half that observed in male sheep one year ago. This difference may be related to body weight, sex, diet, and/or potassium content of the diet.

Plutonium

When DTPA dissolved in dimethyl sulfoxide (DMSO) was applied to the shaved skin of rats, the urinary excretion of subcutaneously administered Pu^{239} nitrate was increased. This increase, however, was no greater than that obtained when DTPA alone was administered intravenously. The tissue retention of the Pu^{239} was reduced in the DMSO treated animals. When Pu^{239} oxide was employed instead of the nitrate, the results were inconclusive.

Neptunium

Nicotinic acid and ATP were tested for their ability to prevent Np^{237} induced fatty livers in rats. Both compounds, when injected following Np^{237} administration, were effective in lowering liver fat accumulation. Neither compound prevented the rise in blood isocitric dehydrogenase (ICD) or glutamic-pyruvic transaminase (SGPT) that is seen following toxic doses of Np^{237} . Adrenalectomy and hypophysectomy were effective in decreasing liver fat accumulation and also prevented the rise in blood ICD and SGPT.

Inhalation Studies

In studies with rats it was shown that administration of NH_4Cl enhanced the effectiveness of thiazolidine-4-carboxylic acid in clearing I^{131} from the thyroid. Further experiments are necessary to determine optimum doses and regimen.

After taking cyanide in their drinking water for three months, rats inhaling $\text{Fe}_2^{59}\text{O}_3$ particles deposited > 25% more of these particles than non-cyanide treated controls. The rate of clearance of $\text{Fe}_2^{59}\text{O}_3$ particles from the lungs was not significantly altered by the cyanide treatments nor was it altered in other rats treated with high concentrations of cigarette smoke.

In vitro solubility tests with Sr^{85} compounds used in inhalation studies showed that the titanate was much less soluble in blood plasma than the oxide, carbonate, phosphate, or fluoride. These results are consistent with in vivo studies in which Sr^{85} titanate was retained in the lung to a much greater extent than the other Sr^{85} compounds.

Immediately, or one day after exposure to uranium ore dust, the uranium and thorium of the deposited ore particles in rat lungs are virtually in secular equilibrium. The average ratio of Th^{230} to U^{238} was 1.03, which is essentially the same as observed in the ore dust before inhalation. As earlier reported, this ratio increases to about 9 after several months' retention in the lung.

Two dogs exposed to "10 μ " and "20 μ " Fe^{59} -labeled microspheres were sacrificed to study site and size of particles retained in the lungs. The particle suspensions to which these animals were

exposed contain small percentages of particles smaller than the nominal size. The "20 μ " suspension contains particles as small as 7 μ and the "10" suspension contains some 5 μ particles. Preliminary indications are that deposition in the dog lung is not due to selection of these smaller particles.

Analyses of tissues from a dog that died two years and eight months after exposure to Pu^{239} oxide showed 63% in lung, 17% in bronchial lymph nodes, 14% in liver, and 47% in bone. The total body burden was 2.9 μCi and compares well with estimates of 3.1-3.3 μCi obtained with the chest counter being developed by the Physics Department.

Dr. D. Englund, a geneticist from Oregon State University and a consultant to the Biology Department, reviewed our dog breeding program. He recommended continuation of the three main lines in our colony and suggested methods for maintaining the genetic background and improving the homogeneity of the colony by hybridization.

Extensive study of the suspected hookworm infestation of the beagle colony has led us to question the identification of the eggs found in the fecal specimens. While they have certain morphological similarities to the ancylostoma (hookworm) egg, other factors do not substantiate this identification. It will probably be necessary to consult a parasitologist to solve our problem. Until it is solved, there can be no relaxation of the stringent isolation precautions we have invoked in the dog colony.

Cocarcinogens

Further experiments were initiated to clarify the effects of $\text{Ce}^{144}\text{-Pr}^{144}$ on induction of hepatic tumors by DMAB. The level of DMAB feeding was increased from earlier experiments in an effort to increase tumor incidence and decrease induction period. Both male and female rats are being studied with the radionuclide injected at two different time periods during DMAB feeding. Other groups of rats are being fed DAF or ethionine with and without $\text{Ce}^{144}\text{-Pr}^{144}$ to determine whether these carcinogens might be better adapted for studies of this type. Three analogs of DMAB (the 2'-, 3'-, and 4'-methyl derivatives) are being studied for their effects on liver retention of $\text{Ce}^{144}\text{-Pr}^{144}$. These derivatives exhibit a wide range of carcinogenicity and it will be interesting to determine if their effect on radionuclide retention correlates with their carcinogenic properties.

Gastrointestinal Radiation Injury

The movement of tracer Na^{22} and H^3 from the lumen of the intestine to the blood and from the blood to the lumen is being studied in rats previously subjected to various radiation exposures with and without external drainage of bile. Preliminary results demonstrate the feasibility of the procedures employed but are, as yet, insufficient to support any conclusions concerning the effect of the various treatments.

Particle solubility studies

Procedures were developed for precise measurement of the solubility of slightly soluble particles of interest in connection with SNAP and SNPO hazard evaluation programs. The method employs small numbers of particles handled on an essentially individual basis without intervention of membranes or filters and permitting microscopic observation and photography during the course of the measurements. A reasonable simulant for human gastric juice has been developed and some preliminary tests of the apparatus performed on Pu^{238} oxide particles.

Plant Nutrition

Native soils high in sodium (taken from a greasewood area), high in potassium (taken from the vicinity of hopsage), and the usual or normal calcareous soil were used to grow barley in the laboratory. All soils were spiked with Cs^{137} . The uptake of this nuclide from the greasewood soil was reduced over three-fold as compared to the other two soils. A similar reduction was previously noted for the uptake of calcium and Sr^{90} from the high sodium and high pH greasewood soil.

Effects of dimethylsulfoxide (DMSO) on seed germination were evaluated for rye, barley, peas and beans. No germination occurred at or above 10% DMSO, while at 5% and below, germination was comparable with controls. Some retardation of root length and of seedling height was noticed with increasing concentrations of DMSO.

Microbiology

It was previously reported that methionine increases the relative synthesis of RNA in E. coli. The technique of sucrose density gradient ultracentrifugation is being employed to examine qualitatively the nature of the RNA synthesized in the presence of methionine as well as in the presence of other amino acids. Results are not yet available but should indicate whether methionine is causing the synthesis of specific kinds of RNA or an increase in the same kinds as normally observed.

A technique was developed for measuring the kinetics of amino acid transport in N. crassa. Preliminary results suggest that L-tryptophan transport does not require an external energy source. There is, however, high endogenous metabolism in this organism so that the distinction between metabolically or non-metabolically mediated transport is not clear. Inducible transport is being examined.

Radiation Effects on Insects

Dimethylsulfoxide (DMSO) gave no radiation protection to X-rayed female flour beetle germ cells whether fecundity or fertility was scored. An ovicidal effect was observed when females were cultured

on food containing DMSO. The onset of reproduction was delayed as compared to controls. Cytological observation of ovarioles showed that DMSO treatment resulted in abnormal oocyte development. The more metabolically active trophocytes, compared to oocytes, are expected to be more adversely influenced by both radiation and DMSO, as compared to effects on the oocytes.

Preliminary review of data from the combined DDT and X-ray experiment indicates a higher weight per beetle in those exposed to 4 kR than in the controls or the low exposure groups. This is not unreasonable since the higher exposure reduced the number of adults so that more food was available for those which did live. Further analysis of the data will be made.

Terrestrial Ecology

Periodic sampling of the shrub, Mountain Maple, of the Blue Mountains during 1964 showed no increase in leaf concentration of Cs^{137} during the period the leaves were on the shrub. Either the major accumulation occurs at the time the leaves are first unfolded and little subsequent contamination occurs, or the rate of contamination through the year is offset by concomitant loss of activity from the leaves.

These data for deciduous leaves are in slight contrast to the previously observed increase in fallout levels in leaves of conifers exposed for more than one year.

Alaskan Studies

Statistical analysis of Eskimo Cs^{137} body burdens was begun. Under the assumption of a constant Cs^{137} intake during several periods of the study, an estimate of effective half-life for Cs^{137} of about 70 days is obtained. The significance of this estimate is uncertain at this time.

R. C. Thompson
BIOLOGY DEPARTMENT

RCT:sw

APPLIED MATHEMATICS DEPARTMENT
MONTHLY REPORT - MARCH 1965

ACTIVITIES FOR OTHER HAPO COMPONENTS

N-Reactor Department

Data were analyzed relative to the increase in defective fuel ends attributable to rewelding the end caps.

A previously used sampling procedure for steam generator tube bundles was evaluated for use in the April 1965 outage.

Irradiation Processing Department

An analysis of data was completed for the first stage of a response surface study for an optimal fuel mix. The second stage experimental layout was designed.

A study was initiated to adapt certain recent developments relative to the Weibull model, to the design and analysis of current run-to-rupture fuel experiments.

A study was completed relative to quench and anneal effects in terms of irradiation-induced distortion of fuel elements.

An analysis was initiated on experimental data relative to the effects of certain bonding procedures in terms of irradiation-induced distortion of fuel elements.

Mathematical models pertaining to stratified fluid flow from large reservoirs are currently under investigation. Such models have application to release studies from Lake Roosevelt behind Grand Coulee Dam.

Chemical Processing Department

Work is nearing completion on the EDPM program for the previously developed mathematical model of the chemical counter current crystallization process. Process parameter studies with the model should begin in approximately two weeks.

ACTIVITIES WITHIN BATTELLE-NORTHWEST2000 Program

Modifications leading to increased efficiency and versatility have been completed on the EDPM program which calculates the transient heat transfer characteristics of a source-laden slab of material.

Data analysis was initiated for the purpose of contrasting the sulfate concentration in the Columbia River at Hanford with that at Vernita.

4000 Program

Ultrasonic Studies

Collaboration with members of the Physical Measurements Unit on preparing an expository treatment of stress wave transmission and attenuation is nearing completion. In addition, the EDPM program "WAVES" has been modified so that input, fitted, and attenuated pulse data are automatically plotted on the Benson Lehner equipment.

Other

Seven separate indices of Pu burn-up are currently employed in interpreting the results from certain PRTR experiments. A study was initiated to (i) assess each of the seven indices in terms of propagated experimental errors and (ii) develop a "best" single index.

Data analysis was initiated to estimate parameters in a non-linear model of certain chemical data.

Data analysis was initiated on PRTR fuel particle data obtained in a pre-nupac experiment.

Data analysis was initiated on data obtained in an experiment concerning the nupac and vipac subsystems.

A non-linear model was developed to express the relationship between the extent of cracking and the force applied to reactor metals. Several sets of experimental data were analyzed in accordance with this model.

5000 Program

Statistical Methods in Radiochemistry

Work continued on the power function calculations for the uniformly most powerful similar test to detect radioactive decay in the presence of random background. A Fortran language program was completed and is currently being debugged for

calculating unconditional power for both fixed and random time counting procedures. The Fortran language program for calculation of critical points for the uniformly most-powerful unbiased test of the null hypothesis that the variance of a normal distribution lies in a specified interval was completed. The format for tables of these critical values is now being worked out and calculation of the critical points will start momentarily.

The GEM program is being modified so that the weights used in the least-square estimation of nuclide decay rates reflect instrument instability as well as counting statistic variation. Mathematical work on this problem was completed and the Fortran language machine program is currently under revision.

Other

Least squares estimates and confidence intervals were obtained (by iterative search procedures) for the parameters in a model employed in the actinide research program.

6000 Program

Data analysis was initiated on experimental data relative to the protection afforded by cystamine during irradiation.

Prototype experimental data on the retention of Cs^{137} in trout were studied to compare least squares parameter estimates obtained for (i) an exponential model and (ii) the linear model based on the log transformation.

Specifications are being written for a computer program to convert counting data from gastrointestinal studies into corrected counts-per-minute for samples with more than one radionuclide present.

A computer program was written to assist the analysis of experiments on the effect of root temperatures on the uptake of Sr^{85} and Cs^{137} .

Sample survey data are being analyzed relative to the level of Cs^{137} retained by villagers in Northern Alaska and the amount of Cs^{137} in caribou meat.

An exponential model was formulated for the relationship between the gross beta and P^{32} retained by Columbia River whitefish.

Progress is being made on developing an EDPM program to calculate the time integrated exposure and deposition over a large area due to a continuous point release.

Work is continuing on obtaining and numerically evaluating the solution to a mathematical model for the diffusion, delay, and dissolution of gaseous contaminants that have been injected into the earth.

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Work is nearing completion on EDPM programs which calculate and generate equipotential and stream lines for two and three dimensional ground water flow systems.

GENERAL

Work was continued on the analysis of the data from General Electric Cincinnati concerning the effect of neutron radiation on certain properties of alloys.

Carl A. Bennett

Manager

APPLIED MATHEMATICS DEPARTMENT

CA Bennett:lg

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RADIATION PROTECTION DEPARTMENT
REPORT FOR MARCH 1965

Organization

Mr. G. E. Backman terminated the end of the month to accept a position with General Electric Company in CPD. Phyllis M. Schrotke accepted a position with Dosimetry Studies and Evaluation Section effective March 22. Joan Palmer, who was on temporary assignment in Radiological Development and Calibrations Section, accepted a permanent assignment in RPD effective March 22.

Laboratory Experience

There were two incidents involving two employees which required special bioassay sampling for plutonium analysis. In addition to the incidents involving plutonium, there were two incidents involving three employees that required evaluation of possible intake of other radioisotopes.

Three employees received hand exposure ranging from 2.5 to 5.8 rem at the Plutonium Development Laboratory, 308 Building, while loading aluminum cylinders with Pu-Al and borated plastic discs. The discs contain 20 wt. % Pu of which 26% was Pu^{240} and were fabricated for physics tests at the 305-B Test Reactor.

Environmental Evaluation

The average concentration of fallout materials in the air in the vicinity of the Hanford project during the four week period ending March 19 was 0.4 pCi/m^3 as determined by beta counting of air filter samples. This value represents a slight increase over the average values of 0.2 pCi/m^3 recorded for the past three months and reflects the expected spring influx of longer-lived fallout materials.

Levels of radioactivity found in the Columbia River and in fish have been normal for this time of year. No unusual circumstances were noted during the month.

Nuclear Safeguards

A draft of the design nuclear safety criteria for the Fast Flux Test Reactor was issued. These criteria will serve as guidance for the design and nuclear safety review of the facility.

Further calculations were made of estimated damage to farm lands and buildings on the Wahluke Slope in the event of accidents at K Reactors and at Purex, for the guidance of the Atomic Energy Commission in deciding whether to open the Slope lands.

A study of flow stoppage in the Rupture Test Loop of the Plutonium Recycle Test Reactor was made, differing from that in the Safeguards Analysis in that the fuel element was a 19-rod cluster and the basket tube was omitted. The possibility of geyser-like ejection of coolant from the stagnated tube was considered. It was concluded that the time for operator action to save the fuel element could be very short. A temporary deviation was prepared requiring immediate operator action (instead of within two minutes), and addition of instrumentation and controls to make the action automatic was recommended.

The AEC was informed that the Rupture Test Loop in-reactor test section will be moved from the periphery to the center of the reactor.

A proposed outline for the revised safeguards analysis report for the PRTR was issued for comment.

The general revision of the PRTR Process Specifications begun in the fall of 1963 was completed.

The response of the PRTR electrical system to the BPA power dip of March 3 was evaluated. The dip lasted for 18-60 cycles. The electrical system responded as designed. When the system was tested on March 4 by simulating a BPA outage, it again responded as designed.

Work is continuing on the high power density core analyses. Analog computer runs indicate that some melting of fuel at the center of several rods of 19-rod clusters at the highest power level will take place. Studies are continuing on whether center-line melting is tolerable, and if so, to what degree. A major problem yet to be resolved is the effect of a reactor excursion on the temperature profile in the fuel.

Work continued on preparation of the High Temperature Lattice Test Reactor Final Safeguards Analysis. Material was given to Graphics for illustrations, and a critical description of the computer-type safety circuit was prepared.

A special condition to permit remote manipulation of a test device worth more than 5% $\Delta k/k$ was prepared for PRCF flux traverse experiments.

Dosimetry Studies

A study to determine the limitation of current whole body counting procedures and equipment for in vivo measurement of plutonium lung burdens was initiated using approximately 0.4 μCi (10 x MPBB) of normal grade plutonium and a counting time of one hour. It was determined that the 60 kev peak from the Am^{241} daughter can be detected. However, sensitivity was poor with the photopeak spread over a large number of channels. The study will be continued to better define the detection level.

Individual reports summarizing the whole body penetrating exposure received during 1964 were prepared and distributed to employees of Battelle-Northwest, General Electric, J. A. Jones, Vitro, WPPSS and RLOO-AEC. The reports to Battelle-Northwest employees also contained a summary of their internal exposure.

Signaling dose alarm instruments patterned after a Chalk River design were fabricated and their performance under varying environmental and radiation conditions was studied. Initial studies conducted at dose rates of 100, 600, and 1800 mR/hr indicated satisfactory dose integration over this dose rate range. Temperature and battery voltage effects are currently being studied.

Development of the fission fragment neutron dosimeters continued. This dosimeter, which consists of fissionable material placed in contact with Lexan plastic, continues to show promising possibilities for use as a personnel neutron dosimeter and for the detection of extremely small quantities of fissionable material. As a part of the study, several long exposures on the top stringer of the sigma pile were made. From this data it was possible to determine the number of tracks on the Lexan film per incident neutron for several uranium isotopes. If exposures at different neutron energies are made, a more meaningful quantity is the number of tracks per neutron-barn. For uranium-235 there is an average of 5.8×10^{-6} tracks/neutron-barn. If one considers that only the fissions occurring within one fission fragment path length can cause tracks, then one can calculate the number of "effective fissions" per unit area. For a fission fragment range of 6.7 microns, the number of tracks per "effective fission" was 0.16 to 0.21. This number is less than unity because only the largest etch pits are counted in evaluating the effects on the Lexan plastic. Uranium-235, uranium-238, and neptunium-237 fission foil dosimeter units were exposed to a few rads of 0.5 Mev, 1.0 Mev, 1.3 Mev and 1.5 Mev neutrons. As expected, the Lexan films showed a relatively flat energy response for uranium-235; the uranium-238 showed a large difference from 1.5 Mev to 1.3 Mev.

The calibration of the Co^{60} radiation facility of the Biology Department was completed.

Environmental Studies

A study is in progress of the effects of reactor effluents on Columbia River quality. Congelation and reduction of routine operating and meteorological data continued. Two dye tests were run. One from B Area for equilibrium concentration measurements downstream, and one from D Area for time of travel measurements as far as the 300 Area. Additional temperature probing was done of the B, C, K, and D effluent plumes for direct measurement of effluent concentrations in the River.

A study of the cause of high inlet temperatures at the 181-N river pumphouse has been in progress. Additional measurements were made to confirm the significance of the K trench seepage on the inlet temperatures. A final report is in preparation.

Applied Reactor Engineering, GE-IPD, is studying the possibility of predicting 181-D inlet temperatures in advance. J. P. Corley, Environmental Studies & Evaluation Section, was asked to consult with the IPD representative and to assist in any way possible. The possibility was discussed of a dye test from the WPPSS outfall now under construction. Such a test would provide pre-operational heat distribution data for comparison with reactor-caused temperature increases now experienced at 181-D.

Hanford's whole body counter has provided a considerable file of data on the radionuclide body burdens of adults living in the Tri-City area. However, there is much less counting data for children. A study of environmental exposure mechanisms must include children among the population sample used and an effort is planned to remedy the problem. Possible approaches using the public school system to assist with large-scale counting of children are under investigation. At the same time, a method for investigating the diet of children counted is being developed. The use of whole-body counting also appears to be the most reliable method for verifying the amount of fish consumption by successful fishermen. To provide enough such data to permit statistical analysis, whole-body counts must be obtained for at least fifty or sixty such subjects. It is planned to locate the mobile whole body counter near popular fishing locations on the river and invite fishermen to volunteer for whole body counts. Officials of the State Game Department will cooperate with the project.

The preparation of a Manual on monitoring requirements for power reactors continued. The Applied Mathematics Department is providing assistance on a section that will deal with sequential sampling techniques.

Radiological Development

Several new approaches to the use of LiF for neutron dosimetry are now under investigation. These studies are directed toward increasing the neutron sensitivity and detection limit for LiF. For exposures where the gamma dose is low, a detection limit of about 70 mrad was observed with one of these techniques. These results are encouraging and indicate that programs now planned with LiF have a good probability of meeting the performance criteria for personnel neutron dosimeters.

Work on the electron Van de Graaff pulse circuit was continued to provide for pulse durations in the microsecond region. A switching amplifier coupled to

a photomultiplier tube can be used to provide remote switching and maintain the 2 million volt isolation required between various components of the accelerator.

The type of criticality alarming instrumentation to be provided in the Fuels Recycle Pilot Plant was reviewed. The use of neutron sensitive detectors in place of gamma detectors offers advantages of increased criticality detection reliability and minimum false alarm possibilities that may arise due to the expected highly fluctuating gamma radiation dose rate in various parts of this facility.

A complete recalibration of the Richland Water Plant monitor was initiated for correlation of dose rate determinations made with this monitor and the dose rate calculated on the basis of radioanalysis results. Arrangements were made with the Standards Laboratory to provide duplicate samples of those radionuclides prepared for the calibration of the U. S. Testing Company counting equipment. Good intercalibration should thus be obtained between the water plant monitor and the analytical sample results. The ratemeter drive circuit for operation of a graph recorder dose rate readout was completed by Technical Shops and was installed in the monitor system.

Outside Consultations

Two meetings were held with school officials concerning a project for counting school children in the whole body counter. A generally cooperative attitude was expressed. No problems were detected from their point of view with the proposed project.

A meeting was held with members of the State Game Department concerning their cooperation with our fishermen contact and creel survey programs. The Department is interested in maintaining the cooperative venture.

Radiological Development and Calibrations Section personnel assisted the AEC Division of Biology and Medicine in evaluating the performance of criticality dosimeters used at various AEC contractor sites. As a part of this performance demonstration, the Hanford Criticality Dosimeters which were developed, calibrated and thoroughly evaluated several years ago were used. Agreement of preliminary results between criticality dosimeter systems from Battelle-Northwest, Oak Ridge National Laboratory, Y-12, K-25, Phylatron Corporation and Savannah River was within $\pm 25\%$ for two reactor bursts. The reactor bursts were 7.7×10^{16} and 6.8×10^{16} fissions over a time period of about 70 μsec . The second burst was shielded by 8" of graphite to provide a moderated neutron spectrum.

Three requests were made by U. S. Testing Company, Inc. for assistance in checking out equipment for routine performance. A total of about one man-day was

provided. This checkout resulted in adjusting the badge processing machine, the film x-ray identification coder and the mechanized film densitometer. The temperature control on the developer solution of the film processing machine was also adjusted to provide the same processing temperature as that used when the equipment was operated by Battelle-Northwest. Processing temperatures were approximately 6°C above recommended processing temperatures resulting in increased film densities per unit dose.

Staff Activities

R. F. Foster attended a meeting of the Health Physics Society Program Committee in New York City on March 4-6. He also conferred with Dr. John Harley (HASL) and with AEC-Washington staff members of the Divisions of Operational Safety and Biology and Medicine on March 8.

R. F. Foster described parts of the "Mechanisms of Environmental Exposure" program to AEC's Advisory Committee on Biology and Medicine on March 27.

R. F. Foster and R. B. Hall met with Mr. W. Towne and Mr. A. Moore of the Portland office of the U. S. Public Health Service and representatives of RLOO-AEC on March 18, to discuss sampling and analysis of the Upper Columbia River.

J. P. Corley attended the biennial Industrial Waste Conference held at the University of Washington on March 18-19.

J. K. Soldat taught one class session of the course "Control of Radioactive Wastes" (R559) at the University of Washington Graduate Center in Richland on March 2. The lecture presented was entitled "Effluent Sampling and Environmental Monitoring."

Two papers, "The Hanford Emergency Dosimetry System," coauthored by H. V. Larson and A. R. Keene, and "The Hanford Criticality Accident: Dosimetry Techniques, Interpretation and Problems," coauthored by H. M. Parker and C. E. Newton, Jr., were presented at the IAEA Symposium on Personnel Dosimetry for Accidental High-Level Exposure to External and Internal Radiation in Vienna, Austria on March 8-12.

H. V. Larson participated in a Symposium on Personnel Dosimetry for External Radiation at AERE, Harwell, England on March 17-18.

During an extended visit in Europe, H. V. Larson visited the following people: Dr. R. Maushart and J. Vaane at the European Transuranium Institute, Karlsruhe, Germany on March 15-16 to discuss dose evaluation and measurement techniques; Dr. J. Rundo and Dr. A. J. Lister at AERE, Harwell, England on March 19 to discuss whole body counting and other internal dosimetry techniques; Dr. A. E.

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Souch at the Berkely Nuclear Laboratories, Berkely, England on March 22 to discuss dose evaluation and measurement techniques with emphasis on high-energy gamma radiation (> 3 Mev) and thermoluminescent dosimetry; C. O. Peabody at Winfrith Laboratories, Winfrith, England on March 23 to discuss thermoluminescent dosimetry.

L. G. Faust visited the Lawrence Radiation Laboratory, Berkeley, California on March 7-9 to arrange for an experiment utilizing their 88-inch cyclotron.

P. E. Bramson and G. W. R. Endres visited Oak Ridge National Laboratory, Oak Ridge, Tennessee March 19 through March 27 to participate in a comparison study of the performance of criticality dosimeters at the request of AEC Division of Biology and Medicine.

C. M. Unruh visited Dr. Margarete Ehrlich at U. S. Bureau of Standards, Washington, D. C. on March 24 to discuss calibration methods and equipment. A visit was also made to the Germantown AEC facility to discuss calibration of radiation protection equipment and civil effects test programs with Dr. N. Barr and Mr. J. Deal on March 25 and 26.

R. F. Foster
for Manager
RADIATION PROTECTION DEPARTMENT

AR Keene:RFF:np

REACTOR DEVELOPMENT - 04 PROGRAMPLUTONIUM RECYCLE PROGRAMPlutonium Recycle Test ReactorOperation

Reactor output for March was 1067 MWD for an experimental time efficiency of 65% and a critical efficiency of 52%. There were seven operating periods during the month, two of which were terminated manually, four were terminated by scrams, and one extended through month-end. A summary of the fuel irradiation program as of March 31, 1965, follows:

	<u>Al-Pu</u>		<u>UO₂</u>		<u>UO₂-PuO₂</u>		<u>Other</u>		<u>Program Totals</u>	
	<u>No.</u>	<u>MWD</u>	<u>No.</u>	<u>MWD</u>	<u>No.</u>	<u>MWD</u>	<u>No.</u>	<u>MWD</u>	<u>No.</u>	<u>MWD</u>
In-Core			3	1174.5	76	16401.3			79	17575.8
Maximum				414.7		463.7				
Average				391.5		215.8				
In Basin	7	572.5	30	3806.2	62	9580.6			99	13959.3
Buried							1	7.3	1	7.3
Chem. Process.	68	5465.8	35	1965.8					103	7431.6
Program Totals	75	6038.3	68	6946.5	138	25981.9	1	7.3	282	38974.0

(Note: MWD/Element x 20 $\approx$ MWD/TU for UO₂ and UO₂-PuO₂.)

Heavy water loss and indicated helium loss for the month were 468 pounds and 118,000 scf., respectively.

Equipment Experience

A total of 1963 craft manhours were utilized during the March reactor outages as follows:

Repair	736 Manhours
Improvement	535 Manhours
Operations Assistance	363 Manhours
Preventive Maintenance	329 Manhours

Improvement Work Status (Significant Items)Work Completed

Tube 1946 Inlet RTD
Supplemental Emergency Water Addition

Work Partially Completed

Corrosion Loop Installation
Air Lock Door Operators
Modification to PRTR Warehouse 3718-C
PRTR Steam Utilization
Additional Fuel Storage and Examination Facility
Creep Test Facility
Flux Wire Scanning System
D₂O Cleanup Facility
Pneumatic Irradiation Facility

Design Work Completed

Decontamination Building (CAH-116)
PSCD Sample Station
Gasometer Condensate Tank and TKA Vent Line Revision
Shim Well Shielding
Instrumented Fuel Element Thermocouple Installation
Boric Acid Prototype Facility
Relocate Rupture Loop

Design Work Partially Completed

Instrument Power Supply
PRTR Experimental and Building Facility Addition
PRTR Increased Power Level

Process Engineering and Reactor Physics

Seven of the 58-inch UO₂-2 w/o PuO₂ elements (including the salt cycle) are under irradiation. The maximum exposure as of March 31, 1965, was 92.9 MWD (2802 MWD/TU). The power generation on these elements continued to be in the 1000 - 1350 KW range.

PRTR Test 63-1, Reactivity Worth of Coolant Removal from fuel channels, was prepared, issued, performed and reported during March. This test covered a measurement of the reactivity effect of removing coolant from tubes containing unirradiated HPD fuel elements. The measured reactivity change upon loss of coolant was small and negative, which implies that the coolant voiding effect in the HPD core will still be negative.

A continuing trend of primary system activity increase has been observed since about October 1964. A program was set up with CSDO for evaluation of the cause of the increase and the necessary support activities within PRTRS initiated. This will include crud sampling, specific activity analysis, and continued monitoring of activity buildup. Since pH reduction as a result of angle valve leakage was coincident with the activity increases, steps have been taken to maintain primary pH above 10.0.

Experimental Reactor Services

The status of the various test elements at the end of March 1965 is shown below. Those elements discharged prior to March 1, 1965, have been deleted from this table.

Test Channel No.	Location	Element Number	Description	Date Initial Charge	Date Dis-charged	Approximate Accumulated MWD
14	1956	5097	Swaged-0.48%	4/2/62	--	285.5
14	1758	5099	Vipac-0.48%	5/8/62		294.6
37	Basin	1097	UO ₂ -Physics	5/12/62	3/6/65	317.2
48	1156	5150	0.48% ($\frac{1}{2}$ " x $\frac{1}{2}$ " pads)	8/1/62		295.2
54	1542	5116	0.48% (clip-on pads)	5/8/62		302.8
54	1554	5118	0.48% (clip-on pads)	5/8/62		463.7
61	Basin	5186	0.48%-Physics	5/28/63	3/6/65	318.4
61	1552	5192	0.48%-Physics	6/13/63		309.2
67	1047	5117	0.48% (Repaired wire)	10/20/63		265.5
72	1253	5253	1% (Zr coupons)	9/1/64		168.7
85	1855	5230	Vipac - 1%	1/30/64		232.2
108	1954	6501	EBWR Cluster	1/30/65		19.0
FERTIF 6	1946	6500	Salt Cycle, 2%	3/16/65		7.8

Thirty-two fuel elements were examined in the basin. Three elements were rejected for further irradiation.

Twenty-seven process tubes were examined. One showed a .005" fret mark at the lower fuel element support.

Fuel Element Rupture Test Facility

A standard 58-inch length Mark I fuel element containing six outer rods of 2% PuO₂ in UO₂ fuel material obtained by the salt cycle process was charged for the reactor startup of March 17. The estimated power generation of the salt cycle element was 875 kw at full reactor power.

Irradiation was continued on UO₂ #1067 with an extended longitudinal slit until the outage of March 5. During this time the element had accumulated about 3.4 MWD exposure in 173 hours of operation.

Chemical Processing of Spent PRTR Fuels

Arrangements have been made with the Redox Operation of GE-CPD to conduct a chemical processing campaign in late May or early June. Approximately 8 kg of plutonium containing 20% Pu²⁴⁰ would be recovered from the selected fuel elements.

TECHNICAL SHOPS SECTION

A total of 25,868 hours of shop work was produced with a backlog at the end of the period totaling \$195,081 or 21,675 hours. To meet emergency requests for service, it was necessary to work 1,409 overtime hours or 6.5% of the total hours produced in Technical Shops.

LABORATORY MAINTENANCE SECTION

Total productive time was 30,200 hours of 32,400 potentially available. Craft overtime worked during March was 3.4% of total available hours. Manpower utilization (in hours) was as follows:

A. Shop Work		2 800
B. Maintenance		10 500
1. Preventive Maintenance	3 000	
2. Emergency or Unscheduled Maintenance	2 000	
3. Normal Scheduled Maintenance	5 500	
C. R&D Assistance		16 900

WD Richmond

Manager

Test Reactor and Engineering Services

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.

INVENTORTITLE OF INVENTION OR DISCOVERY

D. R. Green and
H. L. Libby
L. T. Pedersen

Emissivity Independent Infrared Thermal
Testing Method, HWIR-1822.
Energy Conserving Flywheel

**DATE
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