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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1962

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OAK RIDGE NATIONAL LABORATORY
STATUS AND PROGRESS REPORT
APRIL 1962

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
UNION CARBIDE CORPORATION
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U.S. ATOMIC ENERGY COMMISSION

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1962

This Status and Progress Report summarizes the unclassified portion of the Laboratory's work. Some of the topics are included every month, but the majority are reported on a bimonthly schedule.

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROGRAM

EGCR Core Top and Bottom Flow Distribution Test. — Tests performed on the adjustable orifice of the bottom dummy for the EGCR core have shown that the range of control it provides is not adequate. Therefore a series of tests has been started for obtaining a design that will give a control range of 500 to 2350 lb/hr. Additional tests of a top dummy containing 21 machined grooves 0.245 in. wide and 0.48 in. deep have shown that this configuration gives the least variation in flow for concentric, eccentric, and slanted positions of the top dummy in the EGCR fuel extension sleeve and thus permits predictable flow control for the annulus coolant.

EGCR Control-Rod-Drive Testing Facility. — Installation work continued on components of the testing facility for the EGCR control-rod drive. The test vessel, which includes the heater and graphite assembly, is complete except for installation of the reactor nozzle extension. Electrical and instrumentation control panels and wiring and the coolant piping are being installed. The compressor and the motor-generator set were installed. The estimated date for delivery of the rod drive mechanism for testing is now October 1962.

GCR-3 Design Study. — Preliminary layouts were made of the primary cooling system of a large-scale advanced gas-cooled power reactor (GCR-3) with a thermal power level of approximately 2000 Mw. Based on a triangular lattice pitch of 9.4 in., the core diameter is 27 ft 9 in.; the core can be contained in a spherical reactor vessel 52 ft 16 in. in diameter. Four primary coolant loops with horizontal shell-and-tube steam generators can be fitted into two horizontal cylindrical containment vessels 64 ft in inside diameter and 236 ft long. The coolant system is designed to operate at a pressure of 400 psia with reactor inlet and outlet temperatures of 577 and 1150°F respectively. Four steam cycles for the plant were analyzed by the Westinghouse Electric Corporation, and thermal efficiencies of 36 to 38% were calculated.

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GCR-3 Lattice Calculations and Fuel-Cycle Cost Studies. – A survey study was made for a restricted range of parameters to determine the reactivity limitations on fuel exposure and the resulting fuel-cycle costs for the GCR-3 as a function of lattice pitch and enrichment. The attainable fuel lifetime and the resulting fuel-cycle cost were calculated for a range of lattice pitches and fuel enrichments, for a batch-loading cycle, for an idealized equilibrium cycle requiring continuous refueling, and for a stepwise approach to the idealized equilibrium cycle.

The study showed that there was no practical reactivity limitation on the attainable exposure up to exposures of at least 40,000 Mwd per metric ton of uranium. For a 9.4-in. triangular lattice pitch, which is the lattice pitch of the reference core design, a variation of enrichment from 2.2 to 4% results in a lifetime variation from 10,000 to 40,000 Mwd per metric ton of uranium. Within this range there may, of course, be metallurgical limitations on the attainable exposure. For fuel enrichments as high as 4%, the fuel-cycle cost continued to drop as the enrichment was increased. Extrapolation of the calculations indicates that the optimum lattice has a triangular pitch of about 11 in. and a fuel enrichment of about 4.5%. The overall fuel-cycle cost, which includes the capital cost, will optimize at a smaller lattice pitch than this. For the optimum lattice, considering fuel-cycle cost alone, the fuel-cycle cost is approximately 1.2 mills/kwhr for the idealized equilibrium cycle.

GCR-ORR Loop No. 1 Operation. – Fuel element 7B, which has a nominal heat rate of 35,000 Btu hr^{-1} ft^{-1} of element (actual, 27,000 Btu hr^{-1} ft^{-1} of element), was irradiated for one reactor cycle of seven weeks and then withdrawn from GCR-ORR loop No. 1 on April 8. It is now awaiting hot-cell examination.

Fuel element 7C, which is of similar design and heat rate, but has 0.0625-in.-diam thermocouples with insulated junctions at all four temperature measurement points in the cladding, was inserted into the loop. It is now operating at a maximum cladding temperature of 1500°F. All the thermocouples are functioning. The thermocouples were protected during assembly by simplified rubber squeeze seals near the top of the lead tube instead of being sealed at bulkheads by means of silver soldering. This design simplified assembly.

During the reactor shutdown, the loop was thoroughly leak-tested, and a small leak was found between the primary helium system and the cooling-water system of the No. 2 compressor. This was corrected by replacing the compressor. The leak may have accounted for the rise in the hydrogen concentration, from < 3 ppm to a high of 300 ppm, in the helium during the run just completed.

GCR-ORR Loop No. 2 Construction. – During the April reactor shutdown, work continued on the GCR-ORR loop No. 2 facility. The transfer table was positioned and the associated mechanisms and linkages were connected; the cell removal-sleeve rollers were installed; the beam-hole shield plug was removed, modified, and reset; and the cooling-water circuits were revised. Preoperational startup procedures and check lists and the operating manual are being prepared.

ORNL-MTR-48 Test of UC_2-Graphite Fuel Elements. – Irradiation of experimental assembly ORNL-MTR-48-6 was terminated after two three-week cycles in the MTR. The assembly consisted of a sweep capsule that contained a graphite-matrix fuel element with pyrolytic-carbon-coated particles and four static capsules filled with unsupported coated particles. The fueled-cylinder central temperature was held at 2000 ± 50°F for 581 hr of irradiation and between 1000 and 1300°F for 135.5 hr. A maximum temperature range for the fuel particles of 1100 to 1250°F was estimated from the observed graphite-support-cylinder temperature of 575 to 650°F. The highest temperature measured for the one instrumented type 347 stainless steel fission-product-deposition specimen was between 750 and 800°F.

Irradiation Effects on Beryllium Oxide. – Experimental assembly ORNL-41-7, which has been in the ETR since November 1961, is scheduled to be removed early in May. The assembly has already received an exposure of 11,538 Mwd – that is, a fast-neutron dose of approximately 7×10^{20} neutrons/cm^2 – and it is expected to receive an additional 2000 Mwd before withdrawal.

The BeO samples for the eighth and ninth experiments have been successfully installed in the capsules. These samples include BeO specimens from Saclay, ORNL-fabricated BeO pellets, and BeO single crystals. The crystals will be irradiated at the same temperatures and to the same exposures as those statistically programmed for the BeO samples.

Reactions of Type 304 Stainless Steel with Low-Pressure CO and CO_2 in Helium. – Current compatibility studies of type 304 stainless steel with CO and CO_2 in helium have shown that carburization differences do not depend upon heat-to-heat variations but are due mainly to section thickness. It was demonstrated that carburization does not increase systematically with the carbon potential $(P_{CO})^2/P_{CO_2}$ but reaches a maximum at a value of about 0.2. Increasing the CO content above this value uniformly decreases the extent of carburization. These results are concluded to be due to the profound retarding effect of the oxide layers on carbon diffusion.

Corrosion of Molybdenum in Gases. – Exploratory studies of the Mo-MoO_2-H_2-H_2O equilibria have indicated that molybdenum will not be oxidized in an atmosphere containing 10% water vapor if the active gas also contains 90% H_2 at temperatures above about 1070°F. Below this temperature, a slight amount of oxidation occurs.

Beryllium Corrosion Studies. – The corrosion resistance of Be–0.5 wt % Ba alloys in dry CO_2 was found to be pressure-sensitive, indicating that barium may promote the formation of the carbonate layers which have been reported to protect beryllium in the presence of moisture.

Studies of Distribution of Carbon in Beryllium Exposed to Flowing Carbon Dioxide. – Previous work showed that beryllium was carburized in carbon dioxide and tentatively demonstrated a correlation between the carbon concentration and break-away oxidation in wet carbon dioxide. In an effort to further understand these observations, experiments involving C^{14} were performed to determine the location of carbon in beryllium exposed to carbon dioxide. Hot extruded Pechiney Super Purity Flake was

studied over the temperature range 1100 to 1500°F for various times. The reaction of the specimens had proceeded to a stage well in the range of break-away oxidation at the time each test was terminated.

After testing, the specimens were sectioned and prepared for metallographic observation, and NTB emulsion plates were exposed in contact with the polished surface of each specimen. Each developed plate exhibited a very dark image which corresponded with the outer periphery of the test specimen. Hence it was concluded that carbon was present in the reaction layer. Since this layer consists of an interdispersion of oxide and metallic particles, it is not possible to conclude from these studies whether the carbon is in the oxide or the metallic particles.

Further evidence of carbon distribution was found by successive polishing and surface counting of the ends of the test specimens. The counting tube was masked so that emissions from only a 1/4-in.-diam circle in the center of each specimen were counted. It was found by periodic examination of the test specimen that the surface count rate was proportional to the amount of oxide present. These observations indicate rather strongly that the carbon was concentrated in the oxide.

High-Temperature Measurements for Gas-Cooled Reactor. — Previous work indicated that exposure of stainless-steel-sheathed Chromel-P–Alumel thermocouples to pure CO resulted in inhomogeneities in the Chromel wire. The composition changes did not cause emf errors, however, because they occurred in isothermal regions. Long-term drift tests have now been started with the sheathed couples exposed to outgassed graphite in a flowing atmosphere of 100 ppm CO in helium (1 atm). After 31 days of exposure, the couples showed an average drift of +0.47 mv (+12°C). Periodic examination for inhomogeneities will be made.

Tungsten–26% rhenium alloys from two sources were tested simultaneously for emf stability at 1700 to 1800°C in a graphite resistance-heated furnace containing a pure helium atmosphere. When checked against a common tungsten wire, the alloy wires from vendor H have been constant in emf during tests as long as 200 hr. The alloy wires from vendor E have all developed slow negative emf drifts averaging about 10 to 20°C during 100 to 200 hr. X-ray fluorescence analyses of the wire surfaces showed the wire from vendor H to be 26% rhenium and that from vendor E to be 29.8% rhenium. Additional analyses and metallographic examinations are in progress. Preliminary results indicate that the rhenium content of the alloy wire from vendor E was reduced during the test.

THORIUM UTILIZATION PROGRAM

Thoria Pellet Development. — Pellets designed to furnish information concerning the effects of grain size and bulk density on attrition resistance were prepared and were subjected to the accelerated spouted-bed test.

The pellets were formed as small right cylinders to a green density of 4.1 g/cc and then isostatically pressed at one of three pressures: 12,500, 18,000, and 35,000 psi. Three different time increments of heat treatment at 1750°C in H_2 were used to sinter pellets from each of the

pressure series: 41 min, 16 hr, and 65 hr. The sintered pellets were partially spheroidized by wet ball-milling, polished, and then tested in the spouted-bed test.

Results of the tests showed that variations of pellet bulk density from 92 to 97% of theoretical did not significantly affect attrition resistance. However, sintering time, which affects grain growth, had a marked influence. Reduction in sintering time from 65 hr to 41 min reduced attrition losses by approximately 50%.

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — One of the limitations in the investigation of ThO_2-UO_2-O_2 phase relationships has been associated with the lack of a variety of reliable methods for accurately determining the occurrence of phase transitions between the single-phase fluorite solid-solution volume and the polyphase region of greater oxygen content. The transition has been inferred from dissociation-pressure measurements and from the manifestation of cubic-phase lattice parameter discontinuities, and has been deduced elsewhere additionally from electrical measurements and thermodynamic estimates, though for a limited number of mixtures containing a high fraction of uranium.

A new analytical instrument, a metallograph with which both reflected and transmitted light may be used, has been installed to provide improved methods for identifying ThO_2-UO_2 + X_2. Preliminary examinations with this instrument indicate that it offers the best available method for detection of very small concentrations of one solid phase in the presence of another.

The lack of a useful etchant for solid solutions consisting of high concentrations of ThO_2 has been one of the limiting factors impeding examinations by previous investigators of these materials. A combination of reagents, consisting of an aqueous solution of H_3PO_4 and HF, was developed recently at ORNL and demonstrated to be very effective in etching ThO_2-UO_2 + X; grain-boundary outlines in solids containing as much as 90 mole % ThO_2 were clearly discernible in metallographic examinations.

Fuel Cycle Development: Fuel Preparation. — Four hundred grams of dense 4 wt % Pu—ThO_2 was prepared by the sol-gel process. Four 11- by 5/16-in. test capsules having a bulk density of 7.5 g/cc were fabricated by vibratory compaction for irradiation in the NRX reactor; the capsules are now being leak-tested.

The feasibility of preparing dense, compactable 3 wt % UO_2—ThO_2 by a simplifying modification of the sol-gel process was demonstrated on a laboratory scale and on a 7-kg scale. The modification consisted in the addition of thoria powder or a water slurry of thoria to a heated agitated solution of uranyl nitrate and nitric acid, whose nitrate and uranium content had been previously adjusted to give an optimum NO_3/ThO_2 molar ratio and a specified U-Th ratio. Particles prepared in the demonstrations had densities > 9.9 g/cc, desirable size, and total 1200°C vacuum gas release < 0.01 cc/g; the particles were vibratorily compacted to bulk densities > 85% of theoretical.

In the development of the sol-gel process for preparation of UO_2-ThO_2, 12 successive 15-kg runs in the rotary denitrator all gave ThO_2 products which dispersed into sols of excellent appearance and free of

undispersible residue. The calcination and reduction furnace and procedures developed are now believed adequate. Methods for determining the nitrate/ThO_2 requirement for maximum dispersion and stabilization of uranium-thorium oxide sols are being developed. It has been shown that this requirement is approximately equivalent to half the active surface sites available on specific ThO_2 samples prepared by steam denitration of thorium nitrate at 475 to 500°C. The available active surface sites can be estimated from crystallite size as measured by x-ray line broadening. In 2 M sols, ThO_2 is maximally dispersed when the pH is ~3.1. Under these conditions, the pH does not change with ThO_2 concentration. A convenient method of determining the net nitrate demand was that of digesting ThO_2 in a slight excess of HNO_3 and then back-titrating to a pH of 3.1 with ammonia.

Eight 11-in.-long fuel pins vibratorily compacted with sol-gel UO_2-ThO_2 have completed one year, or 10,000 to 17,000 Mwd/ton, in the NRX. Six 39-in.-long specimens for the replacement holder in the NRX have been gamma-scanned for density variations, x-rayed, and wound with a wire spiral to prevent coolant loss from bowing.

Fuel Cycle Development: Fuel Fabrication Studies. — A vibratory-compaction evaluation was made of two 13-kg batches of sol-gel ThO_2—3 wt % UO_2 prepared, respectively, by ammonium diuranate addition (batch 4) and uranyl nitrate addition (batch 6). Compaction was carried out on a Branford pneumatic vibrator by use of stainless steel fuel sheaths (45 in. long, 1/2-in. OD, 0.035-in. wall). The oxide particle distribution in each case was 60% -6/+16 mesh, 15% -50/+140 mesh, and 25% -200 mesh. For batch 4 an average bulk density of 8.98 g/cc was obtained for two runs, and for batch 6 an average of 8.92 g/cc was obtained for four runs. It was demonstrated on another batch of material that the compacted bulk density could be increased by about 1% if the midfraction of the particle distribution was ball-milled rather than used in the as-crushed condition.

Testing of gamma-scanning equipment designed to measure fuel-rod bulk density and density variations has continued. In the scanning of nine 5/16-in.-diam vibratorily compacted fuel rods, an excellent correlation was obtained between bulk density measured by gamma absorption and that measured by fuel-column volume and weight. In six rods the difference between the two techniques was about 0.01%, and the maximum deviation in the group was about 1%. Local density variations in the fuel column could be readily and reproducibly detected.

Two vibratorily compacted fuel specimens, one containing fused and crushed ThO_2-UO_2 and one containing sol-gel-prepared ThO_2-UO_2, were discharged from the MTR on April 2, after an estimated burnup of 13,000 Mwd/metric ton. These specimens are en route to the hot cells for detailed examination.

Thorium-U^{233} Fuel Rod Fabrication Facility. — Phase I construction in cell 4, Building 3019, was begun by the contractor. The removable concrete floor has been poured and armor-plate shielding installed on the first floor. The steel alpha-containment cubicles are being prefabricated in the ORNL shops.

Design of equipment for the facility is about 75% complete, and procurement or fabrication of many items has begun. The vibratory-compaction assembly has been constructed and is ready for testing.

MOLTEN-SALT REACTOR PROGRAM

MSRE Design. — Drain-tank-cell drawings showing salt, steam, water, and gas piping arrangements were issued for information. Pipe-support drawings for the drain-tank cell are nearing completion. Drain-tank-support drawings were approved.

Thermal-shield drawings were issued. Also finished, but not yet issued for construction, are charcoal-bed, heat-exchanger-support, and pump-support drawings.

Heater-wiring schematic drawings are being made; those for the coolant circuit heaters are 75% complete. Cable-tray layout drawings for routing heater conductors were issued for comment.

The transfer scheme for the 13.8-kv feeders was finished. A complete instrument tabulation was issued. Design of the control-rod drive is still in progress.

Work has begun on the miscellaneous site-modification design. All major site-alteration design is complete.

MSRE Procurement and Construction. — All dished heads for MSRE components have been delivered to UCNC shops for machining. Fabrication work is progressing on the reactor, heat exchanger, and drain tanks. The radiator enclosure is approximately 70% complete.

The reinforcing steel and side walls of the drain-tank cell in Building 7503 were completed to the height required for the first pour of the concrete walls. Fabrication of the drain-tank horizontal hold-down beams was completed. Fabrication of steel for modifications to the 24-ft-diam reactor containment vessel is progressing, and on-site installation of this steel is expected to start by May 1.

IBM 7090 computer checks of the critical-path schedule for the MSRE indicate that installation can be completed by the scheduled date of July 1963 if deliveries of critical purchased items continue as scheduled.

Development of Components and Systems. — Construction of the full-scale hydraulic model of the MSRE core was completed. Flow distribution from the volute to the core-wall cooling annulus was found to be uniform, and the fluid velocity around the core at the bottom of the core-wall cooling annulus was found to be constant.

The engineering test loop was deliberately oxidized internally at 1200°F to provide a basis for evaluating the effectiveness of the flush salt during initial startup. Flush salt was loaded into the drain tank, and operation was begun without graphite. The 8-in.-diam graphite-access freeze joint was made successfully during the filling operation. Salt is being circulated past the joint at 50 gpm and 1100°F.

The 5-in.-pipe box heaters, which have a heating capacity of 1500 w/ft, were able to maintain a pipe at 1350°F with a heat rate of only 525 w/ft. Heat loss due to the pipe-support structure was 65 w/ft.

Thermal cracking of the box at the mechanical joint was encountered but was eliminated by changing the bolting arrangement.

The proposed MSRE control rod and drive unit was modified to include a magnetic clutch for a scram release and a drive speed of 0.45 in./sec. Testing was begun under MSRE conditions of temperature and cooling-gas flow.

Preliminary tests on the pair of 5-in. MSRE freeze flanges were completed. At the end of five thermal cycles, the ring-joint gas-seal leakage was less than 10^{-5} std cc of helium per second, and the permanent distortion was less than 0.015 in. at the flange face.

The pipe-handling, -machining, and -brazing operation required to replace remotely a drain and flush tank was successfully demonstrated in a dimensionally accurate mockup under exact conditions except for the substitution of a catwalk and a grating for the portable radiation shield. Visual and leak-detection inspections indicate a satisfactory joint.

The procedure for a typical maintenance task in the reactor cell was written, and design drawings of the in-cell structure for piping alignment, component jacking, and freeze-flange clamp stowing are approximately 80% complete. The hydraulic drive units for operating the freeze-flange clamps were tested successfully. A tool was designed and tested for operation of the heater power disconnects.

A ring-joint flanged disconnect was developed for use in vertical lines which are intermittently subject to salt flow. A special prototype flange joint of carbon steel was built and tested. A series of long-term runs at 1000°F and 70 thermal cycles from room temperature to 1000°F indicated favorable temperature distribution and leak-tightness.

MSRE Pump Program. — The Inconel centrifugal molten-salt pump (PKP type) has operated satisfactorily for 2132 hr in an endurance test with molten salt $LiF-BeF_2-ThF_4-UF_4$ (65-30-4-1 mole %) at 1225°F, 1950 rpm, and 510 gpm.

Endurance testing of one of the canned-rotor-type pumps for the lubrication stands was interrupted by an internal short in the stator winding. The winding is being inspected for indications of the source of the failure. The stator was replaced, and endurance testing of the pump was resumed. A procedure was developed for sealing the joint between the bonnet and body of the valves for the lubrication stands.

Two impeller castings and one volute casting for the coolant pump and one impeller casting for the fuel pump are acceptable. The second coolant-pump volute and first fuel-pump volute castings are being weld-repaired. Two pairs of volutes and impeller castings for the fuel pump are being poured by the founder.

Fabrication of the pump tank for the prototype fuel pump was completed, and the hot-test stand is being assembled. The bearing housing of the rotary element for the prototype fuel pump was modified slightly, and assembly is progressing.

Drawings for the lubrication stands were approved and issued for fabrication. Final versions of the drawings of the drive motors for the MSRE pumps and the leak-test and welding procedures were received from the vendor for approval. Drawings for the coolant-pump tank are being reviewed; those for the fuel-pump tank were released for fabrication.

Metallurgy. — The effects of heat treatment at 1600°F on the mechanical properties of INOR-8 weld metal were studied by using specimens from a highly restrained weldment that had passed the standard weldability test. Stress-rupture tests at 1300°F and stress levels of 27,500 psi indicated the following improvement:

Sample Condition	Rupture Time (hr)	Total Strain (%)
As welded	96	5.5
Heat treated	239	12.5

A series of tensile tests was made on heat-treated specimens of INOR-8 in an effort to gain further understanding of the mechanism responsible for minimum ductility observed in this alloy at intermediate temperatures. Specimens annealed to give a minimum ductility at 1600°F were tested after being held at this temperature for various times. A significant improvement in ductility was observed after the treatment at 1600°F for 1 hr, and increasing time caused a corresponding increase in ductility but only slight changes in tensile strength. Metallographic examination and other tests indicated that this is not a conventional bulk-type aging reaction.

Instrumentation Development. — Testing of the ball-float level transmitter, which was developed for measurement of molten-salt level in the MSRE pump bowl, is continuing. The transmitter has been operated for eight weeks at 900 to 1200°F. The calibration drift has not exceeded 2% of full scale, and there has been no indication of deterioration of the instrument or impairment of its performance.

Seven mineral-insulated Chromel-Alumel thermocouples of the type proposed for use in the MSRE are being furnace-tested to determine the effect of continued operation at high temperatures on their calibration accuracy. The maximum calibration drift observed during 11 weeks of operation at 1200°F was 2°F.

Several mechanical methods of attaching thermocouples to thin-wall tubes on the MSRE radiator were tested. Results indicate that excessive errors will occur unless the junction is covered with insulation. Temperatures measured with uninsulated couples, under condition of full air flow and 1000°F wall temperature, were 213°F low. Insulating the junction reduced this error to 70°F. Both 1/8- and 1/16-in.-OD sheathed materials were tested. No significant reduction in error was obtained by the use of 1/16-in.-OD materials.

Reactor Analysis. — A preliminary analysis of the kinetics of the MSRE was completed, with particular emphasis on the response of the reactor to various reactivity insertions. Situations considered included arbitrary step and ramp insertions of reactivity, as well as insertions which could arise during actual operation of the system. In the latter category were "cold-slug" incidents, filling accidents which resulted in criticality before the core is full, loss of graphite from the core, improper fuel addition, and uncontrolled rod withdrawals. In the case of a

cold-slug accident in which 1-1/2 core volumes of 900°F fuel salt were pumped into the core under normal-flow conditions, with the initial core critical at 1200°F, the mean temperature of the fuel salt would rise less than 100°F above 1200°F; the corresponding maximum pressure rise was about 5 psi.

With the reactor initially at 10 Mw, a pump power failure resulted in a 120°F rise in mean salt temperature. Under such circumstances the secondary salt would freeze in about 2 min if no corrective action were taken; closing of the radiator doors within 30 sec after power failure would prevent the salt from freezing.

REACTOR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. – Continued studies of fission product release during the oxidation of UO_2 to U_3O_8 in air led to a measurement of the diffusion parameter (D') for the U_3O_8 product. A calculation of the energy of activation for the diffusion of rare gases from U_3O_8 was made for the temperature range 500 to 1200°C. The results are reported both as a function of burnup in the irradiated material and as a function of heating time:

Burnup (Mwd/ton)	Heating Time (hr)	Temperature Range (°C)	Activation Energy (kcal/mole*)
1000	4.5	600–1200	35
1000	1.1	500–1200	31.6
4000	4.5	600–1200	40.5
4000	1.1	500–1200	40.7

*Lindner and Matzke report 20 kcal/mole for tracer-level-irradiated U_3O_8.

The values reported here, ranging from about 30 to 40 kcal/mole, as compared with literature values of 20 to 30 for materials irradiated to tracer levels, suggest a slight dependence on burnup level. No clear indication of the same effect in UO_2 has been observed, but re-examination of data on rare-gas diffusion from irradiated UO_2 into helium at temperatures below 1600°C gave an activation energy value of 42 kcal/mole for UO_2 irradiated to the 1000-Mwd/ton level. After a burnup to 4000 Mwd/ton, this value had increased to 60 kcal/mole.

Release of Fission Products on In-Pile Melting of Reactor Fuels. – The fourth and fifth experiments on in-pile melting of UO_2 fuel specimens were conducted at power levels of 110 w/g with 5-min exposures. Complete melting of the specimens was obtained, and UO_2 was deposited on the floor and walls of the melting chamber. The deposited UO_2 had a glassy appearance like that in experiment 3 and in contrast with the crystalline forms observed in experiments 1 and 2.

Experiments 4 and 5 are part of a series of experiments that are to be identical in construction, operation, and analysis. An important objective of this series is to define the reproducibility of results to be expected from the experimental procedures. Operating conditions of the series are: stainless-steel-clad UO_2 specimens; irradiation time before melting, 50 hr; power at melting position, 110 w/g; time at melting point, 5 min; and a helium atmosphere.

Radiochemical analysis of experiments 2 and 3 is 75% complete. On the basis of these incomplete data, there are major differences in the distributions of fission products in these two experiments. These differences can be explained by the facts that experiment 2 was kept molten longer, was exposed without cladding during irradiation before melting, and exhibited UO_2 crystal deposition. Further interpretation of these differences will be possible when analysis of the experiments is complete. Experiments 4 and 5 are being analyzed, and apparatus for experiment 6 is 50% assembled.

Deposition and Removal of Fission Products from Gases. — Investigation of the behavior of radioactive aerosols is continuing, with current emphasis on their characterization and filtration. A number of experiments were performed to develop a suitable method of testing the efficiency of filters for removal of very fine particulate matter carrying radioiodine. Data pertaining to the efficiency with which these particles were filtered by various media are as yet rather sketchy and require further corroboration before reporting; however, the immediate objective — that of development of the method — appears to have been achieved.

In these experiments, particles emanating into an air stream from hot stainless steel, type 347, were exposed to I^{131} vapor and passed through a high-efficiency low-pressure-drop particulate filter with diffusion-channel side streams sampling the air before and after it reached the filter. From the observed diffusion coefficients, obtained from the distribution of radioiodine along the walls of the channels and making use of particle diffusion theory, the I^{131}-labeled particles had a diameter of ~0.003 μ. The use of very fine aluminum oxide particles as an aid in verifying the diffusion-cell technique for radioactive aerosols has been previously mentioned. According to the literature, the diameter of particles similarly prepared was 0.004 μ, which is very close to the value obtained in this work with radioiodine tracer. In both instances, diffusion measurements were used, although the methods for collection and detection of particles were quite different. These aluminum oxide particles have since been examined at ORNL by means of electron microscopy at 68,000×. Their observed diameters were 0.003 to 0.008 μ, representing good agreement with the values obtained from the two types of diffusion measurements.

Reactor Containment Handbook. — A preliminary outline for a reactor containment handbook was prepared. The general philosophy and scope of the text were determined. Existing methods for designing containment systems for the various reactor types will be described, and sufficient information on fundamental processes and appropriate analytical techniques will be included to enable the designer to incorporate new data into the development of improved containment systems.

Nuclear Safety Pilot Plant. — The Nuclear Safety Pilot Plant is intended to serve as an experimental tool for the study of fission product release and transport following the meltdown of irradiated fuel in a model containment vessel. The study will primarily concern the effects of the meltdown of UO_2 in various atmospheres. Processes and methods for the attenuation of fission products, such as deposition, recycle filtration, and nuclei condensation by means of steam injection followed by water sprays, will be evaluated. The fission product capability of the plant will be in excess of 1000 curies.

The heat source will be a plasma torch capable of producing a low-momentum "laminar" flame and of operating in the transfer-arc manner. A furnace capable of handling molten UO_2 is being developed, and tests were performed in which UO_2 was melted by the plasma-flame method. Electron-microscope studies of the resulting particulate matter caught on sample filters showed that the particles were approximately 0.04 to 0.4 μ in diameter. The particles appeared to be agglomerates of basic particles of U_3O_8 approximately 400 to 500 A in diameter. These results are in good agreement with those of previous meltdown experiments performed with induction heating.

Preliminary process flowsheets and layout drawings for the plant were prepared. Basic siting, shielding, and safety criteria work was completed. Instrumentation flowsheets are being prepared, and design and procurement of long-lead-time items have begun.

Reactivity Effects of Small Changes in Fuel Distribution in Pool-Type Reactors. — The last report (ORNL-3269) described the inception of a program of calculation and experiment intended to determine the reactivity effects of small changes in fuel distribution in pool-type research reactors. At that time ten calculations, all utilizing diffusion-theory codes, had been completed. A series of experiments has now been completed in which the fuel density in selected regions of the Bulk Shielding Reactor I was augmented by the addition of special fuel plates to selected fuel elements. Results of the experiments are in significant disagreement with the predictions of the calculations. It has therefore been concluded that diffusion theory is not capable of adequately describing the effects of the small changes under consideration. A test of a one-dimensional transport-theory code, DSN, on a typical problem, however, has resulted in the prediction of radical differences in flux shape in the region of small fuel concentrations. This result has encouraged further calculations, in which a two-dimensional transport-theory code will be used to predict the results of the fuel distributions of the experiments.

Determination of ℓ/β_{eff} by Reactor Noise Analysis. — The random reactivity fluctuations resulting from the random nature of the nuclear processes taking place in a reactor have been utilized, by measuring the spectral density of such fluctuations, to estimate the value of ℓ/β_{eff} (the ratio of the prompt-neutron lifetime to the effective delayed-neutron fraction) for two reactor cores, the Bulk Shielding Reactors I and II. Data were obtained by the use of a wave analyzer constructed of circuits of the analog-computer type over the range 0.05 to 20 cps and by the use of a commercially available wave analyzer for higher frequencies.

The value of ℓ/β_{eff} obtained for the stainless steel BSR-II core was 3.05 ± 0.10 msec; that for the aluminum BSR-I core was 7.93 ± 0.27 msec. These values are in agreement with a previous determination by pulsed-neutron-source methods, which gave 3.2 ± 0.2 msec for the BSR-II and 8.5 ± 0.5 msec for the BSR-I.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Thermal Transient Studies. — Equipment is being procured, constructed, and tested for the investigation of surface thermal instabilities associated with saturated forced-convection boiling of water in a horizontal pipe.

Boiling Studies. — A study of heat transfer inside horizontal, open-ended, electrically heated tubes immersed in a pool of water was undertaken. Thirty-four tests were conducted with tubes vacuum-jacketed externally; the experiments covered three levels of subcooling for each of three tubes of different heated length-to-diameter ratios. Data on the frequency and amplitude of tube-wall temperature oscillations were obtained and are being analyzed.

MARITIME REACTORS PROGRAM

Physics Studies. — The use of a boron burnable poison to extend the life of the NS "Savannah" core was investigated by using a simple burn-up model. The possibility of placing the boron burnable poison in the cladding of fuel elements of the core I type and inserting these elements into a Zircaloy fuel container was considered. Results indicated that six full-power years of life would be possible (within the present control-rod limitation) for a fuel enrichment of about 6 wt % U^{235} and a boron loading of about 250 ppm B^{10} in the cladding. The reactivity lifetime of the core appeared to be fairly insensitive to the initial boron loading. Peak core reactivity occurred after 1-3/4 full-power years of operation. On the basis of these rather promising results, more detailed calculations are being performed.

Iodine Filter Testing. — An experimental program is in progress to determine the efficiency of iodine filters under the conditions of the NS "Savannah" reactor compartment. Twenty-two small-scale charcoal media tests were completed. Of four types of charcoal tested, Pittsburgh BPL and Pittsburgh PCB rated highest in iodine removal efficiency. Three experimental runs on prototype charcoal units manufactured to the same specifications as the charcoal units installed on the ship showed iodine removal efficiencies somewhat less than the desired 99.9%. The iodine removal efficiency was determined both by radiochemical analysis of the entire experimental assembly and by a gas sampling procedure. Preparations were made for in-place testing of the iodine filters presently installed in the reactor compartment ventilation system of the NS "Savannah."

Shielding Survey. — Plans and preparations for the shielding survey of the NS "Savannah" are essentially complete. The detectors and associated electronic equipment were successfully tested in a series of surveys of various types of land-based reactor shields. These surveys also served as a training program in which the nucleus of personnel familiar with operation of the detection equipment was considerably increased. Of the 41 people required for making the NS "Savannah" shielding measurements, all but 16 have had at least a brief familiarization with the equipment. The remainder will be trained at dockside during the week before the survey.

ARMY POWER REACTORS

An interim postirradiation examination was performed on a test fuel assembly which is being irradiated in the SM-1 reactor. The test fuel assembly, designated SM-2B, was prepared by Alco Products, Inc. It is similar in overall dimensions to the SM-1 core I fuel assembly but differs in many details, one of the principal differences being that the fuel plates are attached to the side plates by welding rather than by brazing. Test operation of the SM-2B assembly began in October 1960. In subsequent visual inspections during reactor shutdown periods, marks were noted connecting two welds in one of the side plates and in the welds. Since the possibility of weld failure could not be eliminated with examination equipment available at the reactor site, the fuel assembly was removed to an ORNL hot cell for further examination.

The suspect area was readily located. Exterior surfaces of the element were examined at low magnification (1.4×) with oblique lighting and at higher magnifications (5× to 15×) with a binocular-type microscope and direct lighting. A dye-penetrant examination was made of the suspect areas. It was concluded that the marks were not cracks but merely a series of fine scratches and that the fuel assembly was in generally good condition. Following additional gamma activity and dimensional measurements, the SM-2B assembly was returned to Fort Belvoir for reinsertion into the SM-1 for further testing.

NUCLEAR TECHNOLOGY AND GENERAL SUPPORT

Basic Reactor Shielding. — Two skeleton routines have been written for the OGRE system. One code places gamma-ray histories on magnetic tape for processing at a later time and in any fashion desired. The other code, instead of writing on tape, requires a subroutine to process each collision as it is generated. The first code will be used primarily for research and comparison of tallying procedures. The latter code will be used for production of data.

The latest system for obtaining cross-section data for use in the OGRE system is from data on magnetic tape. This system enables the OGRE codes to have a minimal amount of input data from cards.

A sequence of multilayered infinite-slab codes is being written. One such code written in the older OGRE system (TM-167) was revised recently to correspond to the latest system. These two codes calculate transmitted dose rate due to gamma rays impinging on the slab, either in a given direction or with an isotropic distribution. Another code, which is nearly complete, will incorporate the calculation of transmitted dose rate with the calculation of heating in the slab, as well as the transmitted energy and angular distribution.

Another OGRE system code is being written for the calculation of transport, in an infinite homogeneous medium, of gamma rays which originate from an isotropic point source. The main purpose is to generate air-scattering data similar to those of Lynch et al. (ORNL-2292), but the data will be placed on a computer-oriented medium as well as printed paper.

Some success was achieved in the coupling of a transport code and a multigroup diffusion code to calculate thermal neutron flux. The preliminary investigation showed that the coupled calculations agree much better with experiment than does the multigroup diffusion calculation alone.

Standard data sheets are now available for the neutron transport code RENUPAK; the running of this code is now routine.

Work is continuing on the standard output from transport and diffusion codes. With the present scheme any number of codes may be coupled.

Power Reactor Fuel Processing: Corrosion. – Titanium exposed to Darex dissolver solution for 72 hr at 87°C and containing 0.0025 M $Na_2B_4O_7$ and 0.01 M HBF_4 showed corrosion rates for the vapor, interface, solution, and condenser areas of 3.0–5.5, 53.1–55.6, 90.8–92.6, and 0.02–0.09 mils/month respectively. When the solution also contained 0.04 M Al^{3+}, corresponding rates were 0.62–1.2, 10.6–12.3, 15.3–15.6, and 0.03 mils/month.

Specimens of Ni-o-nel exposed in the vapor above refluxing solutions of 6, 8, 10, 12, and 15.6 M HNO_3 for 648 hr were corroded at overall rates of 1.8–1.9, 3.0–3.4, 6.7–7.2, 20.2–20.5, and > 37 mils/month respectively. Intergranular attack was noted after 168, 120, 120, 48, and 48 hr respectively. In the vapor above refluxing 4 M HNO_3, maximum overall rates for 1344 hr of exposure were 1.3 mils/month for Ni-o-nel vs 0.6 for LCNA. In both materials the onset of grain-boundary attack was between 192 and 360 hr. Condenser specimens showed rates of < 0.1 mil/month in both cases. These results indicate the necessity for potential users of Ni-o-nel to test this material under the exact conditions expected in their process. The results of cyclic Sulfex or Zirflex tests may not be so pessimistic as those reported for tests with nitric acid alone.

Unwelded specimens of type 6061 aluminum and welded specimens of type 309 SCb stainless steel exposed in refluxing 23 M HNO_3–1 M HF for 144 hr were corroded at apparent maximum rates of 0.1 and 0.6 mil/month respectively. Rates were essentially constant over the complete exposure period. All the aluminum specimens gained weight except those exposed in the vapor phase.

Power Reactor Fuel Processing: Solvent Extraction. – The distribution coefficients for thorium between 1 M $Al(NO_3)_3$–0.1 M HNO_3–x M Th and

both 2.5% tributyl phosphate and 2.5% di-sec-butyl phenyl phosphonate (DSBPP) were measured. For solutions containing < 0.65 M Th the coefficient was larger for the TBP system; for feed containing > 0.65 M Th the coefficient was smaller in the TBP system.

In a countercurrent batch extraction of uranium with 2.5% DSBPP in diethylbenzene, decontamination from thorium was 10^4 with four extraction and three scrub stages. One volume of feed with the composition 10 g of Th per liter, 5 g of U per liter, 0.8 M Al, and 0.1 N acid deficient was extracted with 1 volume of solvent, and the loaded solvent was scrubbed with 0.4 volume of 0.8 M Al that was 0.1 N acid deficient.

Power Reactor Fuel Processing: Zirflex Process. — Demonstration of the Zirflex decladding process, including subsequent modified Purex solvent extraction of the core solution, on an engineering scale was completed. In 22 runs with specimens of Zircaloy-clad uranium dioxide pellets irradiated to 168 to 17,700 Mwd per ton of uranium, the cladding was removed by 6 M NH_4F–1 M NH_4NO_3 in 2 to 3 hr. The zirconium end plugs remained. Soluble-uranium and -plutonium losses to the decladding solution averaged about 0.05%. Centrifugation or filtration of the decladding solution is indicated to remove the 1 to 2% of the UO_2 present as fines resulting from the fracture of low-density (93 to 95%) pellets; however, high-density (96%) pellets produced few UO_2 fines.

Approximately 5 hr was required to dissolve the declad UO_2 core material in 4 M HNO_3 to produce a solvent extraction feed of 100 g of U per liter and 3 M HNO_3.

Gamma decontamination factors for the Purex first-cycle uranium and plutonium product streams were 1.3×10^3 and 2.1×10^3 respectively. Pretreatment of the solvent extraction feed with diacetyl monoxime or its degradation product oxalic acid increased these factors two- to tenfold.

Extended re-use of 30% tributyl phosphate in Amsco 125-82 diluent as solvent in a simulation of the extraction-scrub-strip-solvent recovery scheme with 17,000-Mwd/ton feed solutions resulted in the formation of uranium-retaining degradation products in the solvent that could not be removed by the usual solvent recovery procedures of carbonate and nitric acid washing. No degradation was shown when n-dodecane or the commercial equivalent Adakane-A12 was substituted for Amsco 125-82.

Power Reactor Fuel Processing: Shear-Leach Process. — The shear section of the shear-leach mechanical-complex checkout runs, including evaluation of the electrical circuitry for proper articulated operation, was completed. Installation of chemical processing equipment is complete except for insulation of steam and acid lines. Wear studies of the gibs and liners and the stepped blade are still in progress, and operation of the shear and all its auxiliaries continues to be very satisfactory.

Thirteen ORNL Mark I porcelain-filled prototype fuel assemblies were sheared into 1/2-, 5/8-, and 1-1/2-in. sections for further measurements of particle-size distribution and blade wear. The Squarkeen No. 3 stepped blade shows only slight wear after 4271 cuts on the porcelain-filled fuel assemblies. Wear is primarily by rounding of the leading corners of the center step.

Measurements on ~4-kg batches of 3/4-, 1/2-, and 1/4-in. sheared lengths of porcelain-filled elements assembled with both Nicrobraz-50 and Kanigen-brazed spacer ferrules showed no appreciable difference in the particle-size distributions from Nicrobraz-50 or Kanigen-brazed elements. For 1-, 3/4-, 1/2-, and 1/4-in.-long cuts, the fractions of material < 9520 μ in diameter were 11, 16, 33, and 74% respectively; for material < 44 μ, the fractions were 1.5, 2.4, 4.3, and 5.2%.

Results of shear tests on three porcelain-filled Mark I fuel assemblies with 1/8-in.-thick stainless steel tube sheets located 1/2 in. from each end indicate that it may not be advisable to shear fuel assemblies containing tube sheets in the 250-ton prototype shear. In two of the six cuts the shear blade was passed through the tube sheet. In one cut the tube sheet was cut into two pieces, one containing 7 tubes and the other 25 tubes. The larger chunk jammed between the adjustable stop and inner gag on the reverse stroke of the ram, resulting in some deformation of the adjustable stop.

Radioactive Waste Treatment and Disposal: High-Activity Waste Calcination. – The thermal conductivities of a lead phosphate glass were 1.05 and 1.60 Btu hr^{-1} ft^{-1} $(°F)^{-1}$ at 300 and 1050°F respectively. The production of this glass on a semiengineering scale was accompanied by volatilization of 12.5% of the original ruthenium and physical entrainment of < 0.02% of the promethium as determined from counting Ru^{106} and Pm^{147} tracers.

Simulated Darex waste solution was incorporated into glassy and microcrystalline phosphate and borophosphate solids with softening points of 850 to 900°C and densities of 2.73 to 3.75 g/cc, and containing 13.4 to 32.3 wt % waste oxides, representing volume reductions of 2.9 to 6.6.

Decreasing the pressure from 750 to 570 mm Hg had little effect on the relative volatilities of ruthenium from 6 M HNO_3. Addition of 0.1 M H_3PO_3 to nitrate solutions of ruthenium chloride and ruthenium nitrosohydroxide with and without salts occurring in waste solutions decreased the ruthenium distillation by factors varying from 2920 for 6 M HNO_3 containing ruthenium nitrosohydroxide to 38.3 for simulated Darex waste containing ruthenium chloride.

Process flowsheets and an equipment layout were completed for the Idaho Chemical Processing Plant Pot Calcination Pilot Plant. A mercury trap for removal of mercury compounds from the calciner off-gas was designed and fabricated, and several welded closures for sealing pots were leak-tested at the Georgia Nuclear Laboratory; the leak rate was less than 0.2 std cc of helium per year.

Radioactive Waste Treatment and Disposal: Low-Activity Wastes. – In the ion exchange process for removing radioactive fission products from waste, scavenging was improved by adding $CaCO_3$ to the flash mixer, by either adding 25 ppm of $Ca(OH)_2$ or recirculating sludge from the clarifier. Residual hardness was < 18 ppm at 24°C. Increasing the temperature from 4 to 35°C had little effect on the caustic-copperas treatment but decreased the residual hardness from 70 to 7 ppm in the lime-caustic treatment. Results with a laboratory-scale clarifier showed that equipment size could be reduced by a factor of 10 to 20 over the present pilot plant equipment.

The results of five demonstration runs completed in the pilot plant with ORNL process waste water as feed were essentially the same as those obtained previously: Sr^{90} decontamination factor, 2000 to 12,000; Cs^{137} decontamination factor, 77 to 3000. Plant effluent solutions contained < 3% of current MPC_W values for all isotopes at the 2000-bed-volume level (56,000 gal). A resin-column isotope breakthrough run showed an Sr^{90} release to plant effluent after 2916 bed volumes.

Radioactive Waste Treatment and Disposal: Disposal in Natural Salt Formations. — Four plastic-flow measurement stations were installed in the Carey Salt Company mine at Hutchinson, Kansas. Each station consists of nine gages installed in such a way as to measure both floor and ceiling movement and movement of the columns. The stations are installed in areas of varying age in development of the mine. Two additional gages remain to be installed. An array of 20 holes for the study of effect of high temperatures on rock salt in situ was successfully drilled without the use of a liquid drilling medium. Samples of dry shale from the holes contained 10 to 12% water.

Radioactive Waste Treatment and Disposal: Engineering, Economic, and Hazards Evaluation. — A preliminary conceptual design and cost estimate for a solids disposal facility in salt indicated costs of 6×10^{-3} to 30×10^{-3} mill/kwhr (electrical). Of the total costs, 60 to 85% was attributable to salt removal, indicating that the best possible estimates of this item will be required for the more detailed final study.

Heat transfer calculations of shipping containers for pot-calcined wastes of various ages showed that as many as thirty-six 6-in.-diam pots of calcined acidic Purex waste can be shipped in a single finned carrier after 2.4 yr of decay without mechanical cooling en route. Four 24-in.-diam pots of acidic Purex waste can be shipped, per carrier, after 11 yr of decay.

Radioactive Waste Treatment and Disposal: Clinch River Study. — During periods free from storm runoff in the summer months, power waves in the study reach of Clinch River are classified as uniformly progressive flow. For this type of flow the storage of water in the study reach is conditioned equally by the inflow and outflow rates. By expressing the storage as a weighted function of these rates, the variations in flow due to power releases in the Clinch River above Centers Ferry, near Kingston, Tennessee, may be determined from flow records obtained at the upstream gaging station on Clinch River near Scarboro, Tennessee.

Stable chemical analyses of water samples taken during base flow measurements in September 1961 reveal a marked difference in quality of water between streams draining coal-bearing clastic rocks of the Cumberland escarpment and streams draining predominantly carbonate rocks of the Ridge and Valley province in which Oak Ridge National Laboratory is located. Streams draining the Cumberland escarpment contain a greater proportion of sulfate, sodium, and chloride, and have higher pH's than do streams draining the Ridge and Valley province.

Radioactive Waste Treatment and Disposal: Fundamental Studies of Minerals. — The cesium exchange properties of vermiculite were improved by collapsing the vermiculite lattice with ammonium ions or with one of

the heavy alkali-metal cations. Pretreatment of the vermiculite with potassium increased the number of exchange sites at the edges of collapsed basal spacings, where cesium can be incorporated into the lattice by "edge fixation." This increased the cesium K_d in 0.5 M $NaNO_3$ by a factor of 2.7. Addition of NH_4^+, Cs^+, Rb^+, or K^+ to the influent caused "interlayer fixation" of cesium, which resulted from collapse of the lattice and physical entrapment of the cesium sorbed at the basal surface exchange sites. In this case the cesium K_d was increased by a factor of about 4.4.

Dissolver Solution Analyses. — The development of a procedure for determining uranium coulometrically after its extraction into a cyclohexane solution of tri-n-octylphosphine oxide and subsequent back-extraction into a solution of $(NH_4)_2SO_4$ was mentioned in prior progress reports. The application of this procedure to the common types of dissolver solutions is being studied, and in this connection has been applied to the analysis of synthetic dissolver solutions typical of the following reactor fuels:

Fuel Type	Dissolver Solution
Uranium-aluminum	Al, U, Hg in HNO_3
NS "Savannah" and APPR	U, Fe, Ni, Cr in HNO_3 and HCl
Decladded Fuel	
Consolidated Edison Reactor	Th, U, Al, F in HNO_3
Consumers Power Reactor	U, Mo in HNO_3
Fermi Reactor	U, Mo, Fe in HNO_3

The basic procedure, or minor modifications thereof, has been utilized satisfactorily for these analyses. The relative standard deviation is approximately 0.2% at a concentration level of 5 to 10 mg of uranium, with an absolute error of the order of ±0.1%. The application of this procedure to the zirconium-containing and to the more advanced fuels will be made in the future.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Electronuclear Machines: The Mc^2 Cyclotron. — A new conceptual design for the Mc^2 facility is almost complete. The experimental area is in three separate rooms designated for proton, pion, and muon experiments respectively. The beams enter the individual rooms from a heavily shielded corridor containing the beam transport and analysis system. It is estimated that 35 to 40 ft of ordinary concrete will adequately shield the proton areas, but that 15 ft will suffice in the secondary-particle facilities.

New parameters for the Mc^2 cyclotron magnet have been established. The central magnetic field will be 4500 gauss, and the average radius of the beam at extraction will be 19.1 ft. A 0.15-scale model of three magnet sectors has been built. Preliminary measurements indicate that the iron location and the ampere turn distribution are approximately correct.

The rf cavity systems, which will provide the accelerating voltage for the Mc^2 cyclotron, have been studied both theoretically and experimentally. Cavities were designed to minimize the power for a given gap voltage and to provide uniform voltage along the lips. Scale models of two suitable cavity types, the TEM and TE or coaxial, were built and tested. The results are in agreement with the design calculations and indicate that the rf power for the Mc^2 cyclotron will be in the 500- to 800-kw range.

Basic Studies of Cryogenic Electromagnets. — The technique for evaluating superconductors in the high-field magnet at Y-12 is now standardized and results in reproducible I-H curves. The sample, leads, and joints are rigidly mounted in an epoxy casting. Short-wire tests (2 to 3 in. long) for materials evaluation are replaced by 4- to 5-ft samples wound noninductively. The same I-H curves result, with a sensitivity approximately proportional to the increased length. Inductive samples 100 ft long, producing up to 12 kilogauss self-field, have shown that both 25 and 33% zirconium content in hard-drawn Nb-Zr wire carry reduced currents at low fields, higher currents at midfields (20 to 50 kilogauss), and reach a critical field of 71 and 81 kilogauss respectively. Niobium—50% zirconium wire, 0.010 in. in diameter, carries 5 amp at 90 kilogauss. Magnets with Nb-Zr wire producing fields of 100 kilogauss appear possible.

Physics of Fission. — The pulse-height distribution of correlated fragment pairs from thermal-neutron-induced fission of U^{235} has been measured by using two matched 4-cm^2 silicon surface-barrier detectors and a 128 × 128 channel correlation recorder. The uncorrelated pulse-height spectrum exhibits a peak-to-valley ratio of 20:1 for both detectors. The pulse-height contours resulting from a correlation run containing $> 10^6$ events have been shown; these data have been normalized to energies (E) obtained from time-of-flight measurements [Phys. Rev. 110, 476 (1958)] to determine correlated energy spectra, mass distributions, and other pertinent correlations. The pulse-height (PH) response of surface barriers to fission fragments may be represented [Bull. Am. Phys. Soc. 6, 3 (1961)] by $E = a \times PH + \delta$, where a and δ are empirically determined factors

which depend on the time-of-flight data employed. The effect of these normalizations on the data, and the influence of prompt-neutron emission on the determination of mass distributions are being studied.

Scintillation Spectrometry and Instrument Development. — Singles and coincidence gamma-ray spectra of Cs^{132} have been investigated with scintillation spectrometers. The energies in kev (and relative intensities) of the gamma rays observed are 464 (21 ± 5), 509 (5.1 ± 1.5), 511 (12 ± 4), 569 (2.7 ± 0.8), 631 (8.5 ± 1.8), 669 (1000 ± 30), 1035 (1.6 ± 0.3), 1139 (5.2 ± 0.5), 1304 (0.9 ± 0.5), 1319 (6.2 ± 0.6), and 2000 (0.53 ± 0.06). From these studies it is proposed that Cs^{132} decays to levels at 669 ± 4, 1300 ± 10, 1808 ± 13, and 1988 ± 12 kev in Xe^{132} and at 464 ± 5 and 1034 ± 9 kev in Ba^{132}. The angular correlations of four gamma-ray cascades have been measured. They lead to spin assignments of 2 for the 1300-, 1988-, and 1034-kev levels, and of 2 or 3 for the 1808-kev level. The correlations also give values of 10, 0.0060, and ≦200, respectively, for the ratio of quadrupole to dipole radiation for the 631- and 1319-kev transitions in Xe^{132} and for the 569-kev transition in Ba^{132}. The predominantly quadrupole character of the transitions between the second and first spin-2 levels is attributed to the collective behavior of the nuclei. From the ratio of electron capture to positron emission in the decay to the 669-kev level, the energy separation between Cs^{132} and Xe^{132} was estimated to be 2.15 Mev.

High-Voltage Experimental Program: Excitation of Collective States in Sn and Fe by Inelastic Scattering of 14-Mev Neutrons. — A deuteron beam is chopped into 30-nanosecond bunches at a repetition rate of 220 nanoseconds in the terminal of a 200-kv Cockcroft-Walton accelerator. Klystron bunching is used after acceleration to produce a deuteron pulse at the target which is less than 2 nanoseconds in width with a peak current greater than 3 ma. To study inelastic scattering of tin and iron, 14-Mev neutrons from the d-t reaction have been used. A ring geometry is used. Neutron energies are determined by time-of-flight with a 9-m flight path. With tin there is clear evidence for the excitation of the quadrupole states at 1.1 ± 0.1 Mev. The differential cross section increases from 2.6 ± 0.5 mb/steradian at 81° to 6.5 ± 1.2 mb/steradian at 32°. The shape and the magnitude of the cross section agree well with a theoretical calculation of the excitation of the quadrupole states. In addition to the quadrupole state, there is a somewhat stronger excitation of a state at 2.15 ± 0.1 Mev, which may be the octupole state. With the iron scatterer, the 845-kev first 2^+ state is clearly seen. At some angles it is comparable to the elastic-scattering cross section.

High-Voltage Experimental Program: Countdown System for Pulsed Van de Graaff Accelerator. — The optimum repetition rate of pulsed Van de Graaff beams varies quite widely from one experiment to another. For example, whereas fast-neutron (Mev-range) experiments call for rates in the several-megacycle range, recent efforts in measuring fission parameters in the kev range call for a repetition rate of 100 kc/sec. Other neutron experiments are optimum for intermediate frequencies. All experiments, however, call for short burst duration ($\lesssim$ 5 nanoseconds). A system for reducing the pulse repetition rate from 1 Mc to 500, 250, or 125 kc has been developed. This system uses a pair of electrostatic deflector plates

immediately above the analyzing magnet of a 3-Mv Van de Graaff accelerator to sweep the ion beam across a slit at the required frequency. The deflector plates are biased to allow the transmission of only one pulse in each cycle. A tuned ferrite-core antenna coil located in the bottom of the pressure tank picks up a signal from the pulsing equipment in the terminal. This signal is electronically converted to the required frequency and amplitude. Means for selecting the required frequency and for phase synchronization with the incoming pulse are provided.

Multiparticle Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — Experiments with 15-hr Ir^{185} produced by the $Os^{186}(p,2n)Ir^{185}$ reaction indicate the existence of rotational structures in Os^{185} based on orbitals $1/2^-$ [510] and $3/2^-$ [512], as was observed in the isotone W^{183}. The most intense transitions in $Ir^{185} \rightarrow Os^{185}$ decay populate the five levels of the $1/2^-$ [510] band based on ground. The three levels of the $3/2^-$ [512] band based at 127.8 kev are established by 14 de-exciting transitions of M1 and E2 character, or a mixture of these. An analysis of the rotational-energy constants indicates a stronger particle-rotation coupling of the above bands than was observed in W^{183}, which is consistent with the smaller energy separation in Os^{185}.

Electronuclear Machines: The Isochronous Cyclotron. — An 8-Mev beam of protons was brought out to full radius on March 18, 1962. Energy determination was by means of range measurement in stacked copper foils. This initial beam was obtained by using a 5-kw test rf amplifier pending completion of the 350-kw unit. The beam made about 450 turns, as compared with the 375 turns required for 75-Mev protons. There is little attenuation after the beam leaves the central region.

A unique device was developed and tested for measuring the phase of the beam with respect to the accelerating voltage without intercepting the beam. This is a capacitive pickup that detects the beam pulses and can be set to any radius in the machine. The output is displayed on an oscilloscope triggered from the accelerating voltage. The position of the beam pulse on the scope trace thus indicates phase angle. The shape of the magnetic field can be adjusted by the circular-trimming-coil currents, with minimum phase shift as a criterion.

The electrostatic deflector-septum assembly has been installed, and extraction studies are now in progress.

Stable Isotopes Development. — The nuclides Ne^{22} and Ar^{36} have been enriched to > 99.95%. A column for the separation of small quantities of Xe^{124} from normal xenon is in operation. The C^{13} thermal diffusion plant has been pressure-tested and is ready for operation.

Special Separations. — Approximately 32 g of Np^{237} was purified by an anion exchange resin from 7 M HCl solution after reduction of the neptunium with sulfurous acid. This procedure reduced the plutonium content from 6000 to 400 ppm. However, due to the necessity of converting to a chloride media from the nitrate, the method is lengthy.

Exploratory work on electroplating alpha materials from aqueous solutions has produced the following results:

1. Uranium up to 0.8 mg/cm^2 was plated on a titanium disk and on the inside of a nickel tube 1/2 in. in inside diameter. It was also plated on an aluminum backing, but could not be plated on magnesium.

2. Neptunium-237 up to 0.1 mg/cm^2 was plated on platinum and nickel.
3. Americium-241 in μg amounts was plated on platinum and nickel. The rate of plating proved to be much more rapid on nickel than on platinum.

CHEMISTRY RESEARCH

Thorium Oxide Studies. – Irradiation of experimental assembly C-43-6B, inserted in the LITR on June 15, 1961, at 2.5×10^{13} neutrons cm^{-2} sec^{-1} is continuing. In this experiment a variety of thorium oxide preparations under D_2O in a single autoclave are separated from each other by thin-walled stainless steel tubes. The preparations include the code P-82 thoria pellets, sintered thoria compacts (prepared by a different method than for the P-82 pellets), fired sol-gel thoria particles, fired sol-gel thoria spheres (44 to 74 μ), arc-fused thoria fragments (44 to 74 μ), and 1600°C-fired thoria powder (DT-46). The expected duration is 12 months of irradiation at full reactor power, at which time the U^{233} content will be 0.5% and the oxides will have received an exposure equivalent to that received in a blanket cycle.

Both wet and dry irradiation appeared to harden the surface of thoria pellets, but when the surface shell was worn away, wear rates were similar to those for unirradiated materials. In a standard spouting-bed test, wet (260°C) irradiated P-82 thoria pellets showed a tenfold increase in wear resistance (≦0.01% wt loss/hr) over the control or original pellets in the first 2 hr, but in the next 3 hr, wear rates of the irradiated and unirradiated pellets (original, not autoclaved) were essentially the same, 0.1% wt loss/hr. For the remaining 9 hr the wear rates of the irradiated pellets were about twice as high as those of the original, that is, 0.5% wt loss/hr. The weight loss of dry-irradiated (3 months) pellets during the first hour was 0.03%. During the next 3 hr the average weight loss was 0.25%/hr, and for the next 9 hr, 0.32%/hr. The differences between the wet- and dry-irradiated pellets probably reflect a wet-autoclaving treatment rather than a radiation effect.

Equipment Decontamination. – Oxalate-peroxide solutions buffered with acetate were not unduly corrosive to carbon steel (0.007 mil/hr at pH 4.0, 95°C) when used as a spray decontaminant, but acetic acid vapors were corrosive to metal not contacted by the spray. Although citrate buffer was corrosive when used in too high a ratio to the hydrogen peroxide (up to 50 mils/hr in 0.4 M oxalate–0.4 M peroxide–0.4 M citrate at pH 4.0, 95°C), a citrate-containing solution was developed which had a maximum corrosion rate of 0.003 mil/hr for at least 12 hr at 95°C and then passed in 1 hr through a maximum corrosion peak of 0.16 mil/hr. A commonly used alkaline descaling formula, basic Versene with triethanolamine, was as effective a decontaminant for Zr^{95}-Nb^{95}, and as ineffective for ruthenium, as was an alkaline proprietary reagent. Oxalate-peroxides are effective on both Zr^{95}-Nb^{95} and ruthenium, can be handled more conveniently, and present fewer problems in waste disposal.

Protactinium Chemistry. – Protactinium distribution coefficients between aqueous sulfuric acid solutions and N-benzylheptadecylamine dissolved in diethylbenzene were determined under conditions of varying amine

and acid concentrations. The coefficients increased approximately linearly with amine concentration up to about 0.1 N, but less rapidly at higher concentrations. They decreased with increasing sulfuric acid concentration, the decrease being proportional to the 1.5 to 2.0 power of the sulfuric acid concentration between 3.3 and 8 N, and to the 6 to 8 power in sulfuric acid concentrations 9 to 12.5 N. The change in dependence on acid concentration was quite sharp. Distribution coefficients with 0.01 N amine ranged from 147 in 3.3 N H_2SO_4 to 1.82 in 12.4 N acid.

Radiation Chemistry of Organic Moderators. — The pyrolysis of pure biphenyl for 48 hr at 425°C has produced lower yields of essentially the same products reported previously (ORNL-3269, p 25) for pyrolysis at 490°C. However, increased sensitivity in the gas analyses has shown the presence of trace amounts of hydrocarbons heretofore undetected.

The gaseous decomposition products totaled about 500 μg per experiment and consisted of approximately 80% hydrogen, 15% methane, and 2% propane, along with trace quantities of ethane, acetylene, and three other components as yet unidentified. Benzene continues to be the sole component detected in the intermediate-boiling fraction. The polymeric high-boiling fraction of the decomposition products was shown to be composed of approximately 1% o-terphenyl, 35% m-terphenyl, 30% p-terphenyl, 11% m-quaterphenyl and/or 1,3,5-triphenylbenzene, and 12% m,p-quaterphenyl. About 11% of the material remains unidentified, but is known to consist of three different components.

The formation of each of the polymeric species identified requires the simultaneous production of hydrogen gas, whereas both the synthesis of benzene and the degradation of the benzene ring to form the hydrocarbon gases can only be accomplished by the consumption of hydrogen. With hydrogen by far the major gas constituent and no evidence for carbon formation, it appears that the polymerization reaction path predominates over the degradation path during the pyrolysis of pure biphenyl at 425°C.

Instrumentation for Analytical Chemistry Research. — An equation was derived for simple, reversible, diffusion-controlled dc polarographic processes which expresses the second derivative with respect to time of the diffusion current as a function of the effective potential difference between the polarized and reference electrodes. It can be shown from the equation for the third derivative with respect to time of the diffusion current that the two peaks in the second-derivative polarogram occur at $E_{1/2} \pm 1.32\ RT/nF$ volts. The ordinate of a second-derivative polarogram at any point is directly proportional to diffusion current (i_d), and the height of each peak is directly proportional to i_d, n^2, and $(dE/dt)^2$, where dE/dt is the constant, controlled, effective potential scan rate.

A second-derivative computer was designed and was utilized in recording second-derivative dc polarograms with the ORNL model Q-1988-ES controlled-potential and derivative polarograph. The forms of experimental polarograms have been observed to compare very well with those predicted from the equations. Comparisons have been made by means of the theoretical equations of the relative resolutions of regular, first-derivative, and second-derivative dc polarograms. While the resolution of second-derivative polarograms is better than that of first-derivative polarograms, the noise level is higher. This improvement in resolution is

seldom sufficient, however, to eliminate entirely the overlapping that may occur in portions of wide ranges of concentration ratios of polarograms which have nearly the same half-wave potential. If overlapped polarograms must be resolved mathematically, then it is more advantageous to work with first-derivative than with regular or second-derivative polarograms.

Aqueous Solution Chemistry. — Spectrophotometric measurements were carried out on hydrolyzed U(VI) solutions in chloride and perchlorate media. The results appear to be consistent with the hydrolysis schemes earlier proposed on the basis of equilibrium ultracentrifugation and acidity measurements; that is, the most important species in perchlorate media seem to be $(UO_2)_2(OH)_2^{2+}$ and $(UO_2)_3(OH)_5^{+}$, while in chloride media an additional species, $(UO_2)_3(OH)_4^{2+}$, seems to be present in substantial amounts.

Radiation Chemistry. — The irradiation of silica gel upon which certain polynuclear aromatic compounds are adsorbed produces colors which suggest the presence of ionic intermediates. In ten of sixteen compounds, such coloration was nearly identical with that produced by adsorption of the same compounds on cracking catalysts, where these compounds are thought to be adsorbed as ions. For two cases, naphthalene and triphenylmethane, this color identity was established by reflection spectrophotometry.

High-Temperature and Structural Chemistry. — The electrical conductivities of solutions of calcium metal in calcium chloride, bromide, and iodide were found at saturation (3.0, 2.3, and 3.8 mole % Ca respectively) to assume values 17, 42, and 36% higher than the corresponding specific conductivities of the pure dihalides. Interestingly, the rates of increase in conductance decrease with increasing metal concentration in a way similar to that in the sodium-sodium halide systems. This may be given a somewhat similar interpretation, namely, that the electrons are trapped in pairs, probably not in diatomic metal molecules, Ca_2, but in the covalent single bonds of slightly stable Ca_2^{2+} molecule ions, which are somewhat analogous to mercurous ions, Hg_2^{2+}.

Transuranium Elements. — In scouting tests on methods for separating americium and curium, O_3 passed through a solution of 3 M K_2CO_3 in which americium and curium hydroxides had been dissolved rapidly oxidized the americium to AmO_2^{2+}, which formed an intense reddish-brown complex. On standing overnight, the americium was reduced to AmO_2^{+} and precipitated as an insoluble carbonate that contained 80% of the americium and carried < 5% of the curium.

Distribution coefficients were about 50 for Ru^{4+} and 0.4 for Ru^{3+} in the system Alamine-336—diethylbenzene vs 11 M LiCl. Between this organic and 6 M HCl, which may be used as a strip, values were 6 for Ru^{4+} and 0.02 for Ru^{3+}. Ruthenium can exist in both valence states in LiCl and HCl solutions.

The extraction positions of californium-einsteinium and fermium were redetermined in the system 30% Alamine-336—HCl—diethylbenzene vs 11 M LiCl. Results agreed with those reported in ORNL-3185, the positions being Cf > Fm > Es.

The extraction positions of einsteinium and fermium in 1 M 2-ethylhexylphenylphosphonic acid—diethylbenzene vs 2 M HCl were Fm > Es > Cf, the same as reported in ORNL-3273.

Uranium-232 Preparation. — Four products containing a total of 32.7 mg of U^{232} were prepared by irradiating 7.3 g and 40.8 g of Pa^{231} in the ORR and processing each after 45 hr of decay and again after 80 to 90 hr of decay. An isotopic analysis has been obtained for only one product, which contained 130 ppm U^{233}, 90 ppm U^{235}, and 12,200 ppm U^{238}.

The irradiated Pa^{231} in HCl-HF solution was processed to separate uranium and protactinium. The Pa^{231} irradiated for 10 hr at a flux of 9×10^{13} neutrons cm^{-2} sec^{-1} and decayed for 40 hr read 6000 r/hr at about 3 ft. The radiation was primarily from Pa^{232} and decayed with the Pa^{232} half-life of 1.32 days.

The uranium was further purified by either a second anion exchange cycle or by tertiary amine extraction from HCl-HF solutions, with conversion to a nitrate system and extraction of uranium into tributyl phosphate as the final purification step. Assay of the final products by gross alpha and alpha energy determinations showed U^{232} providing > 96% of the gross alpha radiation. Other alpha activity was primarily from U^{232} daughters.

Target Preparation. — Foils having a thickness of ~10 mg/cm^2 and containing 50% tungsten powder in a copper matrix were fabricated by rolling. The tungsten powder was contained in the matrix as a suspension, not in alloy or solution form.

A new electron beam gun having a 3-kw output and an associated vacuum chamber was installed and was successfully employed to weld tantalum sheets and to evaporate refractory material such as boron. By using this equipment, Lu_2O_3 was evaporated onto a glass plate; the film was found to be transparent to light.

METALLURGY AND MATERIALS RESEARCH

Fundamental Research in X-Ray Diffraction. — The diffraction pattern of neutron-irradiated BeO exhibits unusual line shifts not explainable in terms of simple lattice expansion, as well as selective line broadening. Certain classes of reflections also show asymmetric line contours. The general features of this pattern have been explained by a model of the damaged material in which the interplanar spacing of about 5% of the hexagonal planes are expanded by approximately 40%, presumably by the selective accumulation of radiation debris.

One of the features of the diffraction pattern not explained by this model is that certain lines at large scattering angles are significantly broader than predicted. This difficulty has now been overcome by modifying the model to allow for a distribution of values of the interplanar expansion about its mean of 40%.

Fundamental Investigation of Radiation Effects in Solids: Dielectric Measurements in Irradiated Glasses. — Dielectric constants and dielectric loss tangents have been measured before and after isotropic gamma- and beta-ray irradiation of five types of glass in the frequency range 50 cps to 10 kc. Radiation has little effect on the dielectric constant. The increase in the dielectric loss is largest at 50 cps, decreasing monotonically to 10 kc. There is an indication of a maximum at a frequency

less than 50 cps. The increase in loss after irradiation decays with time to approximately the original value. The decay time varies with the type of glass. A similar increase and decay of the loss tangent is observed after certain heat treatments in those glasses which show the largest change in their loss tangents with irradiation. The largest change in the loss tangent was observed in a lead glass; smaller changes were observed in a phosphate and a crown glass; little change was observed in silica glass and in Pyrex.

The dc polarization of these glasses has been measured before and after Co^{60} gamma-ray irradiation. The polarization current could be resolved into four processes with four distinctive relaxation times. These relaxation times appear to be a bulk property of the various glasses. Three of the relaxation times are changed only slightly by the irradiation; however, the fourth relaxation time increased by a factor of ~2. The apparent concentration of "dipoles" contributing to each of the relaxation processes was enhanced by a factor of ~6 by a dose of 10^6 r of Co^{60} gamma rays. The glasses in which the largest change in dielectric loss is observed after irradiation are also the ones in which a discharge (Lichtenberg patterns) can be produced.

Fundamental Investigation of Radiation Effects in Solids: Effect of Delta Rays on the Scintillation Process in Alkali Halides. — A theoretical study of the effect of delta rays on the scintillation process in alkali halides was completed. This investigation is part of a continuing study of the scintillation mechanism in inorganic crystals. The scintillation response of NaI(Tℓ) to heavy nuclei (B^{10} through Ne^{20}) has been examined experimentally elsewhere, and it was found that the luminescence efficiency is not a function of dE/dx alone, but is instead composed of a series of discrete functions, one for each incident particle. In the present investigation these results were analyzed, with attention to the effect of high-energy secondary electrons (delta rays) produced by the primary particle. Results of this analysis are consistent with experiment and are supported by additional calculations of electron energy losses.

Fundamental Investigation of Radiation Effects in Solids: Photon- and Thermal-Neutron-Induced Defects in Semiconductors. — The rate of introduction of interstitial atoms and lattice vacancies (Frenkel-type defects) in n-type germanium and silicon is comparable for (1) thermal-neutron (n,γ) recoil damage and (2) cadmium capture gamma-ray-induced defects in samples shielded against thermal neutrons with cadmium metal. It is of interest that (1) can be effectively eliminated by using B_4C-impregnated plastic as a shield. This also serves to effectively eliminate (2), since the boron emits only a soft (0.42-Mev) gamma ray and an easily absorbed alpha particle.

CONTROLLED THERMONUCLEAR RESEARCH

Plasma Physics and Arc Research. — A broader and basic experimental approach that may give a quick, qualitative indication of the relative stabilities of various magnetic arrangements appears possible with recently developed techniques. In plasmas formed with a directed stream of neutrals, ions, or electrons, a type of instability may arise from,

or be enhanced by, the stream traversing the plasma. Injection into a mirror magnetic geometry of ions random in energy and direction has permitted greater density accumulation before turbulence appears. In initial experiments with the pressure-gradient arc as the injector into a mirror magnetic geometry, low-energy plasmas are formed with charge exchange and scattering as the principal loss mechanisms. If no dissipative turbulence in a helium plasma (atomic ions in an atomic gas) is assumed, the scattering and charge-exchange cross sections should play equal roles in dissipating the plasma. Experimental agreement with the calculated e-folding time of the plasma decay was excellent. Helium plasmas with 20-ev ions, densities of 10^9 ions/cm^3, and a decay τ of ~0.5 msec have been observed. Deuterium plasmas with 340-ev ions, densities of 2×10^{11} ions/cm^3, and a decay τ of ~0.5 sec have been observed. Again, the experimental τ for deuterons is in general agreement with the calculated value, since the charge-exchange cross section for atomic ions in a molecular gas approaches zero at temperatures considerably above thermal (340 ev for deuterons in deuterium).

DCX-1 Facility. – Measurements were continued of the energy distributions of energetic hydrogen atoms escaping the plasma region as a result of electron capture collisions. A silicon barrier detector with a wide acceptance angle (about 45°) was mounted on the median plane and was employed in conjunction with a 256-channel analyzer. Data have been accumulated for various conditions of pressure and H_2^+ beam under equilibrium conditions and during buildup and decay intervals. Analyses of the data are not yet complete, but in general the peaks of the derived proton distributions remain near 300 kev, and as the H_2^+ beam is increased, the distributions become broader. For example, characteristic proton spectra for equilibrium conditions at 1×10^{-7} torr have a full width at half maximum of about 20 kev at 0.5 ma of H_2^+ and about 50 kev at 5 ma. The variations with pressure are not uniform; the broadest distributions appear near 1×10^{-7} torr.

Radio-frequency measurements in the near field of the proton cyclotron radiation have been extended to pressures of about 10^{-9} torr. At high pressures (about 10^{-7} and 10^{-6} torr) the coherent signals are continuous, are strongly modulated at frequencies in the order of 100 kc, and are confined to narrow bands about harmonics of the proton cyclotron frequency. As the pressure is lowered, the signals gradually become more discontinuous and the frequency bands become wider. At the lowest pressures, the rf appears in bursts at intervals of about 1/2 to 1 sec, with durations of about 50 to 100 msec; and the fundamental band, which was about 1 Mc wide at high pressure, extends over several megacycles.

Use of the lithium ion beam as a plasma potential probe has shown that the potential increases and continues to fluctuate erratically as the pressure is lowered near 10^{-9} torr. With 8.5 ma of H_2^+ injected at 5×10^{-9} torr, the potential usually fluctuates between 200 and 750 v, but it has been observed to be as high as 2 kv. Lower H_2^+ currents produce lower potentials and smaller ranges of fluctuations. For example, with 1 ma of H_2^+ at 3×10^{-9} torr, the potential was observed to fluctuate between 140 and 270 v.

A 12-unit neutral-particle detector which extended across the entire axial length of the innermost vacuum system was installed in the south detector port. Studies have shown that the signals to these detectors

have three components: (1) a fast component due to neutrals from dissociation of the incoming H_2^+ beam which can be suppressed by a suitably placed shield; (2) a second fast component which cannot be suppressed in this manner and whose origin is not clear; and (3) the usual long-time-constant component. The usual curves of $1/\tau$ vs pressure have been measured for this slowly decaying component. They show reasonable agreement with values calculated from the cross section for charge exchange with 300-kev protons (experimental values from 50 to 80% of the calculated values at 2×10^{-9} and 1.5×10^{-7} torr respectively) and show no dependence on injected current for H_2^+ beam values of 1.25, 5.0, and 8.3 ma. The pressure response of the total signal to the 12 detectors appears reasonable in the cases of 1.25 and 5.0 ma of H_2^+, but at the lowest pressures, no increase in total signal was noted when increasing the current from 5.0 to 8.3 ma. The profiles obtained by plotting the detector signals as a function of the displacement of the detector from the median plane are very sensitive to the H_2^+ current at low pressures. The full width at half maximum of profiles measured with 8.4 ma of H_2^+ are 1-1/2 and 8 in. for pressures of 1.3×10^{-7} and 3.2×10^{-9} torr respectively; and for 1.25 ma of H_2^+ they are 1-1/4 and 1-3/4 in. for pressures of 1.4×10^{-7} and 1.8×10^{-9} torr.

A 15-hr bakeout with superheated steam and repeated thermal cycling of the liquid-nitrogen-cooled surfaces produced a base pressure in the liner region of about 7.5×10^{-10} torr. The H_2^+ beam was not being injected.

DCX-2 Facility. — Shakedown tests are continuing. Measurement of the internal field in the injection-snout canal showed the presence of a region of strong transverse field as well as a strong longitudinal field. The region was located at the edge of the machine and was found to be due to an unexpectedly large radial field at that place. A correcting coil to increase the longitudinal compensation was constructed. Operation now shows the expected very small transverse fields which will allow injection of the beam at full magnetic field. While the correcting coil was being constructed, tests of radio-frequency diagnostic equipment were performed by use of the argon arc as an ion source. Many ion cyclotron emission and absorption lines were observed, including those of H^+ and H_2^+. The radiant lower measurements on the arc were extended. Titanium evaporator tests were made which gave evaporation rates of 1/2 g/hr for each evaporator. Pressures obtained were about 2×10^{-8} mm Hg.

Vacuum. — Tests of diffusion-pump-fluid decomposition products were made on three oils. A dioctyl phthalate (Octoil), a mixed hydrocarbon oil (Convoil), and a pentaphenyl ether (Monsanto Chemical Company, OS-124) were used in a 2-in. fractionating diffusion pump. Only the pentaphenyl ether yielded no decomposition products, as monitored by a mass spectrometer with a sensitivity of 3×10^{-9} torr. Larger-scale tests of this promising material have been arranged.

Ion-gage studies on nude gages continued, with measurements being made of ion current distributions in an ordinary Bayard-Alpert type of gage. It was found that the nude gage operated more satisfactorily when a screen was used around the gage elements.

By evaporation of titanium in a baked system at very low pressure onto walls cooled to liquid-nitrogen temperatures, rates of hydrogen sorption were obtained which were as high as 75% of the theoretical maximum.

BIOLOGY AND MEDICINE PROGRAM

ENVIRONMENT RADIATION STUDIES

Terrestrial Ecology: Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — For the period May to October 1961, the Process Waste Water Treatment Plant accounted for about 92% of the $Sr^{89,90}$, 91% of the rare earths (exclusive of Y^{90}), 72% of the Cs^{137}, 76% of the Co^{60}, and 84% of the Ru^{106} discharged to White Oak Creek in Bethel Valley. Of the smaller contributors, the amount of activity detected in the sanitary sewage effluent was the most significant. The mean concentration of Sr^{90} in the effluent for the sample period was 1.7×10^{-6} μc/cc or about 40% of the MPC (continuous occupational exposure for a 40-hr week). Analyses of samples taken at temporary monitoring stations along White Oak Creek and its tributaries indicate a relation between the suspended-solids load in the stream and cesium transport.

Terrestrial Ecology: In Vivo Glass-Rod Dosimetry-Calibration Studies. — Metaphosphate glass dosimeters were exposed to the radiation field on White Oak Lake bed to determine if the photon energies were high enough to use these dosimeters unshielded to measure the external dose to the resident wild mammals. Five high-Z and five low-Z glass rods were attached to the undersides of 0.125-in.-thick Plexiglas platforms placed 3 cm above the soil surface in seven different locations. One-hour dose readings were made with shielded and unshielded pocket ionization chambers (PICS) several times while the rods were being exposed. The shielding material was 0.040-in.-thick tin, which results in the PICS responding linearly and independently of photon energy down to ~25 kev. No detectable difference was noted when the dose readings of 125 shielded PICS and 125 unshielded PICS were compared, but the doses recorded by the high-Z rods were from 15 to 50% higher than the doses recorded by the less-energy-dependent low-Z rods. A small percentage of low-energy photons apparently make up the energy spectrum of the radiation field. The doses recorded by the low-Z rods were 10 to 30% lower than the doses calculated by extrapolating 1-hr PIC readings to estimate a cumulative dose for the one- to two-week periods. Besides the possible error from extrapolating, high PIC readings tend to result from ionization-chamber leakage during exposure, low winter temperatures, high humidity, and dose accumulated while transporting the PICS through radiation areas to and from the platforms. When low- and high-Z rods were irradiated at 10, 20, and 30 r with a calibration source, the response of the low-Z rods was 2.1 times more sensitive. The response of the unshielded low-Z glass dosimeter supports the hypothesis that they can be used in vivo for accurately estimating the external exposure dose to wild mammals on the lake bed. A method for subcutaneously implanting glass rods contained in small nylon cylinders was used on 30 wild rats and proved to be completely successful.

Terrestrial Ecology: Old and New Fallout in Oak Ridge Forests. — Work on the extent and meteorological interpretation of local and stratospheric fallout on forest foliage near Oak Ridge suggested the need for further analysis of fallout behavior. The resumption of weapons testing

in 1961 made it possible to compare fallout distribution before and after tests. Before test resumption, Cs^{137}, Ce^{144}, and Ru^{106} constituted the main residuum from previous fallout. On freshly fallen foliage litter, levels of I^{131}, Zr^{95}-Nb^{95}, Ru^{103}, and Ce^{141} quickly became the major contributors to radioactivity by October 1961. However, freshly fallen litter materials with lower surface-volume ratios (twig and bark fragments) and old forest humus layers protected by fresh litter were still dominated by the longer-lived nuclides. Further collections have been made to show the changing distribution of nuclides in soil layers, in freshly fallen litter, and in rain water. These are being analyzed by gamma spectrometry in preparation for further estimation and interpretation of movement of fallout materials.

Terrestrial Ecology: Arthropod Studies. – The accumulation of Sr^{85}, Cs^{134}, and Co^{60} was measured for millipedes (*Dixidesmus erasus*) in both laboratory and field experiments. These arthropods are dominant during the fall and winter months in the forest floors of mixed mesophytic forests (predominantly *Liriodendron*), where they influence the rate of leaf litter breakdown through their feeding activities. Whole-body burdens of millipedes feeding upon contaminated leaf litter were measured in the laboratory for a 30-day period. Strontium-85 accumulation resulted in increasing body burdens over the 30-day period; the biological half-life (T_b) for Sr^{85} was estimated to be about 20 days. In contrast, Co^{60} concentrations in millipedes equilibrated with concentrations in leaf litter (T_b = 8 days). Cesium-134 body burdens in millipedes reached a peak after 3 days and then declined at a rate which reflected loss of Cs^{134} from the leaf litter. This loss of Cs^{134} was due to leaching by water added to maintain high humidities; the half-time for this loss from leaf litter was estimated to be 6 days under these laboratory conditions. The T_b for Cs^{134} was approximately 2 days.

Field experimentation centered on accumulation and elimination of radioisotopes by millipedes confined in Lucite pens, 0.5 m^2. Recaptures of marked millipedes released into the pens indicated no movement into or out of the pens during the period of study, but movements of millipedes down into the soil still prevented complete recaptures. Accumulation of Cs^{134}, Co^{60}, and Sr^{85} by millipedes in pens produced results qualitatively similar to those found in laboratory experiments; however, data were less conclusive due to incomplete recaptures and longer elimination rates for the radioisotopes. For Co^{60}, the T_b was 17 days in the field vs 8 days in the laboratory; for Cs^{134}, the T_b was 5 days vs 2 days. Further evalution of field results will require investigation of temperature influences on elimination rates.

Terrestrial Ecology: Fish Biogeochemistry. Clinch River Studies. – When the distribution of stable elements is known between water and fish, it should be possible to predict the concentrations of radioactive isotopes of those elements in the fish living in water and receiving continuous releases of radionuclides. Fish-tagging studies showed that the white crappie tended to remain in the vicinity of capture. Consequently, individuals of this species from the Clinch River are being analyzed for Sr, Sr^{90}, Cs, and Cs^{137} to test the correlation between stable chemical

and radiochemical analyses. Initial results show 6.6×10^{-8} g of Sr per g of fish flesh (wet weight), which represents a concentration factor for Sr of 1.12. Bone samples contained 2.27×10^{-4} g of Sr per g of bone (dry weight), or a concentration of Sr 3.85×10^{3} times that in the water. The average Sr^{90} content of bone was 120 μμc/g. This value in bone is equal to a water concentration which is 31% of the MPC of Sr^{90} in water in the neighborhood of atomic installations. These results agree quite well with monitoring data on river water. An analytical method for low concentrations of Cs has just been developed by the Analytical Chemistry Division for use on fish flesh and other biological samples. The preliminary analyses show that there is about 4.5×10^{-8} g of Cs per g of flesh (wet weight).

Terrestrial Ecology: Fish Population Movements. Clinch River Studies. — Fish-tagging studies were conducted on the Clinch River in the vicinity of White Oak Creek to determine the movement of fish in this portion of the Clinch-Tennessee river system. Fish were caught in hoop nets, marked with numbered tags, and released at the point of capture. There have been 226 (4.31%) recoveries from 5244 fish tagged. Most of the recoveries were white bass and white crappie. Particularly striking was the recovery of 142 (17.5%) tags from 812 marked white bass. Apparently, the white bass were originally captured as they made an upstream spawning run in the Clinch during April and May, and then were recaptured by fishermen after returning to Watts Bar Reservoir. The distance between the point of tagging and the point of recovery for white bass ranged from 14.2 to 60.1 river miles, and the average distance was 38 miles. The average time between tagging and recovery was 50 days. White crappie are more restricted in their movements, since the average distance between tagging and recovery was only 10 river miles, and the average time the fish were at large was 89 days. It is evident that fish which have been in the Clinch River in the vicinity of White Oak Creek move considerable distances in the Tennessee River system.

ISOTOPE DEVELOPMENT PROGRAM

Neutron Activation Analysis Research. — The room originally selected for the installation of a 150-kev Cockcroft-Walton type of neutron generator, mentioned in previous reports, was found to be inadequate because of structural deficiencies which would entail a considerable increase in shielding costs. A new, more-remote place has been selected, and engineering surveys on the feasibility of utilizing this space have been completed. In the new location, not only are the shielding costs lowered, but the adjacent area is also available for the chemical processing of samples. The new facility, now scheduled for completion by May 15, will be situated in the building which houses the Reactor School. This change brings about an unexpected dividend in that the generator can be made accessible to the Reactor School staff for occasional use in its training program.

Recovery of Fission Products. — In continued studies of o-phenylphenol (OPP) as a cesium extractant, improved results were obtained in a batch countercurrent test by adding acid to the last extraction stage to

lower the pH to 6–9. That portion of the OPP which had dissolved in the aqueous phase as the sodium salt redistributed into the organic phase, thereby maintaining a concentration as high or higher than the initial 1 M feed. This not only avoided excessive loss of extractant but also resulted in considerably higher cesium extraction coefficients in the system than those previously obtained, since the cesium coefficient has a high power dependence (probably third) on the phenol concentration. In this test with 1 M OPP in diisopropylbenzene (and in a similar test with 4-chloro-2-phenylphenol), cesium recovery was > 98% from tartrate-complexed simulated Purex waste adjusted to pH ~12.7, with six extraction and two scrub (0.3 M NaOH scrub solution) stages at feed/organic/scrub ratios of 5/5/1. The cesium was completely stripped with a single 0.05 volume contact with 0.05 M HNO_3, yielding a product solution containing 2.4 g of cesium and ~0.07 g of sodium per liter (overall Cs/Na separation factor of $\sim 2 \times 10^4$).

In batch extraction tests to determine individual rare-earth extraction behavior with di(2-ethylhexyl)phosphoric acid in the strontium-mixed rare-earth recovery flowsheet, extraction was somewhat slow (> 75% of equilibrium amount in ~1 min) from tartrate-complexed simulated Purex waste, with the order of extraction being Pm > Ce > Eu > Y. By contrast, rare-earth extraction from $NaNO_3$-HNO_3 solution (simulated second-cycle feed) was rapid and the order was the reverse of that above: Y > Eu > Ce, Pm. In individual rare-earth batch stripping tests with various HNO_3 concentrations, equilibrium was reached rapidly in a noncomplexed aqueous system, and the order of stripping was as expected: Pm, Ce > Eu >> Y.

Radioisotope Research and Development. — An air-pulsed mixer-settler was designed and tested. The stage efficiency of an HNO_3—tributyl phosphate system was found to be ~93%.

Six cesium-137 borosilicate polyglass cylinders ~1-1/4 in. high by 2-1/2 in. in diameter were prepared at the Fission Products Development Laboratory by the tetraphenyl boron procedure. The units represent 27,000 curies of Cs^{137}, which emits 125 w of heat to be used in a thermoelectric power unit.

A new procedure was developed for preparing cesium silicate glasses from cesium oxalate. The method consists in precipitating $Cs_2H(C_2O_4)_3$, calcining at 500°C to form $CsHC_2O_4$, blending with SiO_2, and fusing at ~1200°C to form a glass. Other oxides can be added to vary the properties of the glass.

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