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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1962

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OAK RIDGE NATIONAL LABORATORY
STATUS AND PROGRESS REPORT
FEBRUARY 1962

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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U.S. ATOMIC ENERGY COMMISSION

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 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.

SPECIAL NUCLEAR MATERIALS PROGRAM

Chemical and Metallurgical Plant Criticality Studies. — As many as 125 five-liter units of concentrated aqueous uranyl nitrate solution were assembled in critical cubic arrays. The solution, having 415 g of 92.6% U^{235}-enriched uranium per liter and a specific gravity of 1.555, was contained in right circular cylinders of Plexiglas with 1/4-in.-thick walls. The dependence of the number, N, of units required for criticality as a function of the spacing of the units has been determined, as well as the effect of various thicknesses of hydrogenous reflectors on N. In unreflected arrays, N was found to obey the relation

$$N = 2.24\ (\rho_u/\rho)^{1.925},$$

where ρ_u is the solution density in the unit, and ρ is the average solution density in the array. The effect of a reflector is to change the value of the coefficient and the exponent; for example, 6 in. of paraffin causes the coefficient to change to 0.58 and the exponent to 1.692.

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROGRAM

Reactor Physics. — The source required for EGCR startup was estimated for various startup conditions, which included the empty core, the core with 20 fuel channels loaded, and the core with the control rods inserted and with and without fuel elements inserted. The effect of the position of the source in the fueled core on the thermal flux was also investigated. The calculations indicate that a Po-Be source of 10^7 neutrons/sec will provide a thermal flux adequate to ensure proper operation of an in-core fission chamber over the entire range of startup conditions.

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The sensitivity of reactivity lifetime to fission product cross sections was investigated to determine how much the reactivity lifetime of fuels under consideration for gas-cooled reactors will be affected if the aggregate cross section of the fission products is higher than that presently assumed. The sensitivity had been found to be in the range 0.2 to 0.5 for graphite-U^{235}-Th^{232} fuel, with the composition optimized with respect to lifetime, if a graded-exposure equilibrium cycle was used. An approximate analysis indicates that the sensitivity is proportional to the fraction of total neutrons absorbed in fission products and inversely proportional to the reactivity deficiency at the end of the cycle. In addition, the sensitivity is reduced if the fission products saturate (burn out) appreciably during irradiation. Factors of the same type govern the sensitivity to fission product poisoning of batch cycle operation, and the sensitivity of these systems is apparently similar to that of equilibrium systems.

Component Analyses and Tests. — The photoelastic model of the EGCR pressure vessel was sectioned and examined. The data derived from this model are being used to verify and extend the information gained from the strain-gage analysis of the 1/6-scale aluminum model of the same vessel. Excellent agreement between the stresses from the two models was found in the region of the junction between the hemispherical head and the cylindrical portion of the pressure vessel. Discrepancies between the maximum stresses in this region are partially attributable to differences between the Poisson ratios for the two materials.

The photoelastic analysis of the gas-outlet nozzle verified the strain-gage data. Effects associated with the adsorption of moisture in the photoelastic model (time-edge effects) interfered, however, with comparisons between stress distributions for the nozzles in the cluster region, and additional studies must be made before the photoelastic data for these elements can be properly interpreted.

The photoelastic model allowed for a complete examination of the stresses in the shell in the cluster region. Since it was not possible to obtain enough data for ascertaining the stress distributions from the strain-gage analysis because of limited space for gage installation, the additional data help to verify the important conclusion that the highest stresses are in the cluster nozzles instead of the shell, and they add greatly to the information regarding the stresses in the vessel. Reasonable agreement was found to exist between the two sets of data at those points where comparisons could be made.

Flow distribution tests of the EGCR bottom dummy mockup were performed for fixed orifice diameters of 0.143, 0.156, 0.170, 0.180, and 0.190 in. Some of the tests are being rerun to demonstrate reproducibility of the data, and then testing of the prototype and production-model dummies will begin. A series of flow tests on the top dummy was started. A labyrinth geometry is being used to generate head loss.

All welds were completed on the EGCR control-rod-drive test vessel, and the lower portion of the vessel and associated support structures were set. Graphite and heater-assembly subcomponents, such as thermocouple leads, heater leads, the graphite cylinder, and the heater control-panel wiring, are essentially complete.

Pebble-Bed Reactor Experiment. — A practical fuel-handling system for the PBRE was planned that utilizes gravity to move the fuel balls to storage and inspection facilities. Pneumatic tubes are used to lift the fuel balls to the top of the reactor for recycle. A simplified mockup system is being constructed to provide information on pneumatic handling techniques, ball counters, and classifying devices.

Coefficients of friction were measured for spheres moving against spheres and for spheres moving on flat plastic plates as a function of depth of sphere bed and for several sphere diameters and materials. Methods for correlating the data are being studied.

GCR-ORR Loop No. 2. — The sidestream cleanup system and associated instrumentation and the evaporator drain and vent lines were installed in GCR-ORR loop No. 2. Helium leak checking of the main loop piping and the Marmon Conoseal flanges is approximately 50% complete. Flow diagrams for the loop were thoroughly reviewed, revised, and corrected to bring them up to date. Construction has started on the cold finger and on the low-power nuclear test assembly. The dummy fuel element is 80% complete. Two radiation analyzer shields are being constructed. Assembly has continued on the 18-in. pressure-piping plug. Drawings for the latch mechanism and appurtenant quick-disconnect operators were completed. The test-stand modification for the hot test of the 18-in. plug was initiated. A small test stand was designed for determining the helium leakage rate and reliability of the "quad-ring" seal which is to be used in the low-power nuclear test assembly. The design of the cold-finger shield is 80% complete.

Irradiation Program. — A prototype EGCR fuel element with a nominal heat rating of 35,000 Btu hr^{-1} ft^{-1} was removed from GCR-ORR loop No. 1 after irradiation for a seven-week ORR operating cycle. The measured heat generation rate was only 23,000 to 24,000 Btu hr^{-1} ft^{-1}. Evidence was obtained that the element had to be well centered in the gas stream to avoid temperature fluctuations. It was possible for this element to move laterally a maximum of 0.020 in., and this movement resulted in temperature changes of ±90°F, with the temperature increasing with a decrease in the gas gap. A similar element is being irradiated.

Irradiation of experimental assembly ORNL-MTR-48-6 was started. This assembly contains pyrolytic-carbon-coated $(U\text{-}Th)C_2$ particles in a graphite matrix in a sweep-gas capsule and four sealed capsules with similar, loose (no graphite matrix) particles or coated UC_2 particles. The central temperature of the matrix is between 1990 and 2035°F, and the estimated heat generation is approximately 1.1 kw.

Experimental assembly ORNL-41-7, which contains 48 cylindrical BeO specimens and a group of single-crystal samples, has been irradiated to 4300 Mwd in the ETR to a dose of approximately 2.5×10^{20} neutrons/cm^2 at temperatures ranging from 550 to 1050°C. This irradiation will be continued for at least one more cycle in the ETR.

Hot-cell examination of a UO_2-fueled capsule (No. 6) irradiated in GCR-ORR loop No. 1 has shown the cladding of the fuel element to be clean. There is a slight bow (0.010 in.) in the capsule, but there are no visible ridges. The gamma scan shows higher activity at the top of the fuel element than at the bottom. Pellets of five different enrichments were used, and the changes in activity caused by these different

enrichments are also evident. Flux measurements on rings cut from the middle and the bottom of the shroud which covers the fuel element gave 7.8×10^{13} (middle) and 6.5×10^{13} (bottom) neutrons cm^{-2} sec^{-1}. A small amount of Kr^{85} was found when the capsule was punctured, which indicates that the capsule had not failed. The fuel element was then sectioned, and no evidence of chemical reactions was found. The cladding, thermocouple, thermocouple well, and fuel were clean or shiny. The UO_2 pellets were broken but not pulverized.

Materials Investigations. — Carburization and decarburization data obtained for type 304 stainless steel in atmospheres of varying carbon potentials are listed below to illustrate the inversion of the carbon content in the metal as a result of exposure to approximately 0.3 vol % $CO + CO_2$ in helium:

Carbon Potential (P^2_{CO}/P_{CO_2})	Carbon in Metal After 850 hr (%)
0	0.016
0.0071	0.040
0.056	0.062
0.227	1.4
0.363	0.73
0.47	0.42
3.79	0.09

It was observed in the tests in which these data were obtained that low carbon contents in the metal were associated with the presence of Fe_3O_4 as a reaction product and with atmospheres in which the impurities were greater than ~33% CO_2. It appears unusual, therefore, to observe Fe_3O_4 as a reaction product in an atmosphere consisting predominantly of CO, where iron oxides are totally unstable.

The fabrication of BeO specimens for irradiation experiments ETR-ORNL-41-8 and -9 was completed. Pertinent properties of the 236 cold-pressed and sintered specimens are tabulated below:

Specimen Type	Average Bulk Density (% of theoretical)	Average Grain Diameter (μ)
Low-density, fine grain	89.7	15
Low-density, large grain	90.1	45
High-density, fine grain	96.4	25
High-density, large grain	97.7	60

A finned beryllium-clad UO_2 fuel element was thermally cycled 12 times between various temperature limits to determine temperature gradients and temperature response to changes in power input. The radial thermal gradients measured between the central cavity in the UO_2 fuel pellets and the grooves in the finned beryllium tubing are shown in the following tabulation:

Cladding Temperature (°C)	Central UO_2 Temperature (°C)	Minimum Gradient (°C)
300	400–450	100
400	535–575	135
500	675–710	175
600	800–825	200
700	925–950	225

The element was cycled with no gas pressure differential across the cladding wall. Metallographic examination of the tubing after cycling revealed fractures in the beryllium along the 10-in. tube. These cracks were intergranular and transgranular failures perpendicular, parallel, and at a 60° angle to the longitudinal axis. The as-assembled diametral gap was approximately 0.001 in. The tangential strain introduced into the beryllium at 700°C because of the expansion of the UO_2 was calculated to be 0.4%. If the cladding were cooled rapidly to room temperature, this strain would amount to approximately 0.55%. The room-temperature fracture strain of beryllium tubing, regardless of the extrusion ratio, is greater than the calculated strain; however, concentration of stresses because of the fins would increase the probability of failure. It is assumed that several cracks existed in the beryllium prior to thermal-cycling treatment, as reported in documents from Nuclear Metals; however, the element withstood pressure testing prior to cycling, and it must be inferred that stresses caused by the thermal cycling propagated the cracks completely through the tube wall.

A corrosion test was run on a Be–0.5 wt % Ba alloy in helium containing CO_2. The test temperature was 750°C, the CO_2 pressure was approximately 50 mm Hg, and the duration of the test was 288 hr. A log-log plot of the weight-gain data indicated protective-film formation for 10 to 20 hr, followed by breakaway oxidation as a result of spalling of the reaction product. The weight gains were, in general, lower than in a prior test at a CO_2 pressure of approximately 5 mm Hg, indicating that the oxidation rate may be pressure-dependent. X-ray analyses showed the oxide films in both cases to be BeO. A third test is being run at a CO_2 pressure of approximately 500 mm Hg.

THORIUM UTILIZATION PROGRAM

Thoria Pellet Development. – Accelerated attrition testing of milled ThO_2 pellets of varying densities has shown that the structural weakness introduced into the pellets as lamination cracks results in increased attrition losses under conditions existing during spouted-bed tests. Preliminary measurements appear to substantiate qualitative observations that for the high-density laminated pellets, attack seemed to be more pronounced in the areas of defects. The sharp impacts on the pellets in

spouted-bed tests apparently accentuate structural effects which are not revealed in the milder, wet ball-milling tests.

An apparent residual effect has been observed of the size of granules in the ThO_2 powder on the milling behavior of sintered pellets made with this powder. To test this qualitative observation, pellets were prepared from −100-mesh granules instead of the −35-mesh granules ordinarily used; the pellets were isostatically pressed and then sintered at 1750°C for 41 min. After wet milling, no evidence of gouging out of granule particles could be seen, and the pellets appeared significantly smoother and freer of surface defects than were the pellets prepared with coarser (−35-mesh) granules.

Fuel Cycle Development: Chemistry of Solid Fuel Element Materials. — Measurements of the unit-cell lattice parameter, a_0, were made on ThO_2-UO_2 specimens which had been equilibrated in air for periods up to 100 days. These values were plotted against the analytically determined uranium oxidation numbers, UON, of the specimens. Discontinuities in the relationship were observed between UON values of 4.8 and 5.0, at which minimal values of a_0 were observed for specimens containing between 10 and 70 mole % thoria. The minimum a_0 value for the specimen containing 70 mole % thoria occurred at a UON of 5.0 and was 0.015 A lower than that at a UON of 4.0; for the specimen containing only 10 mole % thoria the minimum a_0 value occurred at a UON of 4.82 and was 0.055 A lower than that at a UON of 4.0. These discontinuities are tentatively interpreted as indicating phase transitions from solid-solution regions of lower oxygen content to a polyphase region initially containing U_4O_9.

Fuel Cycle Development: Fuel Preparation. — A 1.4-kg batch of 3 wt % uranium–thorium oxide in which uranium was added as ammonium diuranate was prepared by the sol-gel process for compaction experiments by Babcock and Wilcox. The preparation was easily sized in the dried gel stage to fractions pre-estimated to give the size distribution desired when fired. Particle densities averaged 9.88 g/cc; the O/U ratio was 2.032 to 2.040; the gas release at 1200°C in vacuum was 0.003 cc/g. A test compaction in a 5/16-in.-diam × 11-in. tube gave a packed density of 8.6 g/cc.

Further laboratory-scale experiments demonstrated the ability of the sol-gel process to produce particles with densities > 9.8 g/cc which can be compacted to bulk densities > 8.7 g/cc when uranium is added as uranyl nitrate solution. Adjustment of the pH of mixed uranium–thorium oxide sols to approximately 3, either by ammonia addition or by controlling total nitrate, and thorough mixing are required to prevent inhomogeneity with respect to the U/Th ratio.

The optical density of thoria sols at a wavelength of 6700 A varies linearly with thoria concentration, and for a particular thoria preparation shows a reproducible minimum with varying NO_3^-/ThO_2 mole ratio. This minimum is related to the surface area and to the maximum dispersion of the ThO_2 preparation.

In pilot-plant steam denitration tests (15 kg) in the rotary calciner, ThO_2 products were produced which gave excellent thoria sols and uranium–thorium oxide products.

Fuel Cycle Development: Fuel Fabrication Studies. — Particles of ThO_2–5 wt % UO_2 which had been sized in the dried gel stage and subsequently fired to 1150°C were vibratorily compacted to approximately 86% of theoretical density. This is comparable to values obtained under similar conditions, except with sizing as the final operation.

Nine tubes of type 347 stainless steel, 45-1/2-in. length × 1/2-in. OD × 0.035-in. wall, were loaded with ThO_2–3 wt % UO_2. Bulk densities between 8.80 and 8.88 g/cc were obtained in all cases with a Branford pneumatic vibrator.

Nine irradiation capsules were fabricated for insertion in the NRX at Chalk River during May 1962. Seven capsules are 39 in. long, two capsules are 22 in. long, and all have a 5/16-in. OD × 0.025-in. wall. The capsules were loaded with ThO_2–5 wt % UO_2 (fully enriched) to bulk densities ranging from 8.78 to 8.86 g/cc.

Studies to develop design criteria for the crushing and vibratory compaction phases of the BNL Kilorod program were completed. These criteria have been incorporated into a process design which permits a flexible operation with respect to material flow and equipment adjustment.

Thorium–U^{233} Fuel Rod Fabrication Facility. — Detailed design of phase I of the Thorium–U^{233} Fuel Rod Fabrication Facility was completed, and drawings were transmitted to the contractor for cell 4, Building 3019. Phase-I construction will include the removable concrete-pad floor, all of the first-floor processing cubicles, and the vessel off-gas filter system.

With the exception of one item, the density-scanning device, all the equipment for the fuel rod fabrication processes has been conceptually designed. Final design of 60% of this equipment is completed.

MOLTEN-SALT REACTOR PROGRAM

MSRE Design. — Drawings of the pump mount, heat-exchanger mount, and drain-tank-cell layout were reviewed, and necessary revisions are being made. Drain-tank-mounting drawings were issued for approval. Heater and thermocouple-cell disconnect location drawings are still in progress.

The electrical distribution diagram has been instrumented, and functional wiring diagrams for all coolant heating are 60% complete. The primary piping-support mockup was finished, and design, based on the mockup, of actual pipe support and heater structure was begun.

The control-rod drive-mechanism mockup was completed. Testing of this mechanism is now in progress.

The design package for site modification outside the building was completed and reviewed and the necessary changes were noted on the prints issued for comment. The entire package is being revised accordingly.

An alternate core design, incorporating clad graphite, was prepared for study and comment.

MSRE Procurement and Construction. — Dished heads for pump bowls, fabricated by Lukens Steel Company, were received and are being heat-treated preparatory to machining. The 17-in.-diam heads for the heat exchanger, fabricated by UCNC shops at Paducah, were received. The Paducah shops are machining dies to form the reactor-vessel heads.

Fabrication of the grid support structure for the graphite moderator blocks was begun in the Y-12 shops. Considerable progress was made in the fabrication of the radiator enclosure.

The contractor for the major modifications in Building 7503 has set the structural steel beams in the drain-tank cell and is ready to install the reinforcing steel preparatory to pouring the concrete for the cell walls. The reinforcing steel was set for the penthouse walls over the coolant-system cell.

Fabrication of the stainless steel grid and liner for the drain-tank cell is progressing at the Clermont Engineering Company shops in Philadelphia. Fabrication of the six shielding-support beams and the modifications to the 24-ft-diam reactor cell is in progress at the Chicago Bridge and Iron shops in Birmingham.

A preliminary critical-path schedule was completed for the MSRE installation. Critical-path schedules are being prepared for each major component and for the procurement and fabrication of instrumentation for the MSRE.

Development of Components and Systems. — In the engineering test loop, an INOR-8 graphite container was installed and Inconel was replaced with INOR-8 pipe. A dry box for installing samples in the graphite container was completed, and handling procedures are being established by use of dummy elements.

The removal seal for the sampler-enricher transport container, designed to exclude oxygen from the dry box during insertion and removal of the sample capsule, was cycled 1500 times at a stroke of 42 in. with light lubrication. The total leak rate to the atmosphere through the seal with a helium pressure of 15 psig was < 1 cc/min.

The core heater has operated a total of 3750 hr. The surface temperature of the heater pins was held for 1425 hr at 1425°F without difficulty, but the heater tube support buckled, causing some misalignment.

The 5-in.-pipe box-heater test was extended to include a pipe-to-support attachment. Preliminary tests indicated that very small heat losses were due to this attachment. The test now contains three full-scale heaters for heating 6 ft of 5-in. pipe.

A model of the proposed MSRE control rod and drive unit was assembled. Modification to the initial design of the release clutch was found necessary after testing.

The fabrication of the first pair of 5-in. MSRE freeze flanges, which use a ring-joint gas seal, was completed, and the flanges are being prepared for testing.

The bench braze joint made by use of the 0.005-in.-thick sheet braze metal preform was inspected ultrasonically and metallographically and was found to be satisfactory.

A stationary and a movable pipe vise, capable of exerting an axial load of 2000 lb, a pipe cutting tool, and a pipe tapering attachment have been obtained for performing the mechanical operations for making

braze joints. Preliminary tests on stainless steel and INOR-8 pipe have been satisfactory.

One INOR-8 test braze joint, which was subjected to short-term tension loading at various temperatures to simulate extremes in service, failed at 1700°F with a loading of 8000 psi after having been held for 5 min at 1300, 1500, and 1600°F at respective tensions of 28,000, 10,000, and 8000 psi.

Continued testing of the freeze-flange clamp drives resulted in abandoning the bolt and trunion drive in favor of a hydraulic drive. In-cell jacking equipment for the pump and pipe lines, electric power disconnects, and 1-5/8-in.-diam periscope was tried on a preliminary basis without difficulties.

MSRE Pump Program. — The Inconel centrifugal molten-salt test pump (PKP type) was shut down in order to replace an inoperative drain valve. Since the replacement the pump has circulated MSRE fuel satisfactorily for 788 hr at 1225°F, 1950 rpm, and 510 gpm.

During posttest examination, evidence of slight rubbing was found on the journal and bearing of the test pump with the molten-salt-lubricated journal bearing. This pump had been operated for 12,500 hr, circulating $LiF-BeF_2-UF_4$ salt at 1225°F, and had been started and stopped 92 times. The journal and bearing will be replaced and the testing will be resumed.

The fifth INOR-8 casting of a coolant pump impeller was of class I quality and is being machined for use in the MSRE. The fifth INOR-8 coolant pump volute was cast by the founder, and preliminary radiographic and dye-penetrant inspection at his plant indicated the casting to be satisfactory. Defects were removed from another pair of coolant pump impeller and volute castings, and weld repairs are being made prior to solution heat treatment. These parts will be used for a spare coolant pump.

Fabrication of the rotary element for the prototype fuel pump was completed, and the pump is being assembled. The hot-test facility for the prototype fuel pump was modified to incorporate the movable pump mount, MSRE freeze flange, a test thermal well, and an MSRE electric pipe heater. Completion of the facility awaits delivery of the pump tank, which is 80% complete.

Water testing of the MSRE coolant pump design was completed.

Discoloration of the oil was noted during the endurance testing of one of the canned-motor pumps for the lubrication stands, and inspection revealed a varnishlike coating over most of the surface of the rotor. The supply of cooling oil to the rotor was increased several fold by adding three axial grooves to each of the journal bearings, and testing was resumed.

Metallurgy. — Procedural specifications for welding INOR-8 to stainless steel (PS-35), INOR-8 to nickel (PS-34), and INOR-8 to Inconel (PS-33) were prepared and qualified. Mechanical-properties specimens were prepared from the qualification-test welds for room- and elevated-temperature testing. All dissimilar joints were made of the nickel-base filler material, INCO-82. Short-time tensile tests were performed at room temperature, 538°C, and 648°C, on transverse samples of these weldments. All joints of these dissimilar-metal welds were sound, and all

failures occurred outside the weld-metal area in the nickel, Inconel, or stainless steel base metal.

Elevated-temperature creep tests on transverse samples from the INOR-8-to-type 347 stainless steel welds were made at 538 and 648°C. Preliminary examination of the tested specimens indicates that all failures occurred at the stainless steel-INCO-82 interface. Very little strain, as indicated by reduction in area, was seen in these specimens. Some specimens ruptured on loading, the rupture occurring in the type 347 stainless steel base material.

In all cases the weldments exhibited strengths equal to the strength of the weaker base metal, indicating that this joint has suitable mechanical properties for MSRE use.

Instrumentation Development. – Two prototypes of a float-type molten-salt level transmitter were fabricated and are being tested in a facility constructed for that purpose.

The two prototypes use the same principle of operation but differ in that in one the differential-transformer core assembly is suspended below the float, while in the other it is above the float. The latter arrangement eliminates a nondrainable tube and simplifies the problem of differential transformer replacement, but has inferior damping characteristics and may develop excessive hysteresis if salt vapors condense on the core.

Preliminary results indicate that the temperature effects on the span and the zero point, over the range of 800 to 1200°F, are acceptably low and that there is no pressure effect and negligible hysteresis. There has been no evidence of absorption of salt by the graphite float. Further tests are being made to determine the effects of continuous high-temperature operation on the characteristics of the transmitters.

Tests were performed to determine the accuracy of Inconel-sheathed, mineral-insulated thermocouples attached to INOR-8 pipes with Heliarc-welded, side-lug attachments. Seven thermocouples of this type were tested under static conditions between 900 and 1400°F. The maximum error noted was +6°F, which was within the 3/4% limit of error specified for the material used in this test.

Methods of mechanically attaching mineral-insulated sheathed thermocouples to MSRE radiator tubes are being investigated, and tests are being made to determine the error inherent in this method of attachment. A test device was constructed which simulates conditions in the MSRE radiator and which will permit testing of various methods of attachment. A 1/8-in.-diam sheathed thermocouple strapped to a 7/8-in.-OD × 0.065-in.-wall INOR-8 tube was tested under static conditions and with air flow over the tube. With no air flow and with an outer wall temperature of 1150°F, as determined by a 30-gage reference couple spot-welded to the outer wall of the tube, the 1/8-in.-diam sheathed couple read 1065°F. At an air flow of 80 to 90 fps and an outer wall temperature of 1040°F, the 1/8-in.-diam test couple read 850°F.

Reactor Analysis. – Multigroup diffusion-theory calculations are in progress to obtain estimates of reactivity introductions in the MSRE resulting from insertions of various amounts of "cold" fuel into the core, and from filling the core with fuel salt containing a higher-than-normal concentration of U^{235}. Some preliminary results were analyzed, and an

estimate of $-2.7 \times 10^{-5}/°F$ was obtained for the fuel-salt temperature coefficient of reactivity, in excellent agreement with the estimate of $-2.8 \times 10^{-5}/°F$ previously obtained from a simplified two-group model.

Preliminary calculations of the "cold slug" type of accidents were made, assuming that the graphite temperature remains constant as cooler fuel salt displaces salt at normal operating temperature from the core. Reactivity insertions resulting from slugs at 900, 1000, and 1100°F were calculated by use of the multigroup diffusion theory; the results are in good agreement with previous estimates based on elementary perturbation theory.

Estimates were made of the reactivity resulting from partial replacement of the center graphite stringer by fuel salt, assuming that a portion of the stringer is removed from the center region of the reactor. Reactivities of 0.045, 0.083, and 0.11% result from the replacement of center sections 1, 2, and 3 ft long respectively.

Checkout of a revised version of the reactor kinetics program PET-I is in progress. Additions were made to allow for treatment of separate moderator and fuel temperatures, and the input and output sections of the program were modified and considerably enlarged.

REACTOR SAFETY

Release of Fission Products on Out-of-Pile Melting of Reactor Fuels. — In ORNL-3242, p 9, data on the effect of burnup on the release of fission products by oxidation of UO_2 having a 1000 Mwd/ton burnup level were reported and were compared with previous data for UO_2 having only a tracer-level irradiation.

The data presented here were obtained with PWR UO_2 irradiated to a burnup level of 4000 Mwd/ton. The samples were continuously weighed on a thermobalance while they were heated in helium for about 15 min and then in air. The fission-gas release was measured separately during each period and was almost constant (0.2 to 0.6%) during exposure in helium. Exposure in air was continued just long enough to carry the oxidation reaction to completion, as indicated by the weight increase.

The results in the tabulation show about the same release values that were reported for the 1000 Mwd/ton material, with a similar drop in iodine release at 800°C. On the other hand, an accelerated iodine release (from 7.1% at 800° to 48.9% at 900°) occurred at 900° instead of at 1000°C, as in the material at the lower irradiation level. No significant enhancement was apparent.

Time* (min)	Temp (°C)	Percent Released						
		Xe-Kr	I	Te	Cs	Ru	Sr	Ba
23	500	1.5	3.6	< 0.007	< 0.0004	< 0.005	< 0.0004	
18	600	4.4	10.3	< 0.006	0.002	0.075	< 0.001	
15	700	6.7	10.4	0.004	0.001	0.44	< 0.0003	< 0.0006
15	800	13.2	7.1	0.007	0.015	1.0	< 0.0004	< 0.0007
19	900	20.0	48.9	0.4	0.009	17.3	< 0.0001	0.01
15	1000	38.9	84.0	11.5	0.09	72.1	< 0.0003	< 0.001
14	1100	65.7	79.4	15.9	0.17	90.8	< 0.05	< 0.003
16.5	1200	70.6	81.7	37.1	0.76	89.9	< 0.01	< 0.001

*Minimum period required for complete oxidation.

Release of Fission Products on In-Pile Melting of Reactor Fuels. — The first attempt at in-pile melting of a UO_2 specimen, with the power level at 60 w/g for a 20-min exposure, resulted in partial melting of the specimen. Radiochemical analysis of the materials from this experiment is now essentially complete. The fission product nuclides Ce^{144}, Zr^{95}, Y^{91}, and Sr^{90} were for the most part retained in the UO_2; Xe^{133} and Ru^{106} were largely released from the UO_2; Ba^{140}, Sr^{89}, and Cs^{137}, all of which have volatile precursors, were partly released. These results must be considered preliminary because they are based on only one experiment.

The second and third attempts used different flux levels and different uranium enrichments to obtain power levels of 110 w/g. In both these attempts (10 and 5 min of irradiation respectively) complete melting of the UO_2 was obtained, with some penetration of the material into the ThO_2 support structure. Hot-cell examination revealed needlelike crystals, presumably from vaporized UO_2, after 10 min of exposure but not after 5 min. Such variation in behavior of the UO_2 may well affect the release and subsequent deposition of fission products. Radiochemical analyses of the materials from these experiments are in progress, as are designs for experiments to determine the cause for the variation.

The design of the F-3 facility, in which fuel specimens for future meltdown experiments will be preirradiated to obtain high concentrations of fission products, has been approved by the Operations Division, and the facility is ready for installation in the ORR.

Deposition and Removal of Fission Products from Gases. — The behavior of fission products in gas streams containing very fine aerosol particles is under continuing study. A commercial instrument, consisting

essentially of a fog chamber and a photocell circuit, has been used to evaluate methods for removal of alumina and platinum particles from air streams. These particles, according to literature data and diffusion measurements, are about 0.01 μ in diameter. Highly preliminary results suggest that an "absolute" filter reduces their concentration by a factor of 10^2 to 10^4 in a 5-fpm air stream; a 0.75-in. depth of granular charcoal (6 × 16 mesh) is considerably less effective. Additional measurements of diffusional deposition of radioiodine vapor and I^{131}-labeled particles in cylindrical tubes gave results in agreement with previous observations and further demonstrate the value of such measurements in the characterization of aerosols.

Reactivity Effects During Nuclear Excursions. — An investigation is in progress to determine the effect of small changes of fuel distribution on the reactivity of pool-type research reactors, particularly changes that might occur during reactor transients and thus contribute to the severity or to the reactivity compensation of such excursions. As part of this investigation, calculations are being performed with the GNU and MODRIC one-dimensional multigroup diffusion theory codes and the two-dimensional PDQ-2 few-group diffusion theory code, all programmed for the IBM 7090 computer. Thus far, ten calculations have been completed from which definite predictions have been made concerning the effects to be expected in several mockup experiments in the Bulk Shielding Reactor I. For the experiments, which are yet to be performed, eight special fuel plates, each containing approximately one-half the amount of uranium as a regular fuel plate, were fabricated so that they can be inserted between the fuel plates of a BSR-I element and thus augment the fuel density in selected regions of the reactor. If the experimental and calculational results agree for corresponding configurations, further calculations involving more realistic fuel redistributions will be undertaken.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Thermal-Transient Studies. — The techniques of frequency domain analyses originally developed for communications and control engineering problems have been applied to the investigation of thermal systems which experience random perturbations. An analog-filtering device is being used to compute the power spectral density (PSD) of temperature fluctuations at the inside wall of a heated pipe due to turbulent exchange between the fluid and the pipe wall. Here, the PSD is a statistical quantity defined as the mean-square temperature fluctuation per unit of frequency bandwidth.

A relationship has been developed which may be used to relate the PSD of the wall-temperature fluctuations to thermal stress cycling of the pipe wall. By the use of Laplace transform techniques, the frequency response of the pipe wall to temperature fluctuations can be obtained, from which the periodic temperature gradients and hence the thermal stress variations in the pipe wall may be estimated.

Preliminary PSD measurements obtained with water flowing in a 2-in.-diam (sched 40) stainless steel pipe at a Reynolds modulus of 73,000

indicate that most of the transient thermal power generated at the inner surface of the pipe is in the range of frequencies below 5 cps, with some of the power at frequencies as low as a few tenths of a cps.

An analytical model that postulates an unstable boundary layer combined with eddy penetration is being investigated by comparison of the amplitude of thermal oscillations predicted by the model (analog-computer solution) with experimental measurements. Initial comparisons suggest that the model is reasonable. It is felt that the results of this investigation will contribute a better understanding of turbulence in general and of boundary-layer phenomena in particular.

NUCLEAR TECHNOLOGY AND GENERAL SUPPORT

Basic Reactor Shielding. — A general-purpose Monte Carlo code for the solution of gamma-ray transport problems (OGRE) is near completion. This code, with special subroutines, is expected to replace many of the present codes. For example, subroutines being written for the calculation of the transmission and reflection of gamma rays impinging on a multilayer infinite slab, as well as the heating deposited by the gamma rays, will replace an Oracle code (ORNL CF-60-10-37).

Comparisons between various Monte Carlo tallying procedures have been initiated, the first being for gamma rays and neutrons in an infinite medium. These procedures include statistical estimation on the first flight, statistical estimation on the last flight, and a conditional Monte Carlo procedure. One particularly interesting study was for an isotropic source of neutrons in hydrogen. The tallying procedure of statistical estimation on the first flight appears to be the best from the standpoint of low variance, as well as for accuracy for distances out to 90 cm.

Two codes of the United Nuclear Corporation, RENUPAK and NIOBE, have been checked out for use at the ORGDP computing facility. These codes are for the study of neutron transport in an infinite medium and in a spherical multilayered shield respectively. Standard data sheets are being prepared so that the codes will be generally available.

A very important step has been taken toward the use of the output from the general-purpose 05R Monte Carlo code (ORNL-2609, p 87) for the solution of neutron transport problems. A general-purpose routine for reading 05R tapes (ORNL CF-62-1-2) has been written which will manage all the tape-handling procedures and leave only the Monte Carlo tallying procedure to be performed by the user.

A standard output is being designed so that Monte Carlo codes, transport codes, and multigroup diffusion codes can be coupled together. This is possible because the output from each code is the flux or some similar quantity. The output medium will be binary tapes and/or BCD cards with FORTRAN formats. This will eliminate a considerable amount of work now required for transcribing and combining the results of problems.

Codes of general interest in shielding computation have been or are being written for data manipulation (ORNL-3255), computation of certain functions (ORNL CF-62-2-20), and integration (ORNL-3263).

Power Reactor Fuel Processing: Mechanical. – In further shear-leach tests, a mockup fuel assembly of 36 porcelain-filled stainless steel tubes was easily sheared into pieces 1/4, 1/2, and 1 in. long with the 250-ton prototype shear equipped with a stepped blade. Clean pieces of tubing were produced in the 1/2- and 1-in. cuts, but the 1/4-in. pieces were severely flattened, entrapping core material.

Power Reactor Fuel Processing: Solvent Extraction. – Laboratory studies with synthetic U^{233} solutions indicated that a countercurrent-extraction and static-scrub system would satisfactorily decontaminate uranium from Th^{228} for use in fuel recycle studies. Uranium extracted from 1 M $Th(NO_3)_4$–0.8 M $Al(NO_3)_3$ with 2.5% tributyl phosphate in Amsco or with 2.5% di-sec-butyl phenylphosphate in diethylbenzene and scrubbed with 4 M NH_4NO_3 was decontaminated from Th^{228} by factors of 700 and 5×10^4 respectively. The decontamination factor with di-sec-butyl phenylphosphate without scrub was 600. When the feed contained only 0.25 g of thorium per liter, decontamination factors could not be determined, due to the uncertainty of thorium determinations in very dilute solutions.

In a series of irradiated tests in which ThO_2 was washed with 0.5 to 4 M HNO_3, only a small amount of activity was removed from the ThO_2; 0.2% was removed at room temperature and 0.7% at 95°C.

Power Reactor Fuel Processing: Zirflex Process. – Sixteen Zircaloy-2–clad UO_2 pellets, irradiated in the NRX to about 20,000 Mwd/ton and decayed about nine months, were declad with boiling 6 M NH_4F–1 M NH_4NO_3. The pellets were fractured but did not fall apart. Uranium losses were 0.03 to 0.08%, and plutonium losses were 0.01 to 0.04%. The core pellets were 99.5% dissolved in 4 M HNO_3–0.1 M $Al(NO_3)_3$ in 5 hr, which was slightly faster than the rate for unirradiated pellets.

Power Reactor Fuel Processing: Darex Process. – In the Darex dissolution of APPR fuel, which contains ~2% silicon, voluminous quantities of insoluble silica are formed. The silica becomes highly hydrated during chloride removal and plugs the chloride stripping column. The silica in a solution containing 60 g of stainless steel per liter, made by dissolving APPR fuel in 5 M HNO_3–2 M HCl containing 0.005 M HBF_4, was made filterable by digesting it for 2 hr at 90°C. At a filtration rate of 0.5 ml cm^{-2} min^{-1}, silica was 99% removed. Digestion without filtration led to the passage of 87% through the chloride stripper column, the remainder being easily removed from the column by washing it with dilute nitric acid. In both cases, ~17% of the fluoride was volatilized during chloride stripping. Boiling the dissolver solution for 24 hr at total reflux also converted the silica to a form that was 99% removed by filtration, at an average rate of 1.2 ml cm^{-2} min^{-1}. Insoluble stainless steel carbides and oxides were also removed quantitatively. Addition of gelatin or molybdate to the dissolver solution formed sticky, gelatinous precipitates.

Power Reactor Fuel Processing: Corrosion. – Unwelded Corronel 230 exposed to boiling 20 M HNO_3 for 720 hr was corroded at maximum rates of 5.5, 1.5, and 1.2 mils/month in vapor, interface, and solution respectively. All specimens underwent intergranular attack. In boiling

(116°C) 10 M HNO_3–0.5 M HF for 72 hr, uniform rates of 42.6, 43.7, and 62.0 mils/month for vapor, interface, and solution specimens were noted; at 60°C corresponding rates were 0.92, 1.73, and 2.37. In boiling 10 M HNO_3–0.5 M NaF–0.75 M $Al(NO_3)_3$, 408-hr maximum rates were 0.78, 0.71, and 0.71. Corresponding rates for welded titanium-45A were 0.02, 0.16, and 0.28. Neither alloy showed localized attack. In boiling 1 M HF, 48-hr exposure of Corronel 230 showed rates of 4.09, 23.2, and 43.7 mils/month in vapor, interface, and solution respectively. In 120 hr of exposure to boiling, flowing 1 M HF–0.06 M H_2O_2, corresponding rates were 12.7, 10.6, and 2.13. Corronel 230 dissolved at a nominal rate of 2700 mils/month in a 4-hr exposure to boiling (104°C) 1 M HNO_3–3 M HF, whereas type 309 SCb stainless steel was corroded at a maximum rate of 590 mils/month. At 60°C, a 72-hr exposure of Corronel 230 resulted in rates of 10.6, 0.67, and 1.25.

Corrosion rates of INOR-8 in chlorine gas at 508 and 720°C were 0.62 and 103 mils/month, respectively, in 24 hr. These rates are lower by a factor of about 10 than those previously found for Haynes alloy No. 25 and Nichrome V.

Titanium-45A exposed 120 hr to a boiling solution prepared by dissolving 60 g of stainless steel per liter of solvent in 2 M HCl–5 M HNO_3–0.1 M HBF_4 was covered with a hard, adherent deposit. After descaling, corrosion rates were 0.26, 6.02, and 12.5 mils/month for vapor, interface, and solution specimens.

Welded specimens of commercial Ni-o-nel and low-carbon nickel alloy exposed in vapor and condensate above boiling 4 M HNO_3 for 192 hr were corroded at rates of 0.15 and 0.01 mil/month, respectively, for Ni-o-nel and at 0.06 and 0.01 mil/month for LCNA. Both alloys in the vapor phase acquired a brown stain.

Power Reactor Fuel Processing: Graphite-Base and Uranium Carbide Fuels. – Digestion of type GBF graphite with boiling 90% HNO_3 for 50 to 100 hr oxidized < 2% of the graphite. A small amount of dark red-brown, soluble organic oxidation products was isolated from the nitric acid solution by evaporation. These products were acidic, with a neutralization equivalent of about 88 g, indicating that their molecular weights were much higher than those of the products isolated after extended oxidation.

Small castings of uranium monocarbide were analyzed for uranium, tungsten, total carbon, free carbon, and oxygen, subjected to the standard hydrolysis tests, and examined metallographically. The high purity of the castings, estimated as 89% UC, 5% U_2C_3, 4.5% U metal, 1.3% UC_2, and 0.2 to 0.4% WC, renders them suitable specimens for studies of the effect of radiation on the chemistry of UC.

Dissolver Solution Analyses. – Studies were completed on the use of sulfamic acid for destroying nitrite prior to the coulometric titration of uranium(VI). The results have been reported in ORNL TM-113.

A high-sensitivity coulometric titrator was built and is being tested. It is designed for the determination of milligram and submilligram quantities of high-equivalent-weight elements, such as uranium, plutonium, and neptunium.

Work was continued on a procedure for coulometrically titrating uranium, following its extraction with tri-n-octylphosphine oxide in cyclohexane. Uranium can be quantitatively removed from the organic extract by three re-extractions with 3.5 M $(NH_4)_2SO_4$ and titrated without further chemical treatment. For samples containing 5 to 10 mg of uranium, the relative standard deviation of the entire procedure is 0.2% or less. This procedure was applied successfully to the analysis of dissolver solutions of the Foreign Research Reactor type, that is, nitric acid solutions containing Al, Hg, and U; it is now being tested with dissolver solutions of other reactor fuels.

A precise procedure was developed for determining the uranium content of ThO_2-UO_2 fuels and is being used regularly in service analyses. The sample is ground to pass a No. 100 sieve, after which a weighed 2- to 3-g test portion is refluxed with fuming HNO_3 containing a small amount of HF until dissolution is complete. The solution is then diluted to 25 ml, and aliquots are coulometrically titrated for uranium(VI) in a supporting electrolyte containing sulfamic acid, which destroys nitrites. Duplicate determinations by this method were found to differ, on the average, by 0.4%.

Radioactive Waste Treatment and Disposal: High-Activity Waste Calcination. — The product of semicontinuous evaporation and calcination of simulated TBP-25 waste to form a lead phosphate glass (additives: 2 moles of NaH_2PO_2 and 0.25 mole of PbO per liter) in a 24-in.-long, 4-in.-diam stainless steel pot had a density of 2.84 g/ml and a volume about 1/8 that of the concentrated (1.8 M Al) waste solution. The thermal conductivity of the glass varied from 1.05 Btu hr^{-1} ft^{-1} $(°F)^{-1}$ at 300°F to 1.60 Btu hr^{-1} ft^{-1} $(°F)^{-1}$ at 1050°F, with a temperature coefficient that was positive and nonlinear with temperature, indicating an amorphous solid. Ruthenium volatility decreased from 13.7 to 3% during processing, indicating removal of ruthenium contamination of the equipment from earlier experiments.

Stainless steel, type 304, should be a satisfactory material for waste calcination pots, with negligible corrosion in storage. Corrosion was maximum when the last of the acid and water were being driven off at 850 to 1000°C. Overall corrosion in 24 hr of evaporation-calcination of Purex and TBP-25 wastes was 0.26 to 0.34 mil at 900°C (8 to 10 mils/month). Of this about 0.4 mil was attributable to air oxidation. When glass-making additives were present, rates were about 42 mils/month for the TBP-25 lead phosphate glass at 920°C and 145 mils/month for a Purex borophosphate glass at 900°C. Release of SO_3 from the Purex caused aggressive intergranular attack (possibly catastrophic) at temperatures above 900°C, the critical temperature, depending on the melt composition.

In 11 tests with simulated Darex, TBP-25, and TBP-25 wastes containing organic residues, average overall evaporation-calcination rates for the full-scale (96-in.-long, 8-in.-diam) pot calciner decreased with increased filling (evaporation) time. This is attributed to the higher bulk densities and consequently higher thermal conductivities in the final product. Both batch and continuous processes were demonstrated, with satisfactory control of the continuous evaporator. Filling rates averaged 8.6 to 19.4 liters/hr, depending on feed type, concentration, and filling period, with final solid-product bulk densities ranging from 0.61 to 1.42 g/cm^3.

Material- and energy-balance calculations for a hypothetical Purex waste indicated that an 8-in.-diam pot must be used for wastes cooled less than 180 days, but that a 12-in.-diam pot may be used for calcining older wastes. The maximum vapor rates to the fractionator for Purex wastes are ~20% greater than those predicted for TBP-25, but the column will operate below flooding at the higher rate.

Radioactive Waste Treatment and Disposal: Low-Activity Wastes. — Laboratory-scale support studies for the low-level (essentially tap water) waste-treatment pilot plant showed that (1) BO-4 grade vermiculite is subject to plugging and complete stoppage if allowed to contact small amounts of solids; (2) cobalt and ruthenium removal from the waste stream is improved by the addition of an anion exchange column to the system; (3) Nalco reagents are satisfactory, within 1 ppm accuracy, for pilot-plant control analysis; (4) residual hardness of the liquid leaving the clarifier should be decreased by densification of the sludge blanket by recirculation.

Four demonstration runs on ORNL waste were completed in the low-level waste-treatment pilot plant. In all runs, Sr^{90} and Cs^{137} were removed from the solutions by decontamination factors of > 2000 and > 246 respectively. The plant effluent contained < 1.5% of the current MPC_W values for these isotopes for a 168-hr week. Run times were from 71 to 100 hr of continuous operation, with the use of 1800 to 2100 resin-bed volumes.

Radioactive Waste Treatment and Disposal: Engineering, Economic, and Hazards Evaluation. — Space requirements to dissipate the fission product heat in the waste from a 56,000-Mw (thermal) nuclear power economy were calculated for cylinders of calcined waste buried vertically in the floors of rooms in salt media. Requirements calculated for Purex wastes in acidic and reacidified forms in 6-, 12-, and 24-in.-diam vessels range from 20 acres/year after 0.65 year to about 2 acres/year at 30 years age at burial. Corresponding results for Thorex wastes are 10 and 0.7 acres/year. Calculations were made also for an acidic Thorex glass in 6-in.-diam vessels. The primary effect of the form of the waste (acidified, reacidified, glass) and of the vessel diameter is that the minimum burial age is limited, although area requirements may be affected by factors of 2 to 3 by waste form and vessel size near the minimum burial ages.

Radioactive Waste Treatment and Disposal: Disposal in Deep Wells. — Sodium-calcium-strontium exchange was studied by using Richfield sand as the sorbent. Strontium is selectively sorbed over calcium by a factor of ~1.1, while the selectivity of calcium to sodium sorption, at pH 7, is ~50 g/ml. The normalized strontium breakthrough curves corresponded to the anion breakthrough curves, indicating that strontium breakthrough can be predicted on the basis of solution dispersion.

Radioactive Waste Treatment and Disposal: Disposal in Natural Salt Formations. — Laboratory experiments with salt from the Carey Salt Company mine at Hutchinson, Kansas, have shown that large pieces of salt heated to 260 to 280°C shatter with some violence, accompanied by a release of steam. To evaluate the effect of high temperature on rock salt

in place, a length of 6-5/8-in.-OD pipe, electrically heated to simulate a container of high-level radioactive solids, was placed in a 7-7/8-in.-diam drilled hole in the floor of the Hutchinson mine. After 3-1/2 days of heating, the experiment was terminated because of the presence of water and recrystallized salt in the annulus surrounding the container. Temperatures were measured as high as 127°C (106°C rise) at 9.6 in. from the center of the heater. The surface temperature of the hole, extrapolated from this value, was about 200°C.

Radioactive Waste Treatment and Disposal: Clinch River Study. — During a period of steady flow in the Clinch River of 20,000 cfs, and with the level of Watts Bar Reservoir at 735 ft above mean sea level, the time of travel of Au^{198} in the Clinch River from the mouth of White Oak Creek to Centers Ferry is 9.3 hr, which is about 10% less than that predicted from computation of mean velocity.

The effect of power releases from Norris Reservoir, in which the flow in the Clinch River varied from 1700 to 7700 cfs in 24 hr, on the level of radioactivity at a section 15.3 miles downstream from White Oak Creek was found to be insignificant. The deviation in radioactivity during the period was less than 10% of the average concentration.

In 1963, power releases from Melton Hill Reservoir may cause daily variations in flow of the Clinch River from 0 to 23,000 cfs. At the maximum release rate the level of backwater from the Clinch River into White Oak Creek will be 3.5 ft above present summer levels. Unless the control-gate settings at White Oak Dam are changed, the backwater will flow into White Oak Lake. If the gate levels are raised to prevent inflow from the river, the inflow from the creek to White Oak Lake will raise the lake levels.

Radioactive Waste Treatment and Disposal: Fundamental Studies of Minerals. — To explain the apparently opposite roles of sodium nitrate in removing strontium from calcite and rock phosphate columns, calcium determinations were made by using these minerals and solutions with and without sodium nitrate. The amount of calcium released by calcite and rock phosphate was higher in sodium nitrate solutions. The concentration of calcium detected in 1 M sodium nitrate solution was approximately 1.1 mN; this value is almost four times the solubility of calcite in water. While the dissolution of calcite was the same for three successive leaches, the amount of calcium detected in solution contacted with rock phosphate decreased with each of three successive leaches. Equilibrium was reached within 15 min for calcite particles up to 0.5 mm in diameter; rock phosphate samples showed increasing calcium in the solution with increasing contact time up to 5 hr.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

High-Energy Electronuclear Machines: Cyclotron Analogue II. — The characteristics of the beam in this eight-sector electron cyclotron are being studied in detail. With a dee voltage of only 170 v rms, several microamperes of electrons can be accelerated with very little attenuation to the full energy of 445 kev.

The disagreement between the calculated and measured locations of the sum and difference coupling resonances, $\nu_r - \nu_z = 1$ and $\nu_r + \nu_z = 2$, was resolved. A 1% error was found in a shunt calibration; the locations are in agreement when the sector-coil current is corrected to the proper value. Axial oscillations observed at the sum-coupling resonances and the tendency for the beam to drift away from the median plane were both found to result from a distortion of the median plane of the magnetic field. Although the source of the distortion is unknown, a slight unbalance of currents in the upper and lower halves of the magnet virtually eliminates the effects.

Basic Studies of Cryogenic Electromagnets. — The identical Nb—25% Zr wire used in the 1800-ft magnet previously reported was rewound in a magnet cast in epoxy resin. A copper wire secondary, with a turns ratio of one to one, was also cast in the resin. With this arrangement, currents carried in the 10-mil superconductor ranged between 25 and 30 amp, as compared with 18 to 19 amp in the original coil. The improvement is thought to be due to the rigidity with which the epoxy holds the wire in place. The copper secondary greatly reduces the destructive effects of voltages induced when the NbZr "goes normal." These voltages are reduced by a factor of 10, and current decay in the primary circuit (Nb-Zr wire) is faster by a factor of 10. The field energy is dissipated in the copper secondary circuit. After the coil was driven normal over 500 times, the current capacity dropped to 20 amp, and shortly thereafter to 12 amp.

High-Energy Accelerator Shielding. — Several calculations are being developed to study the transport of high-energy radiation through shielding materials used on high-energy accelerators. In one calculation the longitudinal development of electron-initiated electron-photon cascades was studied for machines which accelerate electrons up to 45 Bev. Another calculation is under development that will use Monte Carlo methods to study the penetration of nucleons, pions, and muons with energies up to 30 Bev; however, it is being preceded by separate intranuclear cascade and nuclear evaporation calculations to provide prerequisite, basic data. Another penetration calculation, which consists of the numerical solution of the Boltzmann equation with the straightahead approximation, was completed and used in the study of the shield for the Stanford University Linear Accelerator.

Physics of Fission. — The pulse-height response of silicon surface-barrier detectors to fission fragments has been found not to be strictly

proportional to fragment energy. A linear relationship in the range of fission-fragment energies is exhibited, but extrapolation to zero pulse height yields a finite and positive energy intercept, analogous to the ionization defect for fission fragments stopped in gases [Phys. Rev. 102, 183 (1956)]. Thus $E = aPH + \delta$, where E is fragment energy, PH is pulse height, and δ for these detectors has been termed [Proceedings of Solid State Nuclear Particle Detectors Conference, Asheville, N.C. (1960)] "pulse height defect."

For most counters exhibiting "good" operating characteristics, δ is constant, independent of counter resistivity and applied bias over a wide range, and is equal to ~8 or ~14 Mev, depending on whether the time-of-flight data of Stein and Whetstone [Phys. Rev. 110, 476 (1958)] or of Milton and Fraser [Phys. Rev. 111, 877 (1958)], are used for the energy calibration. A departure up to δ = 26 Mev was observed for a counter whose surface had not been treated properly and hence was not expected to have optimum characteristics. The value of a was found to depend on silicon resistivity and applied bias, but approached constancy for sufficiently large values of maximum electric field E_0 in the barrier. As the critical value of E_0 was approached, charge multiplication effects became evident.

High-Voltage Experimental Program. — A Moxon-Rae gamma-ray detector (Neutron Time of Flight Methods, EURATOM, Brussels, 1961) has been constructed and used with the ORNL 3-Mv Van de Graaff accelerator. The overall time resolution was about 15 nanoseconds (but should be capable of considerable improvement). By use of the $Li^7(p,n)$ reaction near threshold and a 20-cm flight path, neutron capture in several elements was compared. Cross-section results were obtained for Au, Ag, Cd, Sn, Mo, Pt, W, and Ta. Computer calculations of the relative efficiency of the detector confirm the anticipated proportionality to gamma-ray energy to ±25% over the energy range 0.5 to 8.0 Mev. This range covers most of the gamma rays resulting from neutron capture.

The yields of gamma rays resulting from Coulomb excitation have been measured for 15 even-even nuclei of Ni, Zn, Ge, and Se. Coulomb excitation was produced by variable-energy alpha particles (3 to 10 Mev). In favorable cases the absolute gamma-ray yields are determined to an accuracy within ±4%. The reduced electric quadrupole transition probabilities, B(E2), are generally determined to an accuracy within ±9%. The observed values for B(E2) are larger than the single-particle estimate by factors ranging from 11 for Ni^{58} to 51 for Se^{76}. The 2^+ energies and B(E2) values can be interpreted in terms of the collective vibrational model.

Multiparticle Medium-Energy Nuclear Physics: The 86-Inch Cyclotron. — The differential cross sections for the (p,d) reactions from 22-Mev protons on zirconium isotopes were compared with cross sections calculated by distorted-wave theory. Optical-model parameters were found that yield calculated cross sections which fit the shape of the experimental cross sections. The ratio of the experimental to the theoretical cross section is 0.75 for the $d_{5/2}$ pickup in $Zr^{91}(p,d)$, and the ratio is 0.5 for the $d_{5/2}$ pickup for each of the reactions $Zr^{92}(p,d)$, $Zr^{94}(p,d)$, $Zr^{96}(p,d)$, and $Nb^{93}(p,d)$. Since the theoretical normalization is not known precisely, some adjustment of

the absolute values may be justified. If the Zr^{91}(p,d) ground-state cross section is interpreted as a true measure of the $d_{5/2}$ single-particle pick-up cross section, and if the calculations are normalized to this value, then the cross sections for pickup from the even-neutron isotopes is only two-thirds of the expected value. This is probably an effect of the pairing energy which renders inaccurate the description of the ground states of the targets as simply 2, 4, or 6 noninteracting neutrons in the $d_{5/2}$ shell. The effect of the odd $g_{9/2}$ proton in Nb^{93} is to spread the different final states attributable to $d_{5/2}$ pickup over an energy band of several hundred kev. The reduced width summed over this band is the same as that for the $d_{5/2}$ pickup in Zr^{92}(p,d).

The (p,t) cross sections for the ground-state-to-ground-state transistions on the even-neutron isotopes, Zr^{92}, Zr^{94}, Zr^{96}, and Nb^{93}, are all about the same, unlike the (p,d) cross sections, which increase with neutron number. No theoretical cross sections have been calculated, but the near constancy of the ground-state cross section is plausible on the basis of some crude angular-momentum considerations: Since 2^+ and 4^+ final states as well as the 0^+ ground state can be made by pickup of two $d_{5/2}$ neutrons from Zr^{94} or from Zr^{96}, the total $d_{5/2}$ pickup cross section should be distributed among these three states. The effect of increasing the number of $d_{5/2}$ neutrons in the target from 2 to 4 is to make these higher spin states available to the double-pickup reaction. The increase from 4 neutrons to 6 enhances the cross section for exciting these higher spin states.

Electronuclear Machines: The Isochronous Cyclotron. — The magnet measurement program was completed, and the magnetometer was removed to permit installation of the resonator system. Orbit studies based on these measurement data are in progress; in a typical case it was found that the maximum deviation between the average measured field and the average calculated field was only 8 gauss in about 14,500 gauss.

The assembly of the dee and the liner was completed, and these components were aligned and installed; the resonator is ready for testing. With the resonator installed, the complete system was vacuum-tested and was found to be tight; the pumping speed, base pressure (5×10^{-6} torr), and rate of rise (8×10^{-7} torr/sec) are satisfactory.

Characteristics of the rf system determined from model tests have served as the basis of the design of the complete system, from the master oscillator through the amplifiers to the resonator components. Design of the oscillator and the amplifiers was completed, and the components are being fabricated. In the meantime a temporary oscillator and low-power amplifier system was assembled and is being installed to permit essential preliminary tests of all component systems of the cyclotron.

Mass Spectrometry and Related Techniques. — It has been shown, in a previous investigation of the platinum-catalyzed reaction $D_2 + CO_2 \rightarrow D_2O + CO$, that the mass spectrometer offers a direct method for the identification and study of the volatile transient species. In a study of the decomposition of ammonia on platinum, a transient species NH_4^+ was observed evolving from the catalyst throughout the temperature range 500 to 1200°C. No ionizing electrons were being introduced into the ionization chamber during this observation. The NH_4^+ concentration was measured

as a function of H_2 pressure over a tenfold range, with all other variables held constant, and it was found that the intensity of the NH_4^+ ion increased as a linear function of the H_2 pressure. The fact that the rate of NH_4^+ formation is dependent on the first power rather than the 1/2 power of the H_2 pressure suggests that the ammonia reacts with hydrogen molecules rather than with hydrogen atoms adsorbed on the surface.

Stable Isotopes Development. — Analysis of measurements of the magnetic field of the new inhomogeneous-field separator indicated a maximum deviation of 0.4% from the theoretical field sought. The deviation existed primarily in the region of shorter pole-face radius and would adversely affect the trajectory of ions passing through this region. The trajectories derived from computer analysis of the measured field indicate that an undesirable broadening of the beam focus by ion beams having a divergence of > 5° in this direction may be expected.

The separations of lead isotopes in the inhomogeneous-field separator ultimately produced a sample of Pb^{204} enriched to 86.2%, a twofold increase over the previously obtained enrichment of samples separated in the calutron.

The design of the liner required in the inhomogeneous-field equipment was made final, and construction of a rotating-sector device, to provide multiple filament usage in calutron runs, was initiated. The receiver for use in experimental separations of Bi^{208} and Bi^{210} was designed and assembled.

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} plant has been pressure-tested and is ready for operation.

Special Separations. — The experimental determination of sputtering and saturation values of Ca^{46} on various materials is being intensified. Chemical recovery and refinement of microgram quantities of Ca^{46} from the target and from the remaining pocket area show extreme variations between the purity of Ca^{46} in the target and that in the remaining pocket area.

Charge evaluation studies were performed on neodymium, strontium, and molybdenum. Metallic strontium was superior to any of the strontium compounds for use as a feed material. Neodymium and molybdenum offered improved performance when internally chlorinated.

Ion currents equivalent to 2670 g of separated isotopes were monitored in 28 calutrons. Isotopes of Lu, Ti, Ta, Ca, Re, La, Mo, and Nd were separated and collected from a total of 50,742 g of charge material (433 charges).

Chemical refinement and recovery were completed on 33 new inventory lots of Gd, Lu, Mg, Sm, Sn, Ti, W, and Zr. Striking pads from eight Ta^{181} receiver pockets were processed for sputtering studies. Isotope collector pockets from experimental separations of neodymium, molybdenum, and silver were processed in connection with retention and isotope-enrichment studies.

CHEMISTRY RESEARCH

Thorium Oxide Studies. — The catalytic activity for combination of H_2 and O_2 was determined for a slurry prepared from some of the dried,

irradiated solids recovered from loop L-2-27S. The specific activity under a large excess of O_2 (based on palladium concentration) at 280°C was at least as high as and possibly higher than those obtained with other slurry-palladium catalyst systems which had not been irradiated and more than sufficient to account for the low radiolytic-gas pressures observed during in-pile operation of the loop. At H_2/O_2 ratios $\geqq 0.7$ and after H_2 pretreatment, specific activities were much higher, in accord with previous experience with unirradiated palladium catalyst-slurry systems. Hence the simultaneous reactor irradiation and pumping under O_2 caused no apparent decrease in catalytic activity.

Surface-area and porosity data were obtained on the code P-82 thoria pellets irradiated three months in D_2O at 250°C in the LITR ($\sim 3 \times 10^{13}$ neutrons cm^{-2} sec^{-1}) and on pellets heated in water at 260°C for approximately 2000 hr without irradiation. The irradiated material had a BET (nitrogen-adsorption) surface area of 0.32 m^2/g, a void volume of 1.4%, an average pore radius of 219 A, and a median pore radius of 70 A. The unirradiated pellets had a surface area of 0.023 m^2/g, a void volume of 0.6%, an average pore radius of 758 A, and a median pore radius of 344 A. Fifty-nine percent of the void volume of the irradiated material was in pores of radius $\leqq$ 98 A, and only 15% of the void volume of the unirradiated material was in pores of radius $\leqq$ 112 A. If the data are correct, irradiation of the pellets under water apparently greatly increased the specific surface area, somewhat increased the void volume, and produced a large number of small pores. Despite the increases in surface area and void volume, wear resistance of the irradiated pellets (1-hr standard spouting-bed test) was better by about a factor of 10 than that of the unirradiated pellets (see ORNL-3223).

Equipment Decontamination. — An electrode-potential method was developed for determining the concentration of hydrogen peroxide necessary to induce metal passivity at all formulations and temperatures. Oxalate-peroxide decontaminating solutions were found which were slightly corrosive at room temperature when rubbed on mild steel with a rag or sponge, but to which the steel became passive when rubbing ceased. A typical solution of this type, which also dissolved ordinary rust, was 0.4 M ammonium oxalate—0.2 M ammonium acetate—0.7 M H_2O_2 at pH 2.0.

Acetate was unsuitable as a buffer in oxalate-peroxide solutions for use on carbon steel at elevated temperatures, because of the corrosive nature of the acetic acid in the vapor above the solution. Nonvolatile buffers such as citrate and glycolate were suitable, as was the maintenance of acid reserve by lowering the pH. In the latter case, corrosion was controlled by lowering the temperature and raising the peroxide concentration. For example, the corrosion rate in 0.4 M oxalate—0.4 M H_2O_2 at pH 3.0 and 75°C was 6 mils/hr, but when the peroxide concentration was raised to 1.5 M, the rate dropped to 0.001 mil/hr. Formulas with a higher pH, with the acid reserve provided by a nonvolatile buffer, offer greater safety against corrosion when peroxide concentration drops than do those at lower pH with higher peroxide concentrations.

Protactinium Chemistry. — The separation of uranium from protactinium in HCl-HF solutions by amine extraction was shown to be practical in laboratory tests. In 8 N HCl the organic-to-aqueous distribution coefficient

for protactinium extracted into 0.1 N trilaurylamine in diethylbenzene decreased from 0.9 in 0.01 N HF to 7×10^{-4} in 4 N HF. The corresponding uranium distribution coefficients showed a more complex dependence on HF concentration, decreasing from 300 in 0.01 N HF to about 45 in 0.26 N HF, then increasing to about 100 in 0.8 N HF, and finally decreasing with further rise in HF concentration. Separation of uranium and protactinium with 0.1 N trilaurylamine was best from 8 N HCl–0.8 N HF. The separation factor was 8×10^4 with one equal-volume extraction of a solution containing initially 9 mg of Pa per milliliter and 2 mg of U per milliliter.

Radiation Chemistry of Organic Moderators. – Additional products from pyrolysis of pure biphenyl at 490°C were identified by gas-chromatography techniques.

Hydrogen and methane were present in the low-boiling fraction, and benzene was the only intermediate-boiling component. The following compounds were found in the high-boiling fraction: ortho-terphenyl, meta-terphenyl, para-terphenyl, ortho-quaterphenyl, and meta,para-quaterphenyl. These results indicate both the rupture of the bond between phenyl groups and the degradation of the benzene ring.

Aqueous Solution Chemistry. – Ultracentrifugation of hydrolyzed Pb(II) solutions, 1 M in $NaClO_4$, of hydroxyl number n [moles of hydroxide bound per mole of Pb(II)] = 0 to 1.32, indicated average degrees of polymerization of about four at n = 1. The molecular weights (indicating the degree of polymerization) were lower at lower n and higher at higher n values. These results are generally consistent with a scheme in the literature, based on potential measurements, which postulates $Pb_4(OH)_4^{4+}$ and $Pb_6(OH)_8^{2+}$ as important species in this concentration and pH range.

The nonlinear least-squares technique was used to fit osmotic-coefficient data on a variety of electrolytes to the semiempirical equation

$$\phi = 1 - \frac{\mathcal{S}}{A^3 I}\left[(1 + AI^{1/2}) - 2\ln(1 + AI^{1/2}) - \frac{1}{1 + AI^{1/2}}\right] + BI + CI^2 + DI^3 .$$

From the parameters so obtained, activity coefficients for these electrolytes were computed over a wide range of ionic strength. The method appears very powerful, in that activity coefficients at lower concentrations may be calculated with good accuracy with parameters obtained by fitting osmotic-coefficient data in the range 1 to 3 m. The method is to be greatly preferred over graphic integration if a high-speed computer is available to perform the nonlinear fits.

Radiation Chemistry. – Information on the chemical nature of the intermediates in the radiolysis of water was obtained by comparison of the photolysis and radiolysis of solutions containing H_2O_2, H_2, and O_2. At low H_2O_2 concentration and high light intensity, the overall quantum yield for peroxide decomposition, which proceeds via initial dissociation to OH radicals, is 1.0. On addition of H_2 the quantum yield increases with increasing ratio of concentrations $(H_2)/(H_2O_2)$; hydrogen competes with H_2O_2

for OH radicals. The H atoms produced react with H_2O_2 to re-form OH, thereby raising the quantum yield. The ratio of rate constants in photolysis, $k_{H_2+OH}/k_{H_2O_2+OH} = 0.93$, is the same as that reported previously for the radiolysis of water, indicating that the OH radicals are the same in both systems.

Oxygen added with H_2 lowers the quantum yield, which decreases as $(H_2)/(H_2O_2)$ and $(O_2)/(H_2O_2)$ increase; O_2 competes with H_2O_2 for the H atoms, thereby inhibiting the reaction. The ratio of rate constants in photolysis, $k_{H+H_2O_2}/k_{H+O_2} = 2.2 \times 10^{-3}$, is much lower than that reported for radiolysis, 0.54. This provides additional evidence that the H formed by oxidation of H_2 (called H') is not the same as that produced in the radiation decomposition of water.

Studies of solutions in $HClO_4$ and NaOH showed that neither of the above ratios of rate constants is sensitive to pH in the range ~0.4 to ~9. Above pH 9 the effect of H_2 (and also of $H_2 + O_2$) diminishes. This is probably associated with the conversion of OH to O^-, which is less reactive toward H_2 than is OH. On this assumption, $k_{OH+OH^-}/k_{OH+H_2} \approx 2$. In sulfuric acid solution the effect of H_2 (and $H_2 + O_2$) diminishes below a pH of ~2. Comparison with $HClO_4$ solution showed this to be due to reaction of OH with sulfate to form a species (presumably HSO_4) which is unreactive toward H_2. For H_2-saturated solution (~780 μM) the effect of H_2 diminishes for $(H_2SO_4) > 0.005$ M and is almost eliminated for $(H_2SO_4) > 0.05$ M. The estimated ratio of rate constants $k_{H_2+OH}/k_{H_2SO_4+OH} \approx 200$.

High-Temperature and Structural Chemistry. — The electrical conductivities of mixtures of $NdCl_3$ with $SrCl_2$ and of NdI_3 with SrI_2 exhibited additive behavior and negative deviation from exact additivity respectively. Thus they do not give the strong positive deviation or the maximum shown by mixtures of $NdCl_3$ with $NdCl_2$ and NdI_3 with NdI_2 respectively. This lends considerable support to the earlier suggestion that the positive deviation and the maximum in the Nd(III)-Nd(II) systems are produced by an electron exchange between ions of different valence.

The conductivity of concentrated solutions of lanthanum in molten LaI_3 is very similar to that of the corresponding cerium system, that is, more than 50 times that of the pure molten triiodide, or more than 98% electronic in nature.

The entropies of fusion of the trichlorides of La, Ce, Pr, and Nd were found calorimetrically to be 11.6 ± 0.2 eu. Together with phase-diagram data of the literature, they indicate that these rare-earth metals dissolve in their trichlorides by forming three particles per metal atom, probably mixtures of M^{2+} ions, from $M + 2M^{3+} \rightarrow 3M^{2+}$, and electrons, from $M \rightarrow M^{3+} + 3e^-$, the former reaction greatly prevailing in the Nd-$NdCl_3$ system.

Among the entropies of fusion of several alkaline-earth halides, 6.85 eu for $CaBr_2$ (SnO_2 type of structure), 9.5 for CaI_2 (layer type), 3.39 for $SrCl_2$ (CaF_2 type), 5.81 for SrI_2 (structure unknown), and 6.44 for BaI_2 ($PbCl_2$ type), that for $SrCl_2$ stands out because it is unusually low, but is fairly close to the entropy of 4.2 for CaF_2, which has the same type of structure. The low entropy of fusion in connection with phase-diagram

data suggests solubility, near the melting points of the salts, of NaF and Ca metal in solid CaF_2 and of Sr metal in solid $SrCl_2$, probably in the form of CaF and SrCl.

Transuranium Elements. — Investigation of the solvent-extraction chemistry of 2-ethylhexylphenylphosphonic acid [2-EH(ØP)A] was continued. Both UO_2^{2+} and Pu^{4+} were very strongly extracted from HCl of all concentrations. Their distribution coefficients are 10^2 to 10^3 times as great as that of Cf^{3+}. Distribution coefficients between 1 M 2-EH(ØP)A in diethylbenzene and HCl for Ce, Eu, Tb, and Tm were minimum at 5.5, 6.5, 7.5, and 8 N HCl respectively. Separation factors between adjacent lanthanides remained constant to the minima and then began to converge at higher acidities.

Californium was separated from americium in a countercurrent batch extraction, with californium recovery of > 99.5% and americium removal of > 99.9%. The conditions were: 3 scrub and 4 extraction stages; feed, 1.4 N HCl; scrub, 1.8 N HCl; extractant, 1.0 M 2-EH(ØP)A in diethylbenzene; and feed-to-scrub-to-extraction volume ratios of 1:1:2.

Results of a disconnect selection test program indicated that either of two designs is about equally acceptable. Leak-tight closures were obtained for repeated assemblies, for interchanged halves, and for misalignment and seal-surface marring exceeding that expected for normal use. In accelerated corrosion tests with aqua regia, the connecting lines failed before the seal surfaces leaked.

The use of a modified impact wrench with manipulators was demonstrated for remote disassembly and/or removal of a gang disconnect, an air-operated valve, and a diaphragm pump.

An 0.82-in.-ID pulsed column, operated with 30% Alamine 336 in diethylbenzene as extractant and 11 M LiCl as the aqueous phase, showed stage heights of 16.5 to 19 in. for scrubbing europium from the solvent, and flooding rates of 9 ml/min of organic plus 3 ml/min of aqueous at 25°C and 18 ml/min of organic plus 6 ml/min of aqueous at 50°C.

Uranium-232 Preparation. — Approximately 6.8 mg of U^{232} was prepared and partially purified. Pellets of Al-Pa_2O_5 cermet containing 7.3 g of Pa^{231} and 10.3 g of Al were canned in aluminum and irradiated in the ORR for 10 hr (nvt = 3.5×10^{18}). After a few hours' cooling, the irradiated slug was transferred to a shielded cell, where the aluminum can was mechanically removed and the pellets were processed for separation and recovery of the Pa^{231} and U^{232}. The process included dissolution of aluminum in 8 M HCl, dissolution of Pa_2O_5 in 8 M HCl–0.6 M HF, selective sorption of uranium on the anion exchange resin column, and elution of uranium product into 0.5 M HCl. Trace protactinium and uranium in the aluminum solution were sorbed on an anion exchange resin.

The time interval between irradiation and processing must be chosen to allow most of the Pa^{232} (1.32-day half-life) to decay to U^{232} but to limit the amount of Pa^{233} (27.4-day half-life) that decays to U^{233}. A protactinium-uranium separation after 45 hr of decay produced 5 mg of U^{232} containing 130 ppm U^{233}, 90 ppm U^{235}, and 12,200 ppm U^{238}. The U^{235} and U^{238} contamination came from the Pa_2O_5, which contained about 8 ppm of natural uranium. A second Pa-U separation, 93 hr after the irradiation, produced an additional 1.8 mg of U^{232}, whose isotopic purity has not yet been determined. Both uranium products require additional purification

from protactinium and iron, for which a tertiary amine extraction is being developed.

Target Preparation. — A 15-in. Fe^{57} target consisting of 56 pieces of foil was prepared.

Forty-eight B^{10} targets with thicknesses of 200 $\mu g/cm^2$ were prepared by using the electron furnace.

The quality of enriched iron targets was improved by introducing iron wool in the hydrogen-reduction tube immediately in front of the material being reduced. The wool removed trace nitrogen from the hydrogen stream, thereby precluding any nitriding of the enriched iron sample during reduction.

The equipment for ion deceleration was reassembled and is ready for use in target-formation experiments after termination of rhenium separations.

METALLURGY AND MATERIALS RESEARCH

Research and Development on Pure Materials. — A 300-g ingot of Li^7F consisting of two crystals was produced in the previously described (ORNL-3190) Stockbarger furnace. The starting material was highly purified, dehydrated, hand-picked Li^7F (ORNL-3242). The uncleaved crystals were loaned to R. L. Sproull, director of the Materials Science Center at Cornell University, for low-temperature thermal-conductivity studies.

Equiaxial, 0.5-mm crystals of MgO, larger than those previously reported (ORNL-3242), have been grown from eutectic LiF-NaF and from eutectic $LiF-NaF-MgF_2$. Monoclinic crystals of ZrO_2 up to $1 \times 2 \times 0.02$ mm, ordinarily difficult to obtain by solid-state transformation from cubic ZrO_2, have been grown from molten $NaF-ZrF_4$. Crystals of ZrF_4, $2.5 \times 2.5 \times 5$ mm, were obtained in the same experiment.

Spectroscopy of Ionic Media. — Studies are being continued on the influence of ionic environments on the lowest-energy electronic transition of the nitrate ion. The Franck-Condon transition energy $h\nu$ for molten mixtures of alkali metal, alkaline-earth metal (except Mg^{2+}), and La^{3+} nitrates was found to obey the relation, at 360°C,

$$h\nu = 0.34 \sum (z_i/r_i)\overline{N}_i + 3.81 ,$$

where $h\nu$ is in electron volts, z_i, r_i, and $\overline{N}_i$ are the charge number, ionic radius (in A), and electrical equivalent fraction, respectively, of the ith species of cation, and the summation is over all species of cations.

Fundamental Investigation of Radiation Effects in Solids. — Through a series of irradiations of several varieties of fused silica with energetic particles, followed by light bleaching and heat treatments, a one-to-one correspondence was found between an optical band at 2120 A and an electron-spin resonance line, g = 2.0006. A comparison was then made between the concentration of centers in this ESR line determined by an ESR

comparison technique and the total concentration measured by a magnetic-susceptibility method. The results of the comparison show that the primary paramagnetic center and the center which gives the ESR line, g = 2.0006, are the same. This center has been called the E_1' center. After appropriate irradiations and subsequent bleaching treatments, the concentration of spins in the E_1' center was measured by the susceptibility technique. The absorption coefficient and the width at half-maximum absorption coefficient were found, and with Smakula's equation, appropriate for a Gaussian function, the oscillator strength, f, was found to be 0.14 ± 0.04.

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. Techniques for reducing the turbulence on the plasma formed at the juncture of an electron beam and a directed gas jet (in a vacuum environment) are being investigated. Generally speaking, tubes of magnetic flux can move across a conducting material only very slowly. It was thought that the turbulence of the plasma would be reduced if a conducting mass (copper was used) were placed in close proximity to the plasma and arranged so that the magnetic flux tubes which were disturbed by the turbulent plasma passed through the copper. The effect was positive. With the copper in place, a plasma which had been turbulent at magnetic field strengths greater than 400 gauss did not become turbulent until the field strength was 800 gauss. In addition, the manner of reducing the turbulence was such that the low-density regions of the plasma were filled. That is, both the average plasma temperature and average density appeared to be raised slightly. Carbon was substituted for the copper to see if its lower conductivity would be apparent. It was, and electrostatic effects were, therefore, apparently small.

DCX-1 Facility. — Most of the operation during this period was without Zeolite in the inner vacuum region. Apparently, the recent improvements in vacuum conditions were primarily the result of the revised geometry of the new vacuum liner and the higher-temperature bakeouts — not the result of the Zeolite. Pressures of about 10^{-9} torr in the liner region are reached more easily in the absence of Zeolite. The lowest liner pressure has been 1.9×10^{-9} torr, where proton-containment times of about 37 sec were measured with an injected H_2^+ beam of 2.5 ma.

More studies were made of the responses of the neutral-particle detector signals during periods of plasma accumulation (buildup) and decay. The observation of linear buildups with only 150 ma of injected H_2^+ current strongly suggested that these buildups were not evidence of dissociation on the trapped protons, as originally suggested. A second model which attributed the linear buildups to dissociation on an initially dense, cold plasma was discounted by measurements of the onset times of rf signals at harmonics of the proton cyclotron frequency. (These onset times are the time intervals between gating on the H_2^+ beam and the observation of significant radiation at the successive harmonics.) Relating these onset

times to the accumulation of electron density in the plasma by the Harris theory indicated only the usual gradual increase in electron density after gating on the H_2^+ beam.

Definite time delays in the buildup responses of neutral-particle detectors located off the median plane were recently observed. Under conditions that produce the linear buildups, the delay in the response of a detector 2 in. off the median plane, as compared with one on the median plane, is of the order of several seconds. A shorter delay is observed for a detector 1 in. off the median plane. These responses indicate a slow, gradual expansion of the plasma volume during the buildup period, and it has been demonstrated that such an expansion can account for many features of the neutral-particle detector buildup traces. Experiments are planned to more accurately assess the rate and magnitude of this volume expansion.

DCX-2 Facility. – Preliminary tests have begun in the DCX-2 facility during this report period. Initial experiments with the injector had to be halted because of failure of a turn in the injector-compensating coil, resulting from a water-passage block. The injector was repaired and reassembled, and a beam of up to 20 ma (with unknown mass distribution) at 500 kv was injected into the plasma volume with the full magnetic field applied. Visual observations were made to establish the injection duct by means of a probe swept through the beam. Further experiments relating to the injection trajectory are anticipated before plasma buildup measurements will be attempted.

While the injector was being repaired, both argon and lithium backstream arcs were run in the vacuum tank, a distance of ~7 m. The power radiated by the argon arc was 4 kw/m with operating characteristics of 150 amp and 250 v. The lithium-arc run was terminated because of a minor construction failure – although sufficient running time was attained to show the feasibility and reliability of the use of such a discharge in the DCX-2 geometry.

The pressure in the system has been improved by repairing various small leaks. The best pressure observed in the liner region was 2.8×10^{-6} mm Hg.

Vacuum. – A regulated, low-emission modification to the standard Veeco RG 31A ion-gage control was made and subjected to preliminary testing on both bare and enveloped ion gages. At emissions of about 8 ma the wall potential appears to be close to zero with respect to ground. This enabled measurement of the residual ion current in a tubulated gage; it was equal to about 10^{-10} amp of grid-collector leakage.

Preliminary work with a mass spectrometer to study oil decomposition in diffusion pumps led to the determination of significant amounts of ethane, as well as methane and higher hydrocarbon residues, above an operating, fractionating diffusion pump containing octyl phthalate. The Consolidated Vacuum Corporation's model No. 21612 mass spectrometer apparently has an as-received sensitivity of 2×10^{-9} torr and unit mass resolution to about mass 28.

Studies of the experimental pump using surface-boiling oil were continued. The pump was designed specifically to use polyphenyl ether oils. In this assembly the oil throughput into the top jet of the three-stage

pump can be varied over a considerable range without greatly affecting the throughput in the two lower stages. Pumping-speed measurements for hydrogen indicated that the pumping speed was indeed a strong function of oil throughput to the top jet as monitored by a thermocouple in the jet stream. Optimum operating characteristics for the surface heaters were obtained.

BIOLOGY AND MEDICINE PROGRAM

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Physics of Tissue Damage. — The recent availability of inexpensive optical interference filters has made possible the study of emission spectra from extended sources too weak for conventional spectroscopic techniques, if high resolution is not required. Because many spectra from biochemicals such as amino acids and proteins are continua, and large areas of sources are available, such filters may be used with a photomultiplier detector to form a simple, very-high-aperture spectrometer. Fifty filters were mounted around the periphery of a rotating disk 1 m in diameter which is enclosed in a light-tight housing having access ports for source and detector. Because the transmission profile depends somewhat on the angle and point of incidence of light on the filter, calibration was carried out by using a translucent diffuser source equal to the sample source in diameter (5 cm) and at the source position; the calibration source was illuminated by light from a monochromator. Most filters had a transmission profile in the shape of an isosceles triangle approximately 30% high at the peak and 100 to 150 A wide at half height for source-to-multiplier spacings from 36 to 10 cm respectively. The spectrometer spans the spectral region from 2400 to 7600 A.

ENVIRONMENT RADIATION STUDIES

Terrestrial Ecology: Evaluation of Fission Product Distribution and Movement in White Oak Drainage System. — Water-level measurements, taken in observation wells in the former bed of White Oak Lake, indicate that the depth from land surface to ground water varies from < 1 ft to 5 ft. Water-contour maps show that the general direction of ground-water movement is normal to the flow of White Oak Creek in the lower lake bed, but the pattern in the upper portion is influenced by stream flow from the intermediate-level waste-seepage pit area.

Terrestrial Ecology: Blood Responses of Mammals in Radioactive Waste Areas. — Calibration and operating procedures for an electronic blood counter were completed. Diluting solutions recommended for blood from humans and from laboratory rats and mice were found to be unsatisfactory. A successful method was developed which consisted in initially diluting the blood sample with acetate-buffered saline. Subsequent treatment for erythrocyte or leukocyte counting utilized a gelatine saline or Triton X-100 (Rohm and Haas) solution. These provided the required stability for both total counts and cell-size distribution.

Mean values for erythrocyte (RBC) and leukocyte (WBC) counts and for microhematocrits have been established for several species of wild rodents. The erythrocyte counts ($\times 10^6/mm^3$) of cotton rats, rice rats, house mice, and white-footed mice were, respectively, 7.1, 8.61, 10.76, and 12.03. The leukocyte counts ($\times 10^3/mm^3$) of these species were 3.7, 3.8, 7.6, and 3.6 respectively. The microhematocrit values were 47.2, 50.8, 48.9, and 48.3% respectively. Intraspecies variation of erythrocyte counts was small, and it appears that interspecific differences in erythrocyte numbers are inversely related to species body size.

A cotton rat with a squamous cell carcinoma involving an inguinal mamma had an RBC count of $6.4 \times 10^6/mm^3$, while the WBC count was $19.3 \times 10^3/mm^3$. A white-footed mouse infested with two parasitic bot-fly larvae also had anemia (9.30×10^6 RBC/mm^3) and leukocytosis (13.7×10^3 WBC/mm^3). Acclimation of adult rats to winter temperatures for one month after being kept indoors at 80°F did not affect RBC or WBC counts.

Terrestrial Ecology: Radionuclide Uptake by Mammals in Radioactive Waste Areas. — Significant concentrations of ten radionuclides have been detected in whole carcasses (less GI tracts) of 26 wild, small mammals trapped from White Oak Lake bed. The nuclides Sr^{90} and Co^{60} were found in all animals; Ru^{106} and Cs^{137} were found in all animals except one. The occurrence of Cs^{134}, Zn^{65}, Zr^{95}-Nb^{95}, and Ba^{140}-La^{140} was limited to a smaller number of the animals. All of the isotopes mentioned except Ba^{140}-La^{140} have previously been found in mammals from the lake bed. The presence of Ba^{140}-La^{140} can no doubt be attributed to waste released at ORNL and not to nuclear testing, because the sample collections antedate the recent atmospheric series by Russia.

Ruthenium-106 concentrations in whole caracsses averaged higher than the concentrations of the other isotopes and were highest in rice rats (500 to 8000 $\mu\mu c/g$, dry carcass). These concentrations of Ru^{106} are explained on the basis of the environmental behavior of released ruthenium and the semiaquatic habitat requirements of rice rats. The Ru^{106} seeps through the Conasauga shale formation underlying the liquid-waste pit system and reaches the lake bed in a highly mobile chemical state. This radioruthenium can be readily detected in surface seeps on the upper lake bed, which constitute an ideal habitat for rice rats. The high Ru^{106} in rice rats from White Oak Lake bed indicates that they confine most of their feeding to the seepage areas. The body burdens of all the mammals are being used to calculate dose rates from internal sources to aid in the evaluation of histological samples taken from the same specimens for possible correlation of radiation-induced pathologies.

Terrestrial Ecology: Development of ECOS-A, an Analog Environmental Computer, Organizer, and Simulator. — The ORNL Analog Computer Facility has provided methods of simulating continuous changes in mass M_J and content of material X_J per unit area of earth surface in compartment J of a model ecological system. One operational amplifier per compartment integrates the algebraic sum of inputs and losses for that compartment, which are simulated by voltages. The losses are simulated by a negative feedback of the output voltage of the integrating amplifier. Earlier work was with simple models which assumed linear differential equations with

constant coefficients. Later studies introduced variable coefficients by use of servomultipliers driven by periodic functions of time. These simulate seasonal changes in rates of growth and transfer rates of materials in the ecosystem. Input and loss rates for plant, animal, and soil or other compartments are set on potentiometers connected between circuit components and subcircuits. The structure of the system can be changed by rewiring removable patch panels which control the three consoles of the computer facility. Many tracer experiments and related studies of the field ecology program at ORNL provide information on the rate parameters and boundary conditions for eventual use with this analog model, or for similar digital computer models.

Terrestrial Ecology: Clinch River Studies. — Knowledge of the distribution of chemical elements in organisms is essential for an understanding of biogeochemical cycles and for an interpretation of the behavior of radionuclides released to the environment. The strontium content of clamshells ($CaCO_3$) is directly proportional to growth rate for several species. Additional analyses showed that the strontium concentration is not homogeneous throughout clamshells. For example, in five *Elliptio crassidens* shells the laminar nacreous layers deposited during the first six years averaged 237.7 ppm of strontium per gram, whereas layers deposited in years seven through nine averaged 300 ppm of strontium per gram. Maximum and minimum strontium concentrations in the laminar nacreous layers were 370 and 215 ppm respectively. The average strontium content of peripheral nacreous layers was 219 ppm, and there was no apparent change in the content with age. The range of strontium concentrations in the peripheral layers was from 180 to 255 ppm. Barium and manganese have distribution patterns similar to that of strontium, while the calcium and sodium concentrations are constant with age in both nacreous layers.

Terrestrial Ecology: Radiation Effects on Aquatic Insects. — Larvae of *Chironomus tentans* (a midge) live in the radioactive bottom sediments of White Oak Creek and the Clinch River, and are exposed to ionizing radiation 20 to 1000 times that of natural background. The frequencies of chromosomal aberrations in salivary-gland chromosomes are being compared in irradiated and nonirradiated populations to determine whether there is an effect of the increased radiation levels on the genetic constitution of the natural populations. Permanent slides are prepared of salivary glands of larvae from each population, and are studied microscopically and scored for the different aberrations observed. A comparison was made of the salivary-gland chromosome slides prepared during October 1961. The 33 larvae collected from White Oak Creek had a total of 43 aberrations of 12 types, and 29 larvae from the control area had a total of 4 aberrations of 4 types. There is a significant difference ($P < 0.001$) between the total number of aberrations observed in the contaminated and the control areas.

Terrestrial Ecology: Influence of Microflora on Turnover of Isotopes in Leaf Litter. — Laboratory experiments showed that fungal growth on decaying leaf litter reduced the rate of loss of radioisotopes from the litter. Isotope loss from sterile litter of tulip poplar (*Liriodendron tulipifera*) increased in the order Co^{60}, Fe^{59}, Ca^{45}, Cs^{134}, Zn^{65}. The percent

of the initial amount of radioisotope protected by fungal mycelium against leaching from leaves increased in the same order from 5% for Co^{60} to 40% for Zn^{65} in cultures 4 weeks old. Almost all of the isotope uptake by fungi occurred during the first 3 days of their growth. After 2 weeks, part of the mycelium became senescent, and release of radioisotopes slightly exceeded uptake. After 4 weeks of growth and leaching, the concentration of Cs^{134} in the mycelium was calculated to be 76 times as high as in the leaves. Results of field experiments differ from these laboratory findings in that Zn^{65} was retained in the litter and was lost at rates closer to those for Co^{60} and Fe^{59} than to the rate for Cs^{134}. Uptake and retention of Cs^{134} by fungi growing on litter of different leaf species were positively correlated with microbial respiration on the leaves, and decreased in the order tulip poplar, redbud, white oak. The percent of Cs^{134} leached from sterile leaves decreased in the same order.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Radiation Physics and Dosimetry: Performance of the Keplertron, a Spherical Electrostatic Analyzer. – Theoretical characteristics of the spherical-condenser charged-particle spectrometer have been detailed recently in the literature. Such a device has been constructed from concentric spherical shells 18.75 and 12.5 in. in diameter. Expected transmissions and energy resolutions are 25% and 6.3% for a point source, and 16% and 7.7% for a disk source 3.75 in. in diameter. The spectrometer was designed to operate from a few electron volts to 40 kev and utilizes a Faraday cup as a detector. Preliminary calibration was carried out with an electron gun that approximated a point radioactive source over an energy range from 1 ev to 3 kev. Transmission, resolution, and line profile compare well with theory if the transmission is corrected for that of a grid which electrostatically shields the Faraday cup from the outer sphere. The spectrometer will be used to measure slowing-down spectra of electrons and positrons in matter exposed to high-energy radiation.

Radiation Physics and Dosimetry: A Calibrator for Concave Diffraction Gratings. – Recent studies of the emission of light from electron-irradiated solids have emphasized the importance of a knowledge of both the absolute and relative transmissions of grating spectrometers. In the visible and near ultraviolet, these may be obtained from a tungsten light source emitting a known black-body spectrum. In the far-ultraviolet and soft x-ray regions, the emission of tungsten is too weak to be usable; transmission must be determined as the product of photomultiplier and grating efficiencies. The photomultiplier efficiency is known to be constant if the photomultiplier is coated with sodium salicylate; the grating efficiency can be simply determined with a unique grating calibrator just completed. A vacuum-tight aluminum box contains an entrance slit, grating holder, exit slit, and photomultiplier, all in the Seya-Namioka geometry. The exit slit and photomultiplier are attached to a movable arm, which enables them to be rotated from the normal position until the exit slit is coincident with the entrance slit. Light, from another monochromator, which

passes the entrance slit may be monitored successively at the two positions of the arm, and the ratio of photomultiplier currents is the grating efficiency at that wavelength. Gratings have been studied from 1000 to 6000 A and have shown large differences in efficiency over this range.

Radiation Physics and Dosimetry: Interaction of Subexcitation Electrons with Water Vapor. — In an electron swarm experiment it was found that electrons are captured by H_2O vapor in a resonance process peaking at 6.4 ev, with an energy-integrated cross section equal to 7.7×10^{-18} cm^2 ev.

Swarm experiments were carried out for electrons in the energy range below 1 ev by suitable mixtures of H_2O vapor with gases such as N_2, C_2H_4, and CH_4. These "subdissociative electrons" are not captured to form negative ions with H_2O; therefore H_2O^- probably does not form as a stable ion. It was found, however, that the transport time for electrons in pure C_2H_4 or CH_4 is greatly increased by the addition of small quantities of H_2O vapor. This leads to a suggestion of "collisions" of long duration in water vapor, due to its permanent electric dipole, and to the prediction that other polar molecules should similarly affect the transport time in C_2H_4. This effect has been observed for methyl alcohol and for acetone; in fact, a good correlation of the increase in transport time with the strength of the electric dipole moment has been found. An electron time-of-flight apparatus was developed and used in an effort to measure the lifetime of the long-duration collisions in water. The results were negative, suggesting that the lifetime is less than 10^{-7} sec, the time resolution of the apparatus.

ISOTOPE DEVELOPMENT PROGRAM

Neutron Activation Analysis Research. — Study on applications of radioisotope-beryllium neutron sources was continued. Eight neutron sources (each consisting of 0.6 g of Am^{241} mixed with 6 g of beryllium), with an average neutron yield of 4×10^6 neutrons/sec, were fabricated, and their applicability in neutron activation analysis was evaluated. With these eight sources in optimum array, a neutron-activation efficiency was obtained which is at least twice that possible with an equivalent single large radioisotope-beryllium source. Consequently, sensitivity can be substantially improved.

The optimum configuration and spacing for an eight-source array were established by measuring the thermal-neutron flux distribution about a single neutron source and various multisource configurations in water. The activation unit selected consists of eight sources uniformly spaced in two parallel planes perpendicular to the axis of a 5-cm-diam well 23 cm in depth, in a 12 by 17 by 19 in. paraffin block. In each plane, four sources are spaced 90° apart on a circle 5 cm from the axis of the well. The distance between the planes is 7.7 cm. This configuration produces a flux of 8×10^4 thermal neutrons cm^{-2} sec^{-1}, which is very nearly uniform throughout a volume of 180 cm^3.

In activation analysis of a 100-ml sample, with thermal neutrons from this array, the limits of detection were 10 ppm Hf; 5 ppm Au, Mn, or V; 3 ppm In.

Neutron-source units of the type described above in combination with devices for measurement of radioactivity, chiefly gamma spectrometers, will probably prove most valuable for activation analyses where rapid and simple analyses are required. For example, small amounts of hafnium in zirconium can be determined in a few minutes. This neutron-source unit, in conjunction with a battery-operated gamma spectrometer, will probably be found useful, also, as a portable field analyzer by agronomists in the study of the role of elements such as manganese, aluminum, or sodium; by geochemists in the examination of minerals; and in similar applications. Such uses are being considered in the evaluation of the source unit; consideration is being given to irradiation time, precision, and economy relative to conventional analytical methods.

Recovery of Fission Products. — Preliminary tests showed that o-phenylphenol has greater ability to separate cesium from sodium and better phase separation behavior than p-dodecylphenol previously tested, and may be suitable for recovering cesium from adjusted Purex 1WW waste solution. More than 99% of the cesium was extracted from tartrate-complexed simulated high-acid 1WW solution, adjusted with caustic to a pH of ~12, in a five-stage batch countercurrent test (organic-to-aqueous distribution coefficient = 3) with 1 M o-phenylphenol in xylene. The cesium was quantitatively stripped in a single contact with 0.1 volume of 0.1 N HNO_3, resulting in an overall Cs/Na separation factor of ~10^3 and a cesium concentration factor of ~3. In separate batch tests, Sr, Eu, Ru, and Zr-Nb were not appreciably extracted.

In a single laboratory test, a double precipitation from synthetic Purex 1WW waste solution recovered 75 to 80% of the strontium with < 1% each of the original Fe^{3+}, Al^{3+}, Na^{+}, and SO_4^{2-}. Sulfate was removed by precipitation after addition of excess ferric ion and increase of the HNO_3 concentration to about 65 wt % by the addition of fuming HNO_3. Strontium was precipitated from the almost pure nitrate system by concentrating the filtrate from the sulfate-precipitation step to a boiling point of ~135°C and diluting the concentrate with fuming HNO_3 to more than 85 wt % HNO_3. The strontium in the nitrate precipitate was dissolved in 1 M HNO_3 to provide ion exchange feed solution.

The strontium yield could probably be increased from 75–80% to 80–90% by minimizing the volume of more than 85% nitric acid from which the nitrates were precipitated, since extrapolated solubility data of the $Sr(NO_3)_2$–HNO_3–H_2O system indicated that more than 15% of the 20 to 25% unrecovered strontium had dissolved in the large volume used.

Radioisotope Research and Development. — A procedure for completely coating the Decalso radioisotope standards with thin metallic nickel (from 0.5-mil to several mils thick) was developed at ORGDP. Attentuation of the radiation from Sr^{90} and Cs^{137} is being evaluated.

A procedure for preparing cesium borosilicate polyglass was developed; it consists of precipitation of cesium as tetraphenyl boron in the presence of SiO_2, calcining to remove the phenol, and then fusing to a glass at 1200°C. The resulting product was low in solubility, yet relatively high in activity.

In a differential extraction process for recovering uranium from spent HRT fuel elements, 15 kg of uranium was recovered from 100 liters of solution with an efficiency of > 99.5% and a decontamination factor of > 1×10^6.

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