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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JANUARY 1964

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ORNL-3568

Contract No. W-7405-eng-26

OAK RIDGE NATIONAL LABORATORY

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DATE ISSUED

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Oak Ridge, Tennessee
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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JANUARY 1964

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

REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

EGCR. — Analysis of the transient response of a reflected heterogeneous reactor to a pulse of neutrons is continuing. We made improvements to the TRAN code which facilitate its use or permit a more precise description of the pulsed source. The code was then applied to analysis of the pulser experiments carried out in the British experimental reactor HERO.

We modified the KAY-CHAIN program for computing the multiplication factor of heterogeneous reactors to reduce and simplify the required input data. The output was expanded to include two-group cross sections and diffusion coefficients, as well as the critical buckling.

Control-rod worth calculations were carried out for various rod patterns in partially loaded cores, and we studied the variation with core size of the worth of certain rods required for reactor physics experiments.

Fast Gas-Cooled Reactor. — Physics studies performed as part of an investigation of fast gas-cooled reactors included a survey of helium-cooled and SO_2-cooled reactors as a function of coolant volume fraction, fuel-to-structure volume ratios, and fuel specific power; more detailed exploration of the physics characteristics of a reference helium-cooled reactor and of a sodium-cooled reactor proposed by the General Electric Company (for purposes of comparison); and a brief inquiry into the effect on reactor performance of several different structural metals. Only a few of the principal results are summarized here.

The reference helium-cooled reactor, selected as a reasonable tentative solution to the usual core design problems of heat transfer, criticality, fuel utilization, mechanical integrity, etc., comprises a core of steel-clad UO_2-PuO_2 pins, 0.22 in. in diameter, with axial extensions of depleted UO_2, surrounded by a reflector of similar but larger pins of depleted UO_2. Core height and diameter are 6.0 and 10.8 ft, respectively, the coolant volume fraction is 0.67, and the oxide-to-steel volume ratio

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is 1.68, allowing for cladding and structure. The radial blanket thickness is 18 in., and the axial blanket thickness is 15 in. Steel reflectors 4 in. thick in the radial direction and 12 in. thick in the axial direction help to conserve neutrons. These design choices result in a fuel feed enrichment of 10.23% to achieve 100,000 Mwd/metric ton burnup, a conversion ratio of 1.36, and an estimated fuel-cycle cost of 0.7 mill/kwhr (electrical), including a plutonium credit of 0.4 mill/kwhr (electrical). It is estimated, by comparison, that a sodium-cooled reactor would have a fuel cost 0.1 mill/kwhr (electrical) higher and a breeding gain about 0.1 lower than the helium-cooled reactor; however, neither is fully optimized, and the comparison must be considered tentative.

A comparison of different structural metals (in a simplified spherical geometry) indicated the following breeding ratios (significant only on a relative basis):

Metal	Enrichment (%)	Median Fission Energy (kev)	Breeding Ratio
Iron	9.38	202	1.61
Type 347 stainless steel	9.52	204	1.57
0.7 Ni–0.3 Fe	9.97	211	1.44
Nickel	10.32	212	1.35
Niobium	13.07	248	1.01
Tungsten	16.21	235	0.78

We have estimated the reactivity effect of complete removal of the coolant to be +0.35% $\delta k/k$ for helium and +0.77% $\delta k/k$ for sodium. We recognized, of course, that the choice of core proportions greatly influences this void coefficient in the case of sodium, and presumably also does in the case of helium. However, the reactivity effect of the coolant is expected to place much less restraint on the core design with helium than with sodium; this, of course, is one of the principal reasons for considering helium, since no purely local density changes are possible with helium, as they are with sodium.

Fueled-Graphite Reactors. — The new multigroup, space-independent depletion code LTM was used to compare large gas-cooled graphite-moderated reactors operating on the U^{235}-Th cycle (no U^{233} recycle) with similar reactors operating on partially enriched uranium feed. Optimum compositions were identified for assumed fabrication and reprocessing costs of $50 (each) per kg of heavy element (uranium or thorium) and also for $500/kg for fabrication and for reprocessing; in the latter case, reprocessing was not worthwhile, and a throw-away cycle was adopted. Results may be tabulated as follows, where "A" refers to the U^{235}-Th feed, and "B" refers to partially enriched uranium:

	A	B
$50/kg		
Exposure, Mwd/metric ton	87,000	102,000
Average conversion ratio	0.720	0.633
Fuel credit, mills/kwhr (electrical)	0.33	0.09
Net fuel-cycle cost, mills/kwhr (electrical)	0.73	0.85
$500/kg		
Exposure, Mwd/metric ton	193,000	189,000
Average conversion ratio	0.555	0.569
Net fuel-cycle cost, mills/kwhr (electrical)	1.32	1.24

The differences are not large and suggest that other factors, such as reactor safety considerations, may prove to be determining. Fuel recycle must also be considered, and comparisons of recycle cases are under way.

ORR Poolside Capsule Irradiations. — Tests of three capsules containing 6-cm graphite spheres fueled with pyrolytic-carbon-coated UC_2 particles continue. One capsule has accumulated a burnup of ~6%; the other two, which we installed last month, have only low burnup accumulation. The release of fission gas from these capsules is slightly higher than in previous irradiations with smaller spheres, but it is within AVR (German pebble-bed reactor) requirements. The spheres in these capsules are operating at central temperatures of ~2500°F.

Eight-Ball Experiment. — The third eight-ball experimental assembly containing 1-1/2-in.-diam spheres fueled with pyrolytic-carbon-coated UC_2 fuel particles is operating at sphere surface temperatures of ~1700°F. We installed this assembly in the ORR last month.

Studies of Stress Distributions in Shells. — A theoretical analysis is being made of the stress distribution in an axisymmetrical shell, which is reasonably close to being a sphere. The shell was subjected to a uniform external pressure. We are making this study to find a theory with which to assess the effects of imperfections in a spherical shell and to form the basis of a stepwise integration procedure in the study of linear creep deformation of initially spherical shells. A computer program was written to perform the analysis, and convergence tests are currently being made.

REACTOR FUELS AND MATERIALS

Nondestructive Test Development. — In modern high-performance reactors, uneven fuel distribution in a fuel element can cause local hot

spots and, hence, failure. Therefore, 100% inspection of homogeneity in fuel pieces for these reactors is required. We are continuing the development of techniques, using x- and gamma-ray attenuation for the determination of fuel homogeneity in rods and plates. A versatile scanning apparatus (ORNL-3436, p. 5) has been used most extensively for fuel plates containing U_3O_8-Al dispersion cores. The speed of this scanner, 20 in./min, has been the maximum that could be followed by the strip-chart recorder used to give a complete record of the fuel distribution. This record was needed for adequate evaluation of both the instrument and the fuel plates during their development. The need now is for as rapid a test as possible, but only as to whether a plate is acceptable or not. We have now developed more sophisticated prototype instrumentation, which will allow automatic comparison to preset references, signal integration, and digital printout of signal level. When certain modifications are made to the circuitry, faster scanning speeds of up to 200 in./min can be used.

Materials Compatibility Studies. — The corrosion resistance of refractory metals to alkali metals can be seriously impaired by the presence of oxygen in the liquid metal. Potassium is being considered as the working fluid in several proposed space reactors. In order to improve the lifetime and reliability of such systems, we are studying methods to remove and to analyze for oxygen in potassium.

Recent bench-scale experiments have demonstrated that vacuum distillation is an effective technique for both potassium purification and analysis for impurities in potassium. We have studied the accuracy of this method for determining oxygen in potassium by adding a known quantity of K_2O, equivalent to 500 ppm of oxygen, to approximately 5 g of liquid potassium (70°C) contained in a nickel capsule. The sample was then distilled to dryness at 350°C under an average pressure of 5×10^{-5} torr. The residue from this sample was analyzed and compared with the residue from an unspiked potassium sample distilled in an identical manner. Complete recovery of the oxygen addition was obtained by this procedure.

In systems using potassium as the heat transfer fluid, it is important to know that an accurate analysis for oxygen in the alkali metal is independent of the prior thermal history of the potassium. Accordingly, controlled additions of K_2O were made to potassium, and the mixtures were thermally equilibrated in nickel capsules for 48 hr at 400 and 800°C. Analysis of these samples by vacuum distillation, taking the residue to be K_2O, indicated that only part of the oxygen was recovered: 77% for the sample equilibrated at 400°C and 65% for the 800°C test. We are now evaluating the reasons for these low results. The effects of metallic impurities in the potassium whose oxides are more stable than K_2O were first considered, but no impurities were detected in sufficient quantity to account for the observed deviations. An analysis of the nickel capsules showed the same oxygen content as before the test. However, the concentration of carbon in the nickel had decreased from 300 to 200 ppm. If elemental carbon is present and in intimate contact with K_2O during distillation, calculations indicate that oxygen can react to form CO. This possible mechanism of oxygen removal during vacuum distillation is being further investigated.

Fuel Element Development. — The high-temperature stability of uranium monocarbide makes this material a desirable fuel for advanced reactor systems. However, production of dense bodies of this material by conventional methods requires excessively high sintering temperatures (at least 1900°C) and still fails to produce a completely satisfactory product. We have previously reported (ORNL-3368, p. 8) highly successful sintering of uranium monocarbide at 1350°C, using UAl_2 as a sintering aid, during which the aluminum volatilized from the compact. We have now discovered another aided-sintering process, which appears more suited to routine production and yields excellent pellets with densities greater than 97% of theoretical. A charge containing 1 wt % U_3Si_2 is sintered in vacuum at 1550°C in the presence of a small concentration of beryllium vapor, which scavenges contaminating films from the surfaces of the powder particles. The silicide dissolves in the carbide to form a solid solution. An attractive feature is that equally good results are obtained whether the uranium monocarbide is powdered arc-cast material or is the commercial material made by reduction of UO_2 with graphite. The process is easily controlled, and there appear to be no serious limits to pellet or batch size.

REACTOR COMPONENTS

Reactor Controls Development. — The multichannel fluctuation analyzer described previously (ORNL-3517, p. 7) has been installed and tested at the Pool Critical Assembly (PCA) in conjunction with a core whose physics has been extensively studied (ORNL-3016, p. 12). Preliminary measurements with a neutron source and with the subcritical PCA suggest that the electronic noise is sufficiently low to permit meaningful reactor fluctuation spectra to be obtained at large ($>10\beta$) negative reactivity.

POWER REACTOR FUEL PROCESSING

Combustion of Irradiated Uranium Monocarbide with High-Temperature Steam. — The high-temperature-steam combustion process, developed as an alternative (semiaqueous) head-end process for spent uranium carbide reactor fuel, is being evaluated in the hot cell. Studies of the reaction of stoichiometric uranium monocarbide (irradiated to 7500 Mwd/metric ton) with steam (700 to 750°C, at atmospheric pressure) indicate almost no difference between the reactivities of irradiated and unirradiated fuel. The overall reaction was complete in an hour for nominally 1-g samples, as evidenced by the cessation of the evolution of gas.

About 5 moles of off-gas (89 to 95% hydrogen) was produced per mole of monocarbide. The analytical results for the nonradioactive off-gas agree closely with those previously obtained when unirradiated monocarbide was used. The free-flowing uranium dioxide powder produced by this process, and the steam condensate, are being held for future analysis for residual carbon and fission products. Previous results (ORNL-3561, p.7)

indicated that uranium can be recovered satisfactorily by solvent extraction from feed solutions derived from dissolution of the uranium dioxide in nitric acid.

Corrosion. – The inertness of graphite and carbon fuels to the usual aqueous chemical reagents has made it necessary to consider high-temperature decomposition (e.g., oxidation for graphite or carbide, hydrolysis for carbide fuels), followed by chlorination or fluorination of the ash with or without a prior leaching of the ash; such procedures for recovering the uranium or thorium from spent fuels result in corrosion, of course. Tests on type 309SCb stainless steel were conducted to evaluate this alloy as a construction material. Welded specimens gained weight (≤ 0.03 mg/cm^2) after seven cycles consisting in the use of oxygen at 800°C for 5 hr, followed by leaching in boiling 5 M HNO_3 for 1/2 hr.

For Nichrome V and Corronel 230, a different exposure was used: oxygen at 800°C for 5 hr, followed by an air-CCl_4 mixture at 600°C for 5 hr, and then a 1-hr leaching in 5 M HNO_3. The results confirmed that the rates for these alloys are unacceptably high (16 to 39 mils/month for Nichrome V and 19 to 23 mils/month for Corronel 230 for three contactings with the chemicals). Reversing the order of contacting with the first two reagents resulted in corrosion rates that were three to ten times higher. Reducing the temperature of the leaching step from that which produced refluxing of the nitric acid to room temperature generally increased low rates and sometimes decreased the high ones. It seems that a corrosion rate of about 20 mils/month is unavoidable.

To breach the stainless steel cladding on carbide or graphite fuels, oxygen that contains small amounts of fluorine is being tested as a high-temperature reagent for attacking the cladding. An "A"-nickel reactor is being used. As to the corrosion resistance of the reactor, welded specimens of "A" nickel were tested in steam at 825°C for 672 hr and in O_2-HF mixtures (O_2/HF ratio of 99/1) at 600°C for 96 hr. The specimens gained an average of 5 mg/cm^2 in the steam and a maximum of 0.1 mg/cm^2 in the O_2-HF. In no case was there any indication of localized corrosion.

Chloride Volatility. – Methods for producing and handling stable, concentrated solutions of Zr, Th, Al, and Nb in hydrochloric acid are being investigated. Such solutions would be obtained if the gaseous metal chlorides produced by chlorinating reactor fuels were recovered by absorbing them with water or water and steam.

Aluminum chloride–hydrochloric acid solutions behave similarly to $ZrOCl_2$-HCl or $ThOCl_2$-HCl solutions during evaporation. When excess HCl is distilled off, constant-boiling solutions are formed, and their properties depend on the concentration of the $AlCl_3$. The temperatures for incipient crystallization of the solutions increase with the concentration of the $AlCl_3$ until, at 4 M Al–11 M Cl, crystals form at just below the boiling point of the solution (about 110°C). The addition (at 100°C) of 50% of the stoichiometric amount of 17 M H_2O_2 needed to remove all chloride removed only 6% of the chloride. The stoichiometric proportions were based on the following reactions:

$$AlCl_3 + 3/2\ H_2O_2 \rightarrow Al(OH)_3 + 3/2\ Cl_2\ ,$$

$$2HCl + H_2O_2 \rightarrow 2H_2O + Cl_2\ .$$

However, the temperature for incipient crystallization was lowered from about 100°C to −20. The addition (at 100°C) of larger amounts of 17 M H_2O_2 (up to 140% of the stoichiometric amount for removing all the chloride) resulted in the same efficiency, 12% (also based on the reactions shown above).

Protactinium Adsorption. — Processes have been investigated for the separation of protactinium from Th-U^{233} fuel solutions in order to avoid the long delay required for all the Pa^{233} to decay completely to U^{233}. Laboratory studies showed that unfired Vycor, a leached borosilicate glass manufactured by the Corning Glass Works, will preferentially adsorb Pa^{233} from solutions obtained by dissolving thorium-base fuels in nitric acid. A least-squares evaluation of data obtained in batch adsorption tests with 60- to 80-mesh unfired Vycor and a test solution showed that the distribution coefficient (DC = counts min^{-1} g^{-1} for the glass divided by counts min^{-1} ml^{-1} for the solution) of the adsorbable Pa^{233} may be calculated as follows:

$$DC = (378 \pm 72) + (384 \pm 9)X - (1.27 \pm 0.13)XY - (1.13 \pm 1.02)Y ,$$

where X is the molarity of the nitric acid, and Y is the concentration of the thorium expressed as grams per liter. The test solution contained Pa^{233} (a million counts min^{-1} ml^{-1}), thorium (100 g/liter), and nitric acid in concentrations ranging from 0.5 to 12 M.

Fluoride Volatility Processing: Aluminum-Alloy Flowsheet Development. — In the salt-recycle type of process for Al-U alloy fuel, the proposed composition of the initial salt used for dissolution is 67-21-12 mole % $KF-ZrF_4-AlF_3$, with the final composition being approximately 52-16-32 mole % (neglecting the small amount of uranium fluoride dissolved in the melt). The advantage of this process would be that all the ZrF_4 could be obtained through the use of inexpensive $2KF \cdot ZrF_4$ (50¢ a pound), with the only other necessary additive being KF. No expensive ZrF_4 per se would be required.

In a laboratory study of the above process, hydrofluorination tests at 600°C with aluminum gave dissolution rates of 7.8, 23, 36, and 43 mils/hr, respectively, at AlF_3 concentrations of 12, 22, 26, and 29 mole %. Two tests at 12 and 20 mole % AlF_3 resulted in H_2 recoveries of 86 and 90% of theoretical. There was little indication of any delayed H_2 evolution as in tests at higher ZrF_4 concentrations, where ZrF_4 reduction and subsequent reoxidation occurred. A complete flowsheet test of the above process, involving both the hydrofluorination and fluorination steps, demonstrated that more than 99.9% of the uranium originally present could be volatilized from the melt in an hour.

Fluoride Volatility Processing: Single-Vessel Studies. — The three corrosion rods exposed during the single-vessel studies were sent to Battelle Memorial Institute for more extensive examination. Detailed results of this examination are in memorandum BMI-X-269 (December 27, 1963), but will be summarized here. The rods (one INOR-8 and two alloy 79-4) were exposed during ten dissolution-fluorination cycles and four additional fluorinations. Losses in thickness for both pairs of sides for

each rod were determined at every inch along the length of the rod. Metallographic sections were taken at 2, 15, and 22 in. from the top of each rod. These represent vapor, interface, and liquid specimens respectively. The exposure time to molten salt for all bars was 1060 hr. During this time, HF and F_2 were also present for 142 and 66 hr respectively. Maximum corrosion occurred in the interface region and was 22 mils for INOR-8 and 17 mils for alloy 79-4. Photomicrographs showed a distinct difference between the types of attack on the two alloys. The INOR-8 was roughened, producing an undulating surface with no pitting or intergranular attack, whereas alloy 79-4 was selectively attacked so that pits several mils in depth and several mils across were formed. In spite of this localized attack, alloy 79-4 specimens were more durable, since, even with the pits, the corrosion was slightly less than that for the INOR-8. The corrosion rate is apparently linear with time, according to micrometric measurements made near the middle of the experiment and at the end.

Fluoride Volatility Processing: Tellurium Removal from Process-Gas Effluent Stream. — The aqueous caustic system for fluorine disposal in the Volatility Pilot Plant is capable of removing some, but not all, of the Te^{127}-Te^{129} activity in the gas effluent before delivery to the stack system. However, more complete removal might be obtainable by passage through a hot nickel bed to reduce and absorb the undesirable tellurium activity, which presumably would enter the bed as TeF_6. Preliminary laboratory tests indicated that this is perhaps possible, but the chemistry and effectiveness of the process need further study. In the initial work, a 15 to 25% TeF_6 mixture (helium diluent) was passed through a nickel reactor (6 in. long and 1 in. in diameter) packed with nickel wool. The flow rate was about 100 ml/min. The tellurium retention in the bed was 63, 73, and 89% complete at 600, 500, and 400°C respectively. At 300°C, there was no retention.

Fluoride Volatility Pilot Plant. — The pilot-plant system is being flushed to obtain a uranium material balance and to decontaminate to the extent required for safe corrosion measurements. The first phase of this cleanout was completed. The entire system was rinsed to remove uranium; measurements indicated that no more than trace quantities of uranium remain.

Cell 1 equipment was flushed with ammonium oxalate solution. Activity at the former "hot spot" (2200 r/hr) was reduced to 5 r/hr. The highest measured activity in the cell (outside the equipment) is now 6 r/hr. The background at the bottom of the cell was reduced from 2500 to about 600 mr/hr.

THORIUM UTILIZATION PROGRAM

Uranium-233 Purification and Sol-Gel Oxide Preparation. — The solvent extraction facility was operated to purify about 5 kg of U^{233} for the Kilorod Facility. The product met all specifications for the sol-gel process. The U^{233} loss was 0.14%. A total of 43 kg of U^{233} has been purified to date: 30 kg for sol-gel operations and 13 kg for the Large Power Reactor U^{233} Physics Program at Westinghouse (Bettis).

Resumption of the thorium nitrate pyrolysis operations is still awaiting the receipt of the replacement steam superheater which burned out in November. Sufficient ThO_2 is available to complete the BNL-Kilorod program.

During December, 190 kg of mixed thoria-urania was prepared, bringing the total to 760 kg.

Pellet Fabrication. — It is deemed necessary to achieve a density of 9.2 g/cm^3 for the pellets to be used in the flux-measurement rods for the Brookhaven National Laboratory criticality tests on ThO_2-$U^{233}O_2$ sol-gel fuel. The dies are being made, and pellet pressing should commence during February. A furnace capable of higher temperatures is being obtained.

Rod Fabrication. — Operation of the remote rod fabrication equipment continued to be completely satisfactory. During December, 150 rods were completed, bringing the total to 720.

Maintenance was required on the welding machine, and advantage of the downtime was taken to replace the iris between the compactor and the fuel feeder. Data from the log book are being analyzed to determine the causes and extent of off-stream time.

Thorium-Uranium Fuel Cycle Development Facility. — Design of the TUFCDF, a facility to be used in the development of processes for remotely processing and fabricating Th-U^{233} fuel assemblies, has been in progress since January 3, 1963.

Title I design is complete, and the bound report was issued. Approval to start title II design has not yet been received. During December a concerted effort was made on a review of the title I design-cost estimate. Deletions, minor criteria changes, and removal of duplications led to sufficient cost reduction to provide for about 15% contingency at the start of title II design.

Design of the equipment for oxide preparation and fabrication is proceeding at a rate commensurate with the critical-path schedule. Where private industry is involved in supplying equipment, criteria discussions are being conducted. The expected completion date for this part of the facility is May 10, 1967.

Bids were received for the vertical tube furnace for gel calcination, and alternative furnace concepts are being studied. There is enough time in the schedule to allow for development and testing of an alternative concept.

Sol-Gel Microsphere Preparation. — Development of methods for preparing reactor fuel in the form of microspheres continued. The applicability of the sol-gel process to thorium fuels containing up to 8% uranium was demonstrated by preparing oxide microspheres, starting with a 4 M thorium-uranium oxide sol.

A two-fluid sol dispersion nozzle was incorporated into a system for forming thoria sol droplets of controlled diameter. This nozzle, in which 2-ethylhexanol flows from an annulus concentric with the sol stream, is superior to a single-fluid nozzle.

A larger column for preparing microspheres was installed and used to prepare several 100-g batches of thoria microspheres. Early fluidization problems were overcome by inserting a tapered metal rod in the glass column to prevent large radial variations in the velocity of the solvent.

Uranium dioxide microspheres 100 to 200 μ in diameter were formed by a modification of the sol-gel microsphere-forming process for thorium-uranium oxide. Satisfactory sol for this study was obtained by using the uranium(IV) hydroxide precipitation-peptization method.

Irradiation of Thorium Oxide Fuels. — No irradiations of bulk oxide fuels were started or terminated since the November report. Postirradiation examination of MTR-I and NRX-III rods is still in progress, and dissolution studies and burnup analyses were started.

The bowing observed in the thorium-uranium-zirconium alloy capsules is of grave concern to the operators of the General Electric Test Reactor, since it could give rise to local boiling. Tests are being conducted to determine the extent of this problem. Designs have been formulated to prevent bowing of future capsules.

Vibratory Compaction. — Modifications to the NAVCO vibrator made it possible to achieve higher densities than were previously obtainable with this equipment and to solve mounting and support problems. A multitube configuration was designed to explore the feasibility of increasing the production rate.

A Tektronix storage oscilloscope and a charge amplifier were obtained and calibrated. This oscilloscope can display a single acceleration pulse for about an hour for observation or photography. Characteristic acceleration pulses will be studied as a function of several variables in the vibrator design.

Thorium Alloy Development. — Billets of thorium containing dispersions of 4, 7, and 13 vol % ThO_2 were extruded. Specimens for hot hardness and creep tests are being machined. Results from some of the tests in progress are as follows:

Temperature (°C)	Hardness Values (DPHN)[a]	
	4 vol % ThO_2	7 vol % ThO_2
Ambient	129	148
300	54	67
600	31	31
800	13	13

[a]Diamond pyramid hardness number.

Preparation and Properties of U(IV) Sols and UO_2. — The preparation of U(IV) sols by electrolytic reduction-dialysis and by the precipitation and subsequent dispersion of the hydrous oxide continued. The objective is to prepare concentrated urania sols that can be formed into spheres by appropriate techniques or which can be concentrated to gels, dried, and fired to dense UO_2 suitable for vibratory compaction into fuel elements. With the electrolytic approach, nitrate-stabilized 0.8 to 1.2 M urania

sols were prepared directly in the electrolytic cell. These sols, however, did not show any tendency to gel on drying. With the precipitation-dispersion approach, 0.7 and 1.1 M urania sols were prepared and were formed into UO_2 gel spheres in a drying solvent that contained a surfactant. The gel spheres were converted to apparently dense UO_2 by air drying and then firing at 1100°C in a reducing atmosphere.

Sol-Gel Preparation and Properties of ThC_2 and UC_2-ThC_2. — High viscosity, thixotropy, and segregation continue to be the principal problems associated with thorium-uranium oxide—carbon sols and their forming into microspheres. A variety of carbon blacks and mixing procedures were studied, and an acceptable, though not optimum, method of forming sols has been developed.

The use of surfactants to prevent the coalescence of sol droplets in the forming column and to prevent the dispersion of carbon or thoria from the sols into the drying solvent (2-ethylhexanol) has been studied extensively; a working range for interfacial tension is 4 to 13 dynes/cm. At about 15 dynes/cm the droplets coalesce; below 4 they are not spherical. To prevent the loss of carbon or thoria, nonionic surfactants must be used in the 2-ethylhexanol. Cationic surfactants promote carbon dispersion, and anionic ones promote thoria dispersion.

Extension of Sol-Gel Process to Newer Fuel Types: Thorium Nitride. — Seven firings were made of ThO_2-C gels in nitrogen gas atmospheres to evaluate the possibility of producing a thorium nitride by the sol-gel process. Gels having C/Th mole ratios of 4 and 3 were used, and firing temperatures ranged from 1690 to 2100°C. Conversions in two preparations were encouraging, but more work is necessary to determine the practicability of this approach.

Thorium-Uranium Chemistry Studies. — Thorium oxide and uranium dioxide have the ability to replace each other in a cubic crystal in any proportion with no major change in the crystal structure. Uranium dioxide has the additional ability to incorporate excess oxygen within its crystal lattice up to a limiting value determined by its temperature. If this limiting value is exceeded, the cubic crystal is transformed into an orthorhombic crystal of triuranium octoxide (U_3O_8). The effect of incorporating thorium oxide into the uranium dioxide crystal on the limiting value of the excess oxygen content had not been studied in any detail. Since both uranium and thorium oxides are nuclear reactor materials of importance, a detailed knowledge of such relationships is of fundamental value.

We have determined the maximum excess oxygen content of the cubic phase containing various amounts of thoria (from 0 to 100% ThO_2) over a temperature range from 700 to 1600°C, when the samples are heated in air. The greatest value, $UO_{2.4}$, is obtained with no thoria present at 1525°C. As thoria is incorporated into the crystal, the amount of excess oxygen decreases and so does the temperature above which the crystal is stable. With 40% thoria and at 700°C, the oxygen per atom of metal has decreased from 2.4 to 2.267. A more complete understanding of the system will be achieved when current experiments involving atmospheres containing much less oxygen than air are completed.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — Assembly of the graphite core for the MSR was completed.

Cracks were found in welds in the core support ring after the reactor vessel was stress relieved. The ring and a small amount of metal from the vessel wall were removed. The wall was repaired, and brackets are being welded in place of the ring to support the core assembly.

Water was circulated through the shell side of the heat exchanger, and we found that the tubes vibrated excessively at flow rates above about 600 gpm, which is half the design flow. The tube bundle was removed from the shell, and preparations are being made to lace the tube bundle to reduce the movement of the tubes.

Satisfactory progress was made in all areas of MSRE installation work. The H. K. Ferguson Company had completed ~70% of their work, and the overall project was ~90% complete on January 1.

Metallurgy. — We are investigating the effects of irradiation on the properties of INOR-8, the MSRE structural material, by comparing the tensile properties of irradiated specimens and unirradiated controls at temperatures from 25 to 900°C.

Test specimens of INOR-8 were annealed and were irradiated in the ORR to neutron doses from 5×10^{20} to 12×10^{20} neutrons/cm^2 at 704°C. Control specimens were given identical thermal treatments out-of-pile. The data obtained to date indicate that the effects of radiation on INOR are similar to those found with other nickel-base alloys and stainless steels. The ultimate strength decreased, the ductility at high temperature decreased, but the yield strength did not change. The uniform elongation at a test temperature of 700°C was approximately 5%; however, this property was strain-rate dependent.

Postirradiation annealing treatments at 700 and 900°C had no effect on the irradiation-induced loss in ductility.

Chemical Development. — The MSRE fuel contains a liberal addition of ZrF_4 to ensure that any crystals formed by contamination with oxide ions will be ZrO_2 and not UO_2. A more exact measure of how much margin is subtended by the currently specified 5 moles of ZrF_4 to 0.9 mole of UF_4 came from continuing studies of the reaction

$$ZrO_2(\text{cryst}) + U^{4+}(\text{soln}) \rightleftharpoons Zr^{4+}(\text{soln}) + UO_2(\text{cryst}) \ .$$

The equilibrium quotient for this reaction, the ratio $[Zr^{4+}]/[U^{4+}]$, is the limiting ratio for no precipitation of UO_2; it was found to vary somewhat with the concentration of Zr^{4+}. At 5 mole %, corresponding to the fuel composition, the limiting ratio was ~1.5, with a small temperature dependence in the range 500 to 700°C. At lower concentrations of Zr^{4+}, corresponding to dilute mixtures of fuel salt in flush salt, it rose somewhat (to ~3), but ample margin still remained. Thus, although the deviations from Henry's law implied in these results have not been fully explored, UO_2 is not expected to form as a stable solid phase under reactor operating conditions.

The observed behavior of melts saturated with both UO_2 and ZrO_2 continued to indicate that the equilibrium solid phases are the separate pure oxides (cubic UO_2 and monoclinic ZrO_2). Though some published phase diagrams of the UO_2-ZrO_2 system suggest that $(U\text{-}Zr)O_2$ solid solutions should be the stable solid phases in contact with melts at 500 to 700°C, our measurements, which included the equilibration of specially prepared solid solutions with melts, have indicated that they are not stable with respect to the pure oxides.

Preparation of MSRE Materials. — All fluoride starting materials that will be required for the preparation of coolant, flush, and fuel mixtures for the MSRE have been received at the fluoride production facility except for UF_4, which is readily available when needed. The initial shipment of beryllium fluoride received (5300 of 12,000 lb ordered) was tested for compliance with purchase specifications and found acceptable. The remainder of this order has been received, and samples have been submitted for chemical and spectrographic analyses.

Installation of equipment for pretreating Li^7F to obtain a more dense anhydrous feed material for the fluoride production plant was completed. A final dry-run check on the equipment and operational variables is being made, with the production operation to follow. The Li^7F will be exposed to a flowing stream of anhydrous hydrogen fluoride mixed with helium, while being heated to 200°C, to remove moisture and to convert hydroxides to fluorides. The material will then be purged with helium alone while being heated to above 500°C to effect densification.

Facilities have been installed for the use of all-plastic suits with individual fresh-air supplies in the dry loading room of the production facility. Molecular-sieve drying columns have been provided for the hydrogen and helium supply lines. Sodium fluoride pellet traps have been placed in the off-gas stream to remove HF from the off-gas. An improved exhaust system has been constructed for removing hydrogen from the off-gas and venting it to the atmosphere.

Radiation Effects. — The chemical compatibility of MSRE components under irradiation, as deduced from continuing examinations of specimens from in-pile irradiations, appears generally reassuring. Some undesirable effects (e.g., CF_4 formation) due to radiolytic F_2 from the frozen fuel at room temperature were reencountered and appeared even more definitely to be characteristic of conditions not involved in reactor operation. A possible exception, primarily because the mechanism is not yet understood, is a deposition of uranium in graphite. This happened without penetration of the graphite by fuel salt and probably occurred as a side effect of F_2 evolution during reactor shutdowns, but because of its potential importance this topic has received first priority in recent and current postirradiation studies.

The amount of uranium deposited in experiment 47-4 (10% burnup) was about 0.5 or 1 mg per square centimeter of graphite, as found by activation analysis and supported by chemical analyses. There was sufficient scatter in the data to render uncertain an anticipated correlation with in-pile exposure, and correspondingly with F_2 evolution. The location of the uranium was revealed by autoradiographs before and after in-pile activation of cross sections of graphite specimens that had decayed for

17 months; the uranium was confined to the outer rim of the graphite, where there was also a large concentration of fission product activity from the compatibility test proper. Much better resolution of the site of the deposits was obtained with x radiography, which, through recent developments, was accomplished with negligible interference from fission product activity, and which showed a relatively uniform though fringed penetration of uranium to a depth rarely exceeding 0.05 mm.

Irradiation Tests. — Design of the beam-hole assembly for the ORNL-MTR-47-6 irradiation experiment is complete, and all parts should be delivered by February 20, 1964. Final design of the system for obtaining gas samples from the capsules and of the instrumentation is in progress, and most of the long-delivery items have been ordered.

MSR Processing. — Design of the MSRE fuel processing system was completed except for the instrumentation. Procurement of the fuel-storage-tank heaters and material for the equipment was started. Fabrication of the INOR-8 fuel storage tank is 50% complete.

Most of the funds assigned to the processing plant in FY 1964 will be used for the reactor, so the completion date was changed from July 1 to November 1, 1964. With the revised schedule, the system will not be completed until after precritical testing of the reactor system with flush salt. This flush salt will then be used for shakedown of the fuel processing system.

Analysis of Radioactive MSRE Fuels. — The testing of all the equipment necessary to prepare MSRE fuel samples for analysis is continuing. Simulated fuel samples in copper ladles are received in the transport container. They are handled and treated remotely until the solidified fuel salt is in a powdered and homogenized form ready for analysis. All the equipment, including an improved transport-container decoupling device, appears to be functioning satisfactorily; however, some corrections are indicated. Conditions necessary to obtain maximum recovery are still being established.

Methods are now being developed for the dissolving of parts of the above prepared samples in preparation for analysis.

Since the determination of uranium in the MSRE fuel salt must be as precise as possible, the recently developed high-sensitivity coulometric titrator, ORNL-MQ-2564, was evaluated for such application. An aliquot of sulfate solution of the MSRE salt which contained about 600 μg of uranium was titrated in the mockup hot cell under simulated hot-cell conditions. The relative standard deviation for the titration was calculated to be 0.8%. The instrument gives every indication of being entirely satisfactory for MSRE application.

HIGH FLUX ISOTOPE REACTOR DESIGN AND DEVELOPMENT

Metallurgy. — A decision was made, and AEC approval obtained, to purchase the first batch of commercial HFIR fuel elements on a cost-plus-fixed-fee contract rather than on a fixed-price basis. This decision was dictated by the impossibility of making reasonable estimates of the various rejection rates, thereby requiring contingency factors larger than could be tolerated.

The requested date for receiving comments on the proposed fuel-element specification has passed, with comments being received from a number of companies. The number of comments or questions received was surprisingly small. These are now being reviewed, and a modified specification will be issued.

In consideration of the small number of comments on the specification, the request for the cost-plus-fixed-fee fuel-element bids has been issued and is to be based upon the present specifications and drawings. The request for a bid was sent to fuel-element fabricators who have shown proficiency in this area.

Physics. — HFIR critical-experiment data (HFIRCE-2) and further calculations on the 9.4-kg core were used to help compile a complete reactivity accountability for the HFIR. Included in the compilation were reactivity variations associated with fuel and burnable poison loading and distribution tolerances, poison (Li^6, He^3) content of the beryllium reflector, variations in the plutonium-target neutron cross sections with time, temperature variations, fission product poisons, and variations in control-rod worth as a function of rod position and rod burnup. The minimum and maximum values of $k_{eff} - 1$ with all rods removed were 0.074 and 0.129. These correspond to a calculated fuel-cycle range of 8 to 20 days.

Integrated rod worths were determined in the HFIRCE-2 critical experiments to be 0.165 with all five rods in and 0.133 with the inner rod out and the four safeties in. (The rods used in the experiments simulated Eu_2O_3-Ta-Al rods that had been used for about 6 months with the reactor at full power.) The corresponding minimum shutdown margin with the central rod removed was $0.004\Delta k$. With all five rods in, the shutdown margin for the maximum core reactivity case was 0.036.

The plutonium target design was modified to reduce the amount of high-velocity (42 fps) coolant flow area. This reduced the amount of positive reactivity (induced by voids entering the island with the coolant) that could be introduced with an effective ramp of about 30 msec. The new corresponding maximum reactivity is 0.013 instead of 0.015. Voids entering with the low-velocity (3.5 fps) target coolant have a comparatively long effective ramp of about 360 msec.

Plans for the third HFIR critical experiment (HFIRCE-3) were essentially completed, and the date for commencement now appears to be about April 1, 1964. The HFIRCE-3 core will contain 9.4 kg of U^{235} and 2.12 g of B^{10} burnable poison. Two sets of control rods will be used: (1) Ag-Cu-Al rods used in HFIRCE-2 (simulate approximately 6-month Eu_2O_3-Ta-Al rods) and (2) Eu_2O_3-Ta-Al rods. The latter are those proposed for the HFIR.

Integrated fission density information and fuel plate temperature histories were obtained from fuel cycle and heat removal calculations for use in evaluating possible radiation damage in the fuel plates. Values obtained so far are for nominal core conditions only; hot-plate, hot-streak, and hot-spot effects are being analyzed. For nominal core conditions the maximum integrated fission density consistent with 14 days of full power operation occurs next to the control region at the horizontal midplane; the value is about 1×10^{21} fissions/cm^3 of fuel-plate fuel core. The corresponding fuel plate temperatures are about 250°F. At the radial center and outlet end of each fuel element the fission density is 4×10^{20} fissions/cm^3, and the temperature is about 300°F.

Component Development. — In the present design of the HFIR control-rod track assembly, cobalt-base preload bearings will be in contact with the 6061-0 aluminum alloy cladding of the control-rod plates. Both static and dynamic corrosion tests indicate that the aluminum alloy undergoes severe galvanic corrosion in contact with Stellite 19, Stellite 3, and Haynes alloy No. 25, which are available outer-bearing race materials. In addition to the pitting of 0.008 to 0.015 in./week, the highly concentrated load deforms the aluminum cladding, which results in additional thinning.

Preliminary results from stainless steel sleeves shrunk on the outer bearing race show a lower rate of attack on the aluminum. Other metallic and nonmetallic materials are being tested in an attempt to resolve this problem.

Fabrication is proceeding on all HFIR components currently certified for construction. Fabrication is 75% complete on the permanent reflector and those portions of the horizontal beam tubes which are outside the reactor vessel. Fabrication of the structural components of the reactor is estimated to be 30% complete. A fabrication development program has been initiated to investigate problems associated with welding and machining of the HFIR control-rod tracks. It is anticipated that this program will demonstrate the feasibility of fabricating hollow (1/4-in. wall) box-shaped track section weldments, using spot welds on 1-in. centers. A stress-free weldment is expected to minimize the difficulties which may be encountered during final machining to produce 38-in.-long surfaces that are flat, parallel, and straight within 0.002 in. total indicated runout.

Preliminary testing of the vertical engineering facility tubes with respect to vibration has been completed in the HFIR mockup. Mocked-up tubes of 1-in. sched-40 and 2-in. sched-80 aluminum 6061-T6 pipe were tested. The tubes were located directly in front of and 90° from the south water inlet. Tests were conducted at both locations with and without strainers over the vessel water inlet openings. Vibration was excessive in all cases, with the tubes moving as much as 1/16 in. at the top of the mocked-up reflector liner. Strain gages mounted at the midpoint of the tubes indicated the frequency to be approximately 20 cps.

Design and Analysis. — A review of the fuel-element hot-spot analysis is being conducted. Separate blister and fuel segregation hot-spot factors for both sides of the fuel plate are being considered. These factors were combined in previous calculations.

Drawings of the removable reflector have been certified for construction. The only major change resulting from the review of these drawings was the use of a solid rather than a split aluminum liner between the inner beryllium cylinder and the control region.

The upper plenum and the outer shroud drawings of the core structure were certified for construction.

Drawings of the control-rod-drive support columns, support platform, position indicators, pressure balance cylinder, and other components were issued for approval.

Fabrication of the pressure vessel was completed by Allis-Chalmers Manufacturing Company in December, and the vessel was delivered to the job site in Oak Ridge.

Reports. — The safety analysis report has been temporarily delayed until an analysis of possible startup accidents has been completed. Following this determination, the discharge of radioactive material can be evaluated.

Ten of the twelve chapters of Volume I of the Reactor Description are in final typing. Volume II, containing some of the actual working drawings, will be ready for final typing when the Singmaster and Breyer drawings arrive from New York. The Reactor Description is scheduled for distribution in March 1964.

PHYSICAL RESEARCH

Reactor Operations. — The ORR operated 78.1% and the LITR 86.4% of the time in 1963.

We installed new viewing ports in the top cover of the ORR tank through which each fuel element can be inspected while the reactor is running. The ports have enabled us to detect pieces of a neoprene gasket that was deposited on several fuel elements and to remove the gasket before it caused serious trouble.

A new set of five hydraulic tubes has been installed in position D-8 in the ORR core. The tubes in position F-8 were removed because they were exerting a lateral force in the core which caused the fuel elements to be tight.

Waste Disposal. — A considerable reduction in routine releases of radioactive materials into the environment occurred during 1963. The average monthly release of Ru^{106}, one of the two main contaminants released into the White Oak Creek drainage basin, was 77 curies, as compared with an average of 447 curies during 1962. There was a small increase in the average strontium release (approximately 90% Sr^{90}), from 0.7 curie/month in 1962 to 0.8 curie/month in 1963.

The average discharge from the four stacks was 4.5 curies/month in 1963 and 10.1 curies/month in 1962. The bulk of the activity in both years was I^{131}. The filterable activity released from the same stacks was 35 mc/month in 1963 and 149 mc/month in 1962, with the active materials being predominantly I^{131} and Cs^{137}.

Hot-Cell Operations. — Operations in the hot cells were routine. Six fueled capsules from the ORR were examined; fuels included graphite balls containing UC_2, pyrolytic-carbon-coated UC_2 particles, and UO_2. One UO_2 meltdown and one experimental Army PM (portable medium) fuel bundle were examined. Two ORR poolside experiments containing tube-burst specimens were disassembled. The gas lines to two of the capsules in MSRE experiment MTR-47-5 were slit and examined for evidence of plugging or corrosion. About 120 irradiated tensile specimens were pulled. Profile measurements were made on several beryllium shim rods and core pieces which had bowed.

Installation and final check of equipment continued at the High-Radiation-Level Examination Laboratory.

PHYSICS AND MATHEMATICS

Charge Spectrometry. — The relative abundances of neon and argon ions formed as the result of inner-shell vacancies have been measured with a newly designed charge spectrometer. This spectrometer, which

uses magnetic analysis, will be compared with an earlier time-of-flight machine. To produce the desired initial vacancies the gases are bombarded with x rays of suitable energies. By measuring the spectra of neon ions as a function of x-ray energy, we have determined that the extent of ionization is independent of the energy of the photoelectron from 0.7 to about 20 kev. The charge spectra obtained from producing vacancies in the K and L shells of argon also have been measured. The results where initial ionization is principally in the K shell are in close agreement with those for the electron capture of Ar^{37}. The gross spectra of argon ions have been successfully related to transition probabilities calculated by Rubenstein. In general, however, more ionization is present in both the neon and argon studies than can be accounted for by the present theories of Auger processes and electron shake-off.

The charge states of the krypton atom following the electronic readjustment to an inner shell vacancy have been measured with a magnetic mass spectrometer. Vacancies were produced either predominantly in the K shell, by radiation from a molybdenum target, or predominantly in the L shell, by radiation from a titanium target. The relative intensities of the ions of charge n = 1, 2, 3, ... are as follows (in percent) for an initial K hole: 1, 2.5, 6.3, 16.1, 19.3, 17.2, 15.4, 11.7, 7.5, 2.3, 0.9; for an initial L hole: 1.5, 3.1, 8.5, 22.0, 23.8, 23.1, 13.5, 3.9, 0.52, 0.11. The results have been interpreted in terms of Auger cascades and electron shake-off. Also noted is the similarity of the K-hole spectrum with the charge spectrum obtained [Phys. Rev. 116, 1548 (1959)] from Kr^{79}, which undergoes electron capture.

Neutron Diffraction. – Neutron diffraction measurements were made on a series of rare-earth intermetallic compounds to determine type of magnetic order and its relationship to the number of valence electrons in the system. The compounds studied included TbCu, TbAg, ErCu, TbZn, and TbHg, all of which have the CsCl type of crystal structure. The first three compounds exhibit antiferromagnetism in which adjacent ferromagnetic (110) planes of moments are oppositely oriented (C-type structure). The Néel temperatures are 115, 100, and 33°K respectively. The last two compounds are ferromagnetic, with Curie temperatures of 160 and 80°K. The lattice constant of ferromagnetic TbZn lies between those of TbCu and TbAg, both of which are antiferromagnetic. This shows that the difference in magnetic behavior is associated with the number of valence electrons rather than with interatomic separations. The expected tripositive moment value of $9\mu_B$ per terbium atom was found for all the terbium compounds. The erbium moment in ErCu was observed to decrease with decreasing temperature from approximately $9\mu_B$ at 298°K to about $6\mu_B$ at 77°K. This moment reduction is probably associated with strong ligand field effects in this compound.

Theoretical Physics. – The (d,He^3) reaction on Ca^{40} and isotopically enriched targets of Ti^{46}, Ti^{47}, Ti^{48}, Ti^{49}, and Ti^{50} has been investigated. Absolute differential cross sections for a number of transitions were obtained over an angular range from 11 to 33°. The angular distributions are compared with distorted-wave calculations which use the optical-model-potential parameters derived from the elastic scattering of 21-Mev deuterons on the same target materials. The data for the elastic scattering are given together with the optical-model fits. The $Ca^{40}(d,He^3)K^{39}$ reaction was used to obtain the normalization coefficients

necessary to extract spectroscopic factors from the comparison of the measured and calculated differential cross sections. The results show that the excitation energies of the $d_{3/2}$ and $s_{1/2}$ proton hole states are surprisingly low and increase with the number of $f_{7/2}$ neutrons in the nucleus. In the case of Sc^{45}, the $d_{3/2}$ hole state which contains the full $d_{3/2}$ strength is found at an excitation energy of less than 50 kev. The excitation energy of the $d_{3/2}$ hole state increases to 800 kev in Sc^{47} and to about 2.4 Mev in Sc^{49}. The excitation energy of the $s_{1/2}$ hole state increases from 0.92 Mev in Sc^{45} to 2.1 Mev in Sc^{48}.

Low-Energy Neutron Time-of-Flight Spectroscopy. — Gamma rays from resonance neutron capture in In, Tb, and Hf have been studied with the ORNL fast chopper and a 9-in.-diam × 12-in. NaI(Tl) scintillation detector at the 25-m flight station. Variations of the relative intensities of the low-energy gamma rays following neutron capture in different resonances in In^{115} and Hf^{177} are known to occur, and the isomeric cross-section ratio for activation by neutron capture in In^{115} also exhibits variations between resonances. This would indicate that variations of the intensities of the high-energy capture gamma rays from resonance to resonance could be expected for these nuclides. S-wave neutron capture in Tb^{159} leads to capturing-state spins of 1^+ and 2^+, permitting a fast E1 transition to the 3^- ground state from the 2^+ state. For an indium target, gamma-ray spectra as a function of neutron energy exhibited prominent gamma transitions at 4.6, 5.6, and 6.3 Mev; but due to the finite gamma-ray energy resolution and the intense continuum, no definite conclusions could be drawn with respect to the change of the relative intensities of these three gamma rays between the 1.457-, 3.86-, and 9.10-ev neutron resonances. The high-energy gamma-ray spectra for neutron capture in Tb^{159} from the 3.35-ev through the 44.2-ev neutron resonance showed no resolved single gamma-ray transition, so that no conclusions may be drawn regarding the spins of the investigated levels. A 4.2-Mev transition following neutron capture in the 7.8-ev resonance of Hf^{178} and a 3.8-Mev transition following neutron capture in the 74-ev resonance of Hf^{180} were resolved. Coincidence measurements between these transitions and the low-energy capture spectrum did not show resolved gamma-ray lines.

Low-Temperature, Nuclear and Solid State Physics. — New measurements of the conversion electrons associated with the 312 ± 2 kev transition from the 40-msec second excited state of In^{114} give a K conversion coefficient of 0.10 ± 0.02 and a K/(L + M) ratio of 3.05 ± 0.50, leading unambiguously to an E3 assignment. Similarly, measurement of conversion electrons from the 2.5-sec second excited state of In^{116} gives a K/(L + M) ratio of 1.6 ± 0.2 for the 165-kev transition; this also permits an E3 assignment for the transition.

High-Voltage Experimental Program: Strength Function and Level Spacing of the Even Zirconium Isotopes. — Recent measurements (ORNL-3425, pp. 32–35) on the even isotopes of zirconium yielded an average s-wave strength function of $(0.7 \pm 0.2) \times 10^{-4}$. This value is substantially larger than that for other nuclei in this mass region. By use of the ORNL pulsed accelerator, the measurements on zirconium have been repeated, principally in order to extend the range of measurements to 45 kev. In the region of energy overlap, there is good agreement on the location of

the principal resonances. The pulsed-accelerator measurements exhibited structure in the region between resonances which, without better resolution, can only be construed as fluctuations in an average cross section. These fluctuations complicate the interpretation of the transmission data. However, treating the average as a potential scattering, a reasonably consistent set of widths is obtained. The average strength function obtained for all four even isotopes is $(1.5 \pm 0.5) \times 10^{-4}$, which is much larger than the previous chopper measurements. The presence of the fluctuations between resonances enables us to deduce a value of the s-wave potential scattering cross section which is about 20% smaller than that based on the average cross section between resonances. Finally, a strong decrease in level spacing is apparent as neutrons are added in excess of 50.

High-Voltage Experimental Program: Differential Elastic Scattering of Neutrons from O^{16}. – Differential elastic scattering of neutrons from zero-spin nuclei, such as O^{16}, allows one to assign quantum parameters for neutron resonances. Apparatus designed for determining the neutron differential cross sections with good energy resolution in the 1- to 2-Mev range [Bull. Am. Phys. Soc. 7, 576 (1962)] has been used to measure angular distributions of neutrons scattered from O^{16} with ~20-kev energy resolution. Data were obtained around the broad 600-kev-width peak at 3.6 Mev, and at the peaks of the 25-kev-width resonance at 3.77 Mev and the 80-kev-width resonance at 4.20 Mev, as well as at several nonresonant energies. At 3.35 Mev the present results are in agreement with those reported in the literature [BNL-400, 2d ed. (1962)]. At 3.77 Mev a least-squares Legendre polynomial fit to the data, corrected for multiple scattering and converted to the center-of-mass system, gives $\sigma(x) = 0.37P_0 + 0.20P_1 + 0.54P_2 + 0.44P_3 + 0.30P_4 + 0.11P_5$, where the P's are functions of x, the cosines of the center-of-mass angles. The coefficients are good to within about 10%.

High-Voltage Experimental Program: The $Be^9(He^3,p)B^{11}$ Reaction. – The differential cross sections for proton groups from the second through the ninth excited states of B^{11} have been measured for He^3 energies between 1 and 3 Mev. As the bombarding energy is increased the angular distributions, in general, show more forward peaking, as has been observed by other workers at higher bombarding energies [Proc. Phys. Soc. (London) 75, 754 (1960); Phys. Rev. 116, 1585 (1959)]. For some of the groups a significant change in the shape of the angular distributions is seen within the measured energy interval. Excitation functions have been measured at 25 and 90° for the observed groups. The yield curves are seen to rise rapidly and smoothly without pronounced resonance structure. The above measurements indicate that this reaction is complex in nature and therefore not easily described in terms of any single type of reaction mechanism.

High-Voltage Experimental Program: Nanosecond Pulsing for Van de Graaff Accelerators. – A pulsed ion source capable of delivering 10-ma proton bursts with durations of approximately 1 nanosec has been built. The system consists of a duoplasmatron ion source, a lens-chopper combination, and a klystron buncher which uses the entire accelerator as a

drift path to produce bunched ion bursts at the target. The equipment has been fitted into the terminal of the ORNL 3-Mv Van de Graaff accelerator. Since no accessory bunching magnets are used, ion bursts can be delivered to many different target stations. An external diverter system is used to vary the repetition rate from 10^6 sec^{-1} to 3×10^4 sec^{-1}.

CHEMISTRY

Effects of Radiation on Analytical Methods. — The effect of gamma radiation on the accuracy of the polarographic, indirect method of determining U(IV) in solutions of U(VI) was investigated. Under the influence of gamma radiation, U(IV) is oxidized to U(VI) in aqueous solution. In solutions containing ~5 mg of UO_2 per ml of 85% phosphoric acid, the conversion of U(IV) to U(VI) at 25°C was ~6% for a dose of 2.4×10^4 rads. Experiments are being carried out to determine the effect of temperature on this reaction.

A study of the effects of gamma radiation on 2-thenoyltrifluoroacetone (TTA) in xylene was initiated. TTA is used in many solvent extraction procedures in analytical chemistry. It has been found that a dose of 3×10^5 rads has no apparent effect on the ultraviolet and infrared spectra of TTA.

The concentrated lithium chloride medium used in actinide separations, which has been investigated for radiolytic formation of free chlorine on irradiation, is proving to be very interesting for the study of many types of reactions in analytical chemistry. There seems to be very little free water in this medium, so that reactions analogous to those in nonaqueous organic solvents are possible. Most metal ions lose their water of hydration in a concentrated lithium chloride solution and remain as chloro complexes. An acid in this system may be completely titrated before the base begins to attack the metal ion, thus making it possible to titrate free acid in a solution containing a hydrolyzable metal ion. It is also possible to titrate metal ions to various end points corresponding to different hydration states. The titration of uranyl ion, for example, shows end-point breaks corresponding to the $UO_2(OH)_2$ and diuranate end points.

Chemical Engineering Research. — A fully instrumented cascade with seven Mark-X-type extraction stages (nominally 3-3/4 in. long) and four polisher stages was built. Line volume was reduced to a minimum for a device with external pumps. The volume per stage is about 160 cc, with a residence time of less than 4 sec per theoretical stage. Stage efficiency at a volume ratio (aqueous/organic) of 3 is about 75%. Comparing this with data for a cascade that has four stages indicates that stage efficiency is not a function of the number of stages.

Separations Chemistry Research. — The use of diethylbenzene (DEB) as a diluent in solvent extraction processing prompted a study of DEB stability toward nitric acid. It was shown previously (ORNL-3388, p. 23) that, under comparable conditions of degradation by 2 M HNO_3, more Zr-Nb fission-product-extracting materials were formed from ortho- and para-DEB

than from meta-DEB. It has now been shown as well that the total amount of degradation products (extracting and nonextracting) from p-DEB is larger than from m-DEB. One of the major products, isolated from degraded p-DEB by distillation and gas-liquid-chromatographic techniques, is 4-ethylacetophenone. However, the extraction properties of closely related acetophenone, used as a stand-in, do not account for the Zr-Nb extraction of degraded DEB. Nitrogen-rich fractions of degradation products from DEB have been accumulated, but the individual components have not yet been identified.

Volatility Studies. — Sodium fluoride is unique among the alkali-metal fluorides (and a number of other likely fluorides tested) in its reversible absorption of uranium hexafluoride; over 99% of the absorbed uranium hexafluoride can be desorbed in the absence of fluorine. While the fluorides of Li, K, Rb, Cs, and Ag absorbed large quantities of uranium hexafluoride, less than 10%, if any, of the absorbed uranium hexafluoride could be desorbed below 400°C. At about 400°C, the yellow complexes of either KF, RbF, or CsF with UF_6 began to turn white, and analyses indicated that the uranium was no longer all hexavalent. As to LiF, the yellow hexavalent complex was not detected; reaction occurred above 370°C, with the product being white. Analysis of the white complex, which had been heated to as high as 390°C, showed almost all the uranium to be pentavalent.

Radiolysis of Metal Fluorides. — X-ray dosages of about 10^{16} Mev/g to the crystalline solids at ambient temperature have been shown, in previous experiments of this study, to liberate F_2 from the (LiF, BeF_2, ZrF_4, UF_4) MSRE fuel mixture but not from pure LiF or ZrF_4. Recent irradiations have liberated F_2 from ThF_4. A well-crystallized ThF_4 specimen (crystallite size, 2400 A; surface area, 0.42 m^2/g; 85% of particles smaller than 2 mils in diameter) was fluorinated for 14 days at 160°C; on irradiation to a dose of 4×10^{15} Mev/g this preparation yielded traces of CF_4 and F_2 in an amount corresponding to a G value of about 0.005.

Tests of the effect of particle size on behavior of the MSRE salt under x rays have been continued. Fine and coarse fractions (0.52 and 0.08 m^2/g respectively) of a simulated fuel batch (crystallite size, 330 to 370 A) were each alternately fluorinated and irradiated. The fine particles ultimately yielded elemental fluorine with G values approaching 0.02; the coarse particles yielded measurable quantities of CF_4 and COF_2 (equivalent to a G value of 0.002 for F_2 evolution) during irradiation to a dosage of 7×10^{15} Mev/g. No evidence for UF_6 was found in any of these tests. It appears that surface area (particle size) and the presence of impurities such as O^{2-} and H_2O (which can be removed by prefluorination) are parameters of importance.

Mechanism of Aqueous and Radiation Corrosion of Zirconium Alloys. — The causes of enhanced corrosion of Zircaloy-2 by aqueous media under heavy-particle irradiation are being investigated. Film-free, pickled, and prefilmed specimens were irradiated by fast neutrons in the LITR for 6 to 10 days while in helium and at 100 or 300°C. The film-free surfaces were prepared by heating pickled specimens in helium-filled Zircaloy-2 autoclaves at 500°C for 16 hr, and were irradiated while still in the autoclaves. The specimens were then oxidized in 300°C steam plus oxygen along with controls, and measurements were periodically made of the specimen weight gain and ac impedance of the corrosion film.

The irradiated, film-free specimen corroded twice as fast as its control for a period of about 5 weeks. After a period of a few days of reduced corrosion, an irradiated prefilmed specimen and irradiated pickled specimens corroded more rapidly than their respective controls. Some irradiation effects on film impedances were observed, but these were not correlated with enhanced postirradiation corrosion.

The results are regarded as evidence that the observed enhancement of corrosion resulted, primarily, from an increase in chemical reactivity of the metal. Experiments to confirm the results are in progress.

Physical Chemistry of Ion Exchangers. — Spectrophotometric examinations and chemical analyses were conducted on a cross-linked polystyrene sulfonate (Dowex 50, 0.5% divinylbenzene) and on a liquid cation exchanger prepared by dissolving dinonylnaphthalene sulfonic acid in toluene, in an attempt to find an explanation for the anomalously large absorption of Fe(III) from concentrated aqueous HBr and LiBr solutions by strong-acid cation exchangers. The characteristic ligand field bands of the tetrahedral anion, $FeBr_4^-$, were observed in both exchangers when they had been brought to equilibrium with solutions more concentrated than 5 M in Br^-. Measurements of the Li^+ (or H^+) content of the liquid exchanger showed that the amount in excess of the exchange capacity was sufficient to give $LiFeBr_4$ (or $HFeBr_4$). It was concluded that the absorption of Fe(III) from concentrated HBr and LiBr involved the $FeBr_4^-$ complex ion, and that the ion pairs, $H^+FeBr_4^-$ and $Li^+FeBr_4^-$, were "salted" into the organic cation exchange phase. Thus, in special circumstances, ion exchangers may extract salts by essentially a solvent extraction mechanism involving ion association complexes.

Nuclear Chemistry: Fission Studies and Characterization of Low-Energy Properties of Nuclei. — A three-crystal scintillation pair spectrometer was constructed for the study of high-energy gamma rays from radioactive nuclei. A gamma-ray spectrum of 37-min Cl^{38} showed the well-known gamma transitions in Ar^{38}, measured here as 1655 ± 5 and 2176 ± 5 kev. In addition, the 3831-kev gamma-ray transition from the 3^- state of Ar^{38} to the 0^+ ground state was seen for the first time. If the intensity of the 1655-kev gamma ray is taken as 1.0, the relative intensities of the 2176- and 3831-kev gamma rays are 1.37 ± 0.04 and $(6.2 \pm 0.5) \times 10^{-4}$ respectively. The transition probability for the 3831-kev gamma ray is greater than the single-particle estimate by a factor of about 2000.

Physical Chemistry of Molten Salts: $HF-H_2O$ Equilibrium with Molten Fluorides. – A study of the equilibrium between $HF-H_2O$ gaseous mixtures and $LiF-BeF_2$ melts has been initiated, as a means of investigating thermodynamic behavior in molten fluorides. The reaction involved,

$$2HF(g) + O^{2-}(soln) \rightleftharpoons 2F^{-}(soln) + H_2O(g) ,$$

has long been employed as a means of removing small amounts of oxide, which may be present as a contaminant. When a melt of LiF and BeF_2 is saturated with an oxide (e.g., BeO), the corresponding equilibrium,

$$2HF(g) + BeO(cryst) \rightleftharpoons BeF_2(soln) + H_2O(g) ,$$

is a function only of P_{H_2O}/P^2_{HF}, a measurable quantity, and the activity of the BeF_2 in the solution. Measurements of such melts, coupled with literature data on the corresponding reaction involving pure liquid BeF_2, indicate that the activity coefficient for BeF_2, γ_{BeF_2}, has a value of ~0.2 for Li_2BeF_4 melts in the temperature range 500 to 700°C, where BeF_2 concentration is expressed as mole fraction and liquid BeF_2 is the standard state. Similar measurements for Li_2BeF_4 melts saturated with ZrO_2, a less soluble oxide, along with thermochemical data for the equilibrium of ZrO_2 and ZrF_4 solids with HF and H_2O, indicate that γ_{ZrF_4} has a value of ~0.02 at low concentrations of ZrF_4. The observed P_{H_2O}/P^2_{HF} quotients for melts saturated with ZrO_2, combined with a previous estimate of the solubility product of ZrO_2 (ORNL-3436, p. 15), give the following relationship between P_{H_2O}/P^2_{HF} (in atmospheres) and the oxide ion concentration (in moles per kilogram) for the composition Li_2BeF_4 at 600°C:

$$[O^{2-}] = 5 \times 10^{-4} \ (P_{H_2O}/P^2_{HF}) .$$

When verified, this relationship can provide a needed, although inconvenient, means of oxide analysis at the low oxide concentrations (< 0.06 mole/kg) encountered in these melts.

Physical Chemistry of Molten Salts: Crystal Structure of $6LiF \cdot BeF_2 \cdot ZrF_4$. – In the program of crystal structure determinations related to the study of phase relationships in fluoride systems, there is current interest in the stereochemistry of heavy-metal and transition-metal ions. Zirconium salts are therefore of interest, and a complete structure analysis of the complex stoichiometric compound $6LiF \cdot BeF_2 \cdot ZrF_4$ was undertaken. This compound is of interest also because it appears as one of the primary phases in the system $LiF-BeF_2-ZrF_4$, which furnishes the fuel solvent for the MSRE.

The compound was found to be tetragonal, having a centrosymmetric unit cell of dimensions a_0 = 6.57 A, c_0 = 18.62 A; the space group is $I4_1/amd - D^{19}_{4h}$. There are four formula weights per unit cell.

Partial three-dimensional photographic x-ray intensity data were collected. The structure was solved by the heavy-atom technique with the

aid of Patterson, electron density, and difference syntheses projected down the [010] direction; the atomic parameters were refined by least-squares adjustment.

Both the Be^{2+} and Zr^{4+} ions have discrete coordination polyhedra. The beryllium ion is surrounded by four equidistant fluoride ions at 1.57 A, forming a nearly regular tetrahedron. In this respect the beryllium coordination is similar to that in Li_2BeF_4, another primary phase in the $LiF-BeF_2-ZrF_4$ system. About the zirconium ion there are four fluoride ions at 2.07 A and four at 2.16 A, arranged as two interpenetrating tetrahedra with a common center; these eight fluoride ions form an irregular dodecahedron. This type of eightfold coordination is common for early transition-metal ions, including that of the zirconium ion in K_2ZrF_6. The other eight-coordination polyhedron frequently observed is the square antiprism exhibited by ZrF_4 itself.

Lithium ions in $6LiF \cdot BeF_2 \cdot ZrF_4$ satisfy electroneutrality and serve to connect the polyhedral ions described above but do not appear to have additional important stereochemical significance. Thus the formula should be written $Li_6BeF_4ZrF_8$.

The structure is not closely packed. Indeed, there are rather large contiguous voids; their possible significance is being investigated with reference to the escape of fluorine atoms during the radiolysis of the solid compound.

High-Temperature Aqueous Solution Chemistry. — By means of an optical cell used previously (ORNL-3477, p. 24), the effect of hydrostatic pressure in raising the temperature of second-liquid-phase formation in UO_2SO_4-H_2SO_4-H_2O solutions has been determined for various solutions containing 0.6, 1.0, and 4.0 $\underline{m}$ SO_3 and having molal ratios, $\underline{m}_{UO_3}/\underline{m}_{SO_3}$, of 0.4, 0.6, and 0.8. The experiments were performed at 300 to 430°C and at 75 to 1500 bars. In all sets of experiments the variation of temperature of immiscibility with exerted pressure was positive and linear, giving values, $\Delta t/\Delta p$, varying from +0.080 to +0.30°C/bar. In agreement with previously reported results for stoichiometric UO_2SO_4 solutions (ORNL-3477, p. 24), $\Delta t/\Delta p$ for these solutions was found to be approximately equal to $\Delta t/\Delta p$ for pure water at a constant density, the density of water at the minimum temperature of immiscibility of the solution. An even better correspondence between the behavior of the solutions and that of the solvent, pure water, was found when a constant value of the product of D and T was used instead of constant density; the product of D, the dielectric constant, and T, the absolute temperature, is a parameter of greater theoretical significance for solution chemistry than is density. With these and the previous results, the system $UO_3-SO_3-H_2O$ [J. Inorg. Nucl. Chem. 24, 995 (1962)] can now be described as a function of composition, temperature, and hydrostatic pressure.

Chemical Separation of Isotopes. — The possibility of fractionating the isotopes of beryllium, potassium, and rubidium by means of the chemical exchange

$$^{1}M_{(Hg)} + {}^{2}M^{+}_{(aq)} = {}^{2}M_{(Hg)} + {}^{1}M^{+}_{(aq)} ,$$

where ^1M and ^2M represent different isotopes of the same element, was examined. The first prerequisite, adequate solubility of the metal in mercury, was satisfied by potassium and rubidium; beryllium dissolved in mercury only to the extent of a few parts per million. Rapid exchange of the isotopic species between phases, another requirement for fractionation, was observed in both the potassium and rubidium systems. A fractionation factor, $(^{39}K/^{41}K)_{Hg}/(^{39}K/^{41}K)_{aq}$, of 1.0006 ± 0.0004 (95% confidence interval) was measured for the potassium system. A fractionation factor, $(^{86}Rb/^{85}Rb)_{Hg}/(^{86}Rb/^{85}Rb)_{aq}$, of 1.00017 ± 0.00007 (95% confidence interval) was measured for the rubidium system. The small values of these fractionation factors preclude the use of the above exchange reaction as an effective means of separating the isotopes of these elements. However, substantial separation of cesium from rubidium does occur in the cesium-rubidium system: subsequent measurements at different temperatures established the relationship

$$\log \beta = -0.679 + 301.2/T ,$$

where β is the elemental fractionation factor, $(Cs/Rb)_{Hg}/(Cs/Rb)_{aq}$. At room temperature, β is greater than 2, and only a few theoretical stages are required to achieve essentially complete separation of these elements.

TRANSURANIUM-ELEMENT PRODUCTION

Chemical Process Development: Tramex Safety. — The present Tramex flowsheet recommends the use of 8 M HCl to strip actinides from the solvent, Alamine 336·HCl in diethylbenzene. The primary reason for the high acidity is to maintain a large extraction factor for zirconium so it will remain in the solvent and not be stripped with the product. It has been found that similar results can be achieved by stripping with 1 M HCl if a dialkylphosphoric acid is used to scrub the strip product. In a laboratory mixer-settler demonstration made with tracer activities, actinides were stripped into 1 M HCl, and the strip product was scrubbed with 0.5 M di(2-ethylhexyl)phosphoric acid in Amsco diluent. Zirconium remained quantitatively in the organic phase. Ruthenium also remained in or was reextracted into the solvent and was below the limit of detection in the strip product. This corresponds to a ruthenium decontamination factor higher than 1000.

Chemical Process Development: Extractants for Recovering Trivalent Actinides from Acid Solutions. — Bidentate compounds such as tetraamylmethylenediphosphonate [TA(MDP)] and dihexylmethylenediphosphonic acid [DH(MDP)A] are strong chelating agents and can be used to extract trivalent actinides and lanthanides from 1 to 4 M HNO_3 solutions. This is in contrast to the monodentate compounds, such as tributyl phosphate, which extract significant amounts of the trivalent actinides and lanthanides from nearly neutral salt solutions or from very concentrated (stronger than 15 M) HNO_3.

Undiluted TA(MDP), which is a liquid with a viscosity similar to that of tributyl phosphate, can be used. Maximum extraction coefficients into pure TA(MDP) were obtained from solutions in the range of 2 to 4 M HNO_3, and were 37, 34, 23, and 45 for Am, Cf, Eu, and Ce respectively. Extraction coefficients into DH(MDP)A were higher by a factor of about 10^5. However, because of its high viscosity, DH(MDP)A can be used only at low concentrations. Distribution coefficients for Am, Cf, Eu, and Ce into 0.07 M DH(MDP)A in diethylbenzene diluent were about 1000 from 1 M HNO_3 and about 100 from 4 M HNO_3.

These and other bidentate extractants may be useful for recovering trivalent actinides and lanthanides from acid waste, such as that from the Purex process. However, much additional information is needed, including the behavior of other fission products and ionic contaminants.

Curium Recovery Facility. — Process equipment for recovery and purification of gram quantities of Cm^{242} and Cm^{244} is being installed in cells 3 and 4, Building 4507. This facility is equipped with a tank for dissolving irradiated targets of americium-241 oxide in aluminum, a feed-adjustment tank, mixer-settlers for one extraction cycle using the Tramex process, an evaporator for converting product from acid chlorides to acid nitrates, and a product removal station. Nearly all the equipment except the dissolver and product removal station has been installed. A number of modifications to equipment and piping were found necessary during cold testing, which is still in progress. The tantalum plating on four Hastelloy C tanks was defective, and the tanks were replaced with glass-lined ones. A nitric acid solution containing about 150 g of Am^{243}, 200 g of Cm^{244}, and 100,000 curies of rare-earth fission products is stored at the facility. This material was recovered at Savannah River from the 10 kg of plutonium irradiated for the Transuranium-Element Program.

HFIR Target Development: Preparation of Oxides for HFIR Targets. — HFIR targets made from pressed pellets of aluminum powder and actinide oxide must have a continuous aluminum phase for satisfactory heat transfer during irradiation. Acceptable prototype targets have been made with PuO_2 particles between 20 and 200 μ; however, with smaller particles the oxide phase becomes continuous. This oxide was prepared by precipitating $Pu(OH)_4$, washing, drying, calcining to the oxide at 1150°C, and grinding. However, about 25% of the oxide was less than 20 μ in diameter and could not be used. Recycle is a problem, since either dried hydroxide or high-fired oxide is difficult to dissolve, even in boiling concentrated HNO_3 containing HF catalyst. The preparation of PuO_2 by a sol-gel method would simplify the recycle problem, because the dried gel can be sized, and fines generated in this process are readily dispersible in water. Preliminary indications are that a satisfactory sol-gel process for the preparation of dense PuO_2 can be developed. Apparent plutonium sols were prepared by digesting freshly precipitated and washed $Pu(OH)_4$ at 60°C for 8 hr at a pH of 1.5. Gels produced on drying such sols at 120°C, unlike dried $Pu(OH)_4$, are readily dispersible in water; however, this gel is porous and friable. Density and hardness of the final oxide can be improved by heating the dried gel, redispersing as a sol in water, and

drying. Calcination of this material, which had been heated at 160°C for 2 hr, produced a hard glassy oxide with a density of 10.4 g/cc ± 10%.

Large-particle AmO_2 was successfully prepared by a hydroxide precipitation method similar to that used to prepare PuO_2. The hydroxide was air dried before calcination, and, during drying, small hard particles formed. After calcination, nearly all particles were between 20 and 200 μ in diameter. It is not yet known whether the formation of particles of the desired size without grinding is typical of this preparation or simply fortuitous. The density of the fired oxide was 10.0 g/cc ± 10%. Attempts to prepare americium sols by methods used for plutonium sol preparation were not successful. Other possible methods of sol preparation have not yet been investigated.

HFIR Target Development: Design and Fabrication. — Progress continued in all phases of the design, fabrication, and mockup work on the target fabrication equipment for cubicles 1, 2, and 3 of the Transuranium Facility. The work is on schedule and is approaching the 50% completion mark.

Detail designs were completed for irradiation samples containing Pu^{242}, which will be exposed in one of the reactors at Savannah River if approval is obtained from the AEC.

Transuranium Facility Design. — The main structural steel for the main building has now been erected at the Transuranium Processing Plant, and construction continues on schedule. Some problems with procuring stainless steel pipe that can pass the required ultrasonic tests have been encountered.

Fabrication of process equipment in the ORNL shops, recently begun, is behind schedule because of some delays in completing the design and because of the limited inert-gas welding facilities. An additional inert-gas box is being assembled, and a welder-qualification program is being accelerated.

Design of the facilities for cell 3, Building 4507, supporting the extraction plant in cell 4 (Curium Recovery Facility), is nearly complete, and fabrication of the equipment is in progress.

Corrosion. — The addition of either 0.0023 M $LaCl_3$ or approximately 0.001 M H_2O_2 to boiling 6 M HCl resulted in a mild acceleration of the attack on welded Zircaloy-2 specimens. In the absence of the additives, the observed 1000-hr rate was 0.2 mil/month. With $LaCl_3$ present, a fivefold increase in the rate was obtained during 1000 hr; for a similar period, the addition of H_2O_2 produced nearly a fourfold increase.

A greater sensitivity to hydrogen pickup was shown by the Zircaloy-2 specimens in the additive tests. Using specimens of similar size and shape in all tests, the hydrogen content for a specimen after 1000 hr in the additive-free boiling 6 M HCl solution was 89 ppm. For a similar period, the values increased to 360 and 540 ppm, respectively, when H_2O_2 or $LaCl_3$ was present in the test solution.

Determination of Free Acid in Process Solutions. — Encouraging results have been obtained toward the development of a reliable method for the determination of free acid in solutions containing hydrolyzable ions. High concentrations of lithium ion free the proton almost completely of

water, thereby suppressing hydrolysis. Both polarographic and potentiometric measurements of the free-acid concentration have been successfully made in the presence of aluminum and other hydrolyzable ions without the addition of complexing agents.

METALLURGY AND MATERIALS

Ceramics Research: Sintering Studies. — As increasingly high operating temperatures are sought for nuclear and other systems, the need increases for high-quality ceramics, which are normally fabricated by sintering. We are seeking improved understanding of the mechanism of sintering, particularly in materials useful in nuclear reactors. We have started to study the rate of grain growth in dense thorium oxide and cerium dioxide and the effect of grain boundaries on the removal of porosity in these materials. Rate data for thorium oxide at 1720 and 1800°C and for cerium dioxide at 1060 and 1400°C may be described by the usual equation for grain growth, $D = Kt^n$, where D is the average grain diameter at time t, and K and n are constants that depend on the specific system and the temperature. The time exponent, n, was found to be 0.77 for the ThO_2 used. A time exponent of 0.09, indicative of a very low growth rate, for the CeO_2 used to date may be attributed to calcium and magnesium impurities. The few data reported in the literature for other oxides indicate time exponents in the range from 1/3 to 1/2.

Physical Properties Studies. — Many modern technical developments, particularly in the field of nuclear energy, involve the transfer of heat at high temperatures and the design of heat transfer elements. These, in turn, require heat transport data that are more accurate and more reliable over a wider temperature range and for more materials than have been available in the past. Accordingly, we are continuing to develop improved methods of measuring and interpreting heat transport properties.

Our radial heat flow apparatus has been modified to permit the use of smaller specimens (2 in. in diameter rather than 3), which are more readily available. The changes, which include a different type of core heater, will be tested in a measurement of the thermal conductivity of type CBG graphite, the moderator material for the MSRE, from 200 to 1000°C. We have been seeking improved thermocouple materials to avoid the drift in temperature measurements that limits thermal conductivity measurements above 1100°C. At 1300°C and 2×10^{-7} torr, 90% Pt–10% Rh vs Pt, 94% Pt–6% Rh vs 70% Pt–30% Rh, W vs 74% W–26% Re, and 95% W–5% Re vs 74% W–26% Re thermocouples have shown changes of less than 5°C in 350 hr.

We have analyzed our thermal conductivity measurements on several mixtures of metals with metal oxides and have found the thermal conductivity to depend on which phase is continuous and on the volume percent of metal present. The thermal conductivity was low when the oxide phase was continuous and high when the metal was continuous. We have started a systematic study of the thermal conductivity of Al-Al_2O_3 mixtures, using the thermal comparator apparatus.

The influence of evaporation and oxidation on the total hemispherical emittance and the electrical resistivity of type 316 stainless steel was measured between 200 and 1000°C. The electrical resistivity values increased uniformly with temperature but were higher than literature values. The total hemispherical emittance of unoxidized specimens increased with temperature to 500°C. Above 500°C the emittance depended on temperature and pressure and changed due to surface oxidation or reduction during measurement. The emittance of the oxidized specimens increased with temperature and was double to triple that of the unoxidized specimens, depending on the amount of oxide present. The thinner oxide films decomposed above 800°C.

CONTROLLED THERMONUCLEAR RESEARCH

DCX-1 Facility. — A number of experiments have been performed to determine the effects of different types of movable-end-wall configurations on gas dissociation plasmas. Three sets of end walls were used; each set consisted of two identical walls, one for positive Z and one for negative Z. Each wall was constructed so that the surface presented to the plasma was a plane, parallel to the median plane, of circular cross section greater than the allowed radial extent of the plasma in the median plane. The simplest end wall was a single sheet of copper at ground potential. The two other configurations were patterned after end walls found to have a marked stabilizing effect on calutron instabilities. In each of these end walls, the surface presented to the plasma consisted of sixty 2-in.-square closely spaced copper plates supported, by insulators, on a back plate which itself was grounded. Small coils wound around the insulators produced parallel resonance of each of the elements within a desired frequency range. For one set this range was 18 to 25 Mc, and for the other it was 125 to 150 Mc. The end walls could be moved in Z, from Z = ±2 in. to Z = ±6 in. for the grounded set and to Z = ±5 in. for the other two.

The charge-exchange neutral currents and the rf signals from an electric probe and two magnetic probes were measured as functions of the end-wall separation. These measurements were performed at various pressures, and they also included observations of the effects of applying a few watts of microwave power at a frequency (26.6 kMc) suitable for electron cyclotron resonance heating in the median plane.

The distributions of the charge-exchange signals showed that the plasma did not shrink away from the end walls as they were advanced toward the median plane. As the ends were advanced inward toward the plasma, the plasma appeared to run into them as it would into any other barrier. There were marked changes in the rf spectra as the ends were advanced. With the widest end-wall separations, the spectra consisted principally of proton cyclotron harmonics (14 Mc, 28 Mc, etc.) with considerably less of certain nonharmonic frequencies (19, 25, 32, 38, and 50 Mc). Differences were noted at different pressures or with different end configurations, but generally as the ends were advanced, the nonharmonics became more prominent in relation to the harmonics. In several cases at the closest spacing, the 25-Mc signals were markedly enhanced. The

electron cyclotron heating suppressed the nonharmonics, but increased the time-averaged amplitude of the proton cyclotron harmonics, as had been previously observed. Again the decreases in plasma scattering obtained by adding microwave power were relatively minor.

The harmonic signals are the result of proton clumping, but the origin of the nonharmonics is not clear. It is realized that the unstable interactions producing them are responsible for the pulselike ejection of electrons out of the mirrors and therefore for the high plasma potential. In addition it is evident that these interactions contribute to the scattering of the plasma.

The facility is being prepared for a series of experiments using Lorentz-force trapping.

Beam-Plasma Interaction. — The apparatus used to generate the hot-electron plasma has been improved by adding a fast-closing valve on the gas feed line at its opening into the electron beam defining aperture. The valve will shut off the gas flow in about 1 msec. After valve closure the calculated time required to reduce the vacuum chamber pressure to 1/e of its valve-open pressure, using existing diffusion pumping only, is about 50 msec. Since the plasma decay constant without the valve was of this same magnitude, some increase in the duration of the plasma x-ray flux was expected and has been observed. X radiation has continued as long as 0.5 sec after beam turnoff. This is an increase by nearly a factor of 2 over the higher-pressure case.

Theory. — The calculation of anomalous plasma diffusion by microinstabilities has been extended to the high-density and -temperature (high β) regime of thermonuclear interest. In view of the accumulating evidence that macroscopic stability can be achieved by proper shaping of magnetic fields, anomalous diffusional transport is expected to be the limiting factor on plasma confinement. At high β, we find that the energy available to drive microinstabilities approaches the total diamagnetic energy, comparable to the thermal energy. Nonetheless, results continue to indicate that, even if microinstabilities do their worst, a macroscopically stable, toroidal configuration of moderate size should provide containment adequate for thermonuclear power production. The predicted critical size is a toroidal minor radius of less than 1 m.

Radio-Frequency Heating. — Further studies with an x-ray pinhole camera have confirmed earlier measurements of the plasma shape and size in the EPA facility. With a center magnetic field of ~2100 gauss, the plasma appears to have the shape of a pillbox (or oblate spheroid) with a diameter of 16 to 18 in. and a length (or minor diameter) of ~12 in. Attempts are being made to mathematically transform the radial intensity distribution from the photographic film to the actual radial density distribution in the plasma. Using the known measured sensitometric properties of the film and comparing these with the calculated bremsstrahlung spectral intensity from the plasma, satisfactory agreement is reached in regard to the number of photons per square centimeter necessary to expose the film. These measurements are also in rough agreement with the bremsstrahlung measurements made with a 3 × 3 in. NaI(Tl) crystal and a phototube, which give a density of $\sim 5 \times 10^{11}$ electrons/cm^3 at 25-kw microwave power input.

Further measurements are being made of the change in plasma size and shape as a function of magnetic-field current and microwave power. Although the studies are incomplete, they tend to indicate that the plasma volume grows as the electron cyclotron resonant heating zones are moved axially toward the midplane, and also as the microwave power is increased.

BIOLOGY AND MEDICINE

CHEMICAL CO-CARCINOGENESIS

Effects of Chemical Carcinogens on Microorganisms. — The thymine deficiency recently introduced into *Aerobacter aerogenes* has been under investigation. Response to this requirement is unusual. At thymine concentrations below 10 μg/ml, death ensues after a low cell yield (less than 5×10^7 viable cells per ml), and most of the thymine available remains untouched. At 20 μg/ml and above, the growth curve is diauxic. With thymidine, on the other hand, much of the nucleoside is converted to thymine, so that culture filtrates, upon inoculation, respond just as a culture in thymine does. Thus one may explain the poor response in thymine as being due to inadequate conversion of thymine to thymidine. Thymidylate utilization proceeds smoothly, and less than 5 μg/ml will cause the formation of a yield of more than 10^9 viable cells per ml. Accordingly, we now utilize thymidylate in lieu of the customary thymine or thymidine for our "thymineless death" investigations. It will be important to know whether the thymidylate is taken into the cell intact (for which there is no precedent) or whether utilization of thymidylate as thymidine can account for the above observations. We shall use P^{32}-labeled thymidylate to measure thymidylate uptake.

RADIATION EFFECTS

Statistical Services and Research. — Experiments have been designed to determine the possible interaction of variables that have been known, or suspected, in the past to have an influence on mortality from secondary disease. The variables selected for study in the first of these experiments are bone marrow dose, day of injection, age of donor, and sex. The experimental design chosen for the initial phase was a 1/2 replicate of a 2×2^3 factorial with four center points. The factor levels are: 10×10^6 and 50×10^6 cells of bone marrow, with center point at 30×10^6 cells; injection at 0 and 3 days after irradiation, with center point at 1.5 days; and age of donor 7 and 91 days, with center point at 49 days. The factor of sex is, of course, at two "levels," male and female. Each treatment combination is applied to 15 mice, the mice are caged 5 to a cage, and the total number of mice in each experiment is 150. The variables being observed are the proportion of mice dead after 30, 60, and

90 days out of 15 at each experimental point. If this variable is designated by $p = d/15$, the analyses are carried out on the transformed variable $\theta = \text{arc sin}\sqrt{p}$, where the variance of θ is $821.7/15 = 54.78$. Data from the first series of experiments are being analyzed at present.

COMBATING DETRIMENTAL EFFECTS OF RADIATION

Mammalian Recovery. — Electron microscopic studies of the cellular basis for an immune response are being continued. Nucleated erythrocytes have been used to determine which cells phagocytize the antigen and their location and relation to developing plasma cells.

Mice were injected intravenously with 1 ml of saline containing a 10% suspension of chicken erythrocytes. Animals were killed at 2, 4, 8, 16, and 24 hr and at 2, 3, 5, 7, and 9 days after the large single dose of antigen. Splenic tissue was removed and fixed for light and electron microscopy. The majority of chicken erythrocytes which reached the spleen were phagocytized by 24 hr, most by histiocytes in the red pulp. Erythrocytes which were injected into the mice were also phagocytized by reticular macrophages in the white pulp, predominantly in the marginal zone bordering the red pulp. Developing plasma cells were numerous in the latter areas at days 2 and 3, presumably arising from phagocytic reticular cells or their progeny, or from cells in close contact with the macrophages that enabled a transfer of immunogenicity. As the plasma cellular reaction subsided, phagocytized plasma cells were observed in both histiocytes of red pulp and reticular macrophages of white pulp. Support is given to the idea that plasmacytes represent terminal cells which proliferate and differentiate in response to antigenic stimulation.

Tissue Culture. — Scoring of human blood samples exposed to 2.5-Mev D-D neutrons and to 50- and 100-Mev protons for chromosome aberrations has been completed. An additional set of human blood samples has also been exposed to fission-spectrum neutrons from the HPRR operated in "burst" mode. Scoring of these samples for chromosomal aberrations is in progress.

Conceptual design has been completed for an experimental "package" in which human blood samples will be irradiated with beta rays in a weightless condition. This experiment is designed for the NASA Gemini-3 flight. Engineering and experimental fabrication of the prototype experiment are under way.

Studies have been made on the frequency of x-ray-induced chromatid aberrations induced at different times of the cell cycle of *Vicia faba*. It has been shown that cells irradiated in G_2 have a significantly higher number of aberrations at the subsequent metaphase than cells irradiated in S. Since two-break aberrations account for the difference, it has been hypothesized that G_2 cells have more sites in which an exchange can occur.

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Biophysics: Ultraviolet Studies with Bacteria. — We have previously noted that *Escherichia coli* B phr⁻, which is not photoreactivable under certain conditions, does exhibit photoreactivation of killing in the logarithmic growth phase at 3341 A but not at 4047 A. This led us to suggest that this "photoreactivation" might be similar in mechanism to the phenomenon of photoprotection. Studies of the dependence of the reaction on the dose rate of the reactivating radiation and on the temperature during the reactivation treatment support this conclusion. Finally, the observation that 3341-A radiation produces a large growth delay in nutrient broth, whereas the same dose at 4047 A has no effect, makes it very likely that the phenomenon observed is indeed similar in mechanism to photoprotection. We call this phenomenon "indirect photoreactivation." Similar studies with the parent strain, *E. coli* B (Harm), indicate that photoreactivation of killing in the logarithmic phase is largely of the usual photoenzymatic type at 4047 A (direct photoreactivation), but largely of the photochemical type at 3341 A (indirect photoreactivation). Thus, indirect photoreactivation may be a widespread phenomenon.

Biophysics: Cell Stage Dependence of Incorporation of H^3-Thymidine into Grasshopper Neuroblasts. — Autoradiographic studies show that grasshopper neuroblasts trapped in midmitosis by colchicine treatment and exposed to x rays (64 to 75,000 r) do not incorporate tritiated thymidine or its derivatives into DNA. The intracellular pool of thymidine derivatives is not detectably changed by exposure to x rays. The data show a correlation between radiation-induced incorporation of thymidine and the cell stage at the time of irradiation.

Biophysics: Thymine Dimers and the Recovery of Cells from the Effects of Ultraviolet Irradiation. — A few thymine dimers block DNA synthesis in *Escherichia coli*. Sensitive strains such as B_{s-1} are permanently blocked, and the dimers remain in the bacterial DNA. Strains that are more resistant to radiation (15T⁻, B, B/r) resume DNA synthesis after a recovery time that increases as the dose increases. During this time oligonucleotides, containing thymine dimers, are removed from DNA. The removal of dimers (a process that does not require DNA synthesis) seems to be necessary for recovery. We guess that another step is the incorporation of normal nucleotides into the gap left by removal of dimers, and so we looked for single chain breaks in the DNA isolated from bacteria at various times after irradiation. The DNA was denatured in formaldehyde and banded in a cesium chloride density gradient. The molecular weights calculated from bandwidth data indicate that the most resistant strain (B/r) closes the gaps that result from dimer removal more rapidly than strain B. Thus the rates of dimer removal and nucleotide replacement are sufficient to explain the wide range of sensitivities to ultraviolet radiation among strains of *E. coli*.

Biophysics: Dimerization of Thymine in the Solid State. — The conditions necessary for dimerization of thymine in the solid state are being studied. Preliminary results indicate that the absorption spectrum of

thin layers of solid thymine differs from that of thymine in solution. Exposure to ultraviolet radiation decreases the absorption, but this change may or may not be due to dimerization.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Basic Instrumentation. — The application of field-effect transistors in charge-sensitive preamplifiers was continued. The noise spectra (equivalent noise resistance R_n as a function of frequency) of several of the new insulated-gate field-effect transistor devices were measured. The R_n of these devices was dominated by a flicker component. In the frequency range from 500 cps to 1 Mc, R_n of the insulated-gate transistor was between 10 and 100 times that of p-n junction field-effect transistors.

A charge-sensitive preamplifier was developed which has bias stabilization adequate for normal operation with the input stage cooled to any temperature between 25 and −200°C (liquid-nitrogen temperature). The input stage of the preamplifier is a p-n junction field-effect transistor whose noise spectrum is a minimum at approximately −150°C. The equivalent noise resistances measured at this temperature were about one-third of the room-temperature values.

Work was continued to develop a semiconductor radiation detector that would yield information concerning not only the energy of the incident ionizing radiation but also the point of its interaction along the two-dimensional surface of the detector. A promising configuration has been determined, and an attempt is being made to fabricate such a device.

ISOTOPE DEVELOPMENT

Application of Radioisotopes in Analytical Chemistry. — A general nuclear method for the determination of sulfur is now being developed. This method is based on the release of radioiodine according to the following reaction:

$$5SO_2 + 2I^*O_3^- + 4H_2O \rightarrow SO_4^{2-} + 4H_2SO_4 + I_2^* .$$

The generated iodine is extracted and counted; its activity is proportional to the amount of SO_2 reacted. Our preliminary work indicates that a practical lower limit for sulfur is 0.5 μg. The technique can be used to determine atmospheric SO_2 for smog studies at a level of 10^{-3} ppm in 1-liter samples of gas.

In addition, we are using this method for the determination of sulfur in many other materials whenever the sulfur can be burned to SO_2.

Analysis of Gamma-Ray Nuclides by Electronic Spectra Resolution. — Programs have been written and tested for the recall of standard spectra from the library in the form of punched paper tapes or graphs. The punched paper tapes are being used to develop assay methods for short-lived nuclides.

The standard response library has been increased to include the spectra of 32 nuclides.

CIVILIAN APPLICATIONS OF NUCLEAR EXPLOSIVES

Chemical Applications of Nuclear Explosions. — Work in this program is centered about laboratory development connected with Project Coach, a planned underground detonation of a 5- to 10-kiloton nuclear device designed to produce maximum neutron fluxes in a bedded salt formation near Carlsbad, New Mexico. The purpose of the project is to produce milligram or larger quantities of transcurium elements. Recovery of the elements will involve mining thousands of tons of debris and chemically reclaiming the isotopes. Methods under study here consist in leaching the salt with water and collecting the insoluble residue. The results indicate that the residue contains the elements sought.

Reaction of the water-insoluble residues with sodium carbonate or sodium hydroxide solutions prior to leaching with acid did not increase the amount of activity over that obtained from a direct acid leach. All the anhydrite in the residue had been metathesized to calcium carbonate or calcium hydroxide, depending on whether sodium carbonate or sodium hydroxide was used.

Leaching of salt samples with well water from the test site produced about 10% more insoluble matter than leaching with distilled water, the result, probably, of the high sulfate content (about 2000 ppm) of the well water.

Petrographic analysis of rock salt samples from the Gnome site showed the principal constituents to be halite, anhydrite, quartz, and polyhalite. Minor amounts of an unidentified microcrystalline material and clay were also noted. Ultimately, we hope to separate the minerals and determine whether any particular one concentrates the transplutonium elements. So far, early results indicate that simple gross gamma counting can be used in selecting the material to be processed. Thirteen samples from different locations in the Gnome debris show virtual constancy of the ratios of plutonium to gross beta activity and of yttrium to zirconium plus niobium. Chemically, yttrium is similar to the transplutonium elements.

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