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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

SEPTEMBER 1965

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STATUS AND PROGRESS REPORT

September 1965

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October 6, 1965

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

SEPTEMBER 1965

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

GCR-ORR Loop 1 Irradiation Experiment. — Experimental fuel assembly 15, which consists of a mixed bed of loose pyrolytic-carbon-coated fuel particles of different sizes and compositions in a graphite container, is being irradiated in GCR-ORR loop 1. The assembly continued to operate satisfactorily at a maximum central fuel temperature of 1370°C and reached a maximum burnup of 1.7 at. % of the heavy metal (4.5% of the ^{235}U was consumed). The fission-gas release rates (expressed as ratio of release rate to birth rate) continue to be approximately 1×10^{-6} to 2×10^{-6} for krypton and xenon isotopes.

The coolant cleanup system has performed satisfactorily, except for removal of CO_2. The CO_2 content in the coolant increased at a rate of 1.5 ppm/day during ORR cycle 61 and reached 45 ppm at the end of the cycle on August 22, 1965. It is apparent that the molecular sieve had become saturated. Since it was regenerated during the reactor shutdown, the CO_2 level has remained below 5 ppm.

LITR Capsule Irradiation Experiments. — Capsule L-CP10, which contains loose pyrolytic-carbon-coated fuel particles in each of four compartments, was removed from LITR core position C-46 on August 26, 1965. The capsule had operated at indicated fuel central temperatures of 820 to 1052°C and had reached the desired burnup of 12.5 at. % of the heavy metal.

ORR Poolside Irradiation Experiments. — Capsule 04-8, which contains EGCR-vendor fuel material, reached an estimated burnup of 6140 Mwd per metric ton of UO_2 and is continuing to operate satisfactorily. Maximum measured cladding temperature is 1560°F.

BeO-Graphite Compatibility Experiment. — The Mark II assembly for the study of BeO-graphite compatibility in the presence of radiation continued to operate satisfactorily, except that the temperature continued to drop slightly and is now 1390°C; design temperature is 1500°C. The rate of temperature drop appears to be diminishing, and the temperature is still within the range of interest. Irradiation will continue.

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High-Burnup Coated-Particle Irradiation Experiment. — A high-burnup irradiation experiment is being conducted to evaluate the performance of loose pyrolytic-carbon-coated fuel particles in a graphite container at high burnup under conditions of interest to the TARGET reactor development program. Irradiation in the ORR F-9 facility continued, and the estimated burnup reached 17.5 at. % of the heavy metal in the UC_2 particles in the lower fuel compartment and 16 at. % of the heavy metal in the sol-gel UO_2 particles in the upper compartment. During ORR cycle 61 the measured fuel temperature was lower than desired. On August 31 the fuel temperature was increased to be within the presently desired operating limits of 1340 to 1370°C by changing the sweep gas from neon to a mixture of argon and helium.

Recently, the fission-gas release rates (expressed as ratio of release rate to birth rate) have increased significantly, from about 10^{-6} to about 10^{-4}, for krypton and xenon isotopes. Since the two fuel compartments have a common sweep gas, it is not possible to determine which type of fuel particle is the source of the increased release.

Test of Compatibility of Oxide Fuel Pellets and Graphite Under Irradiation. — An irradiation experiment is being conducted to evaluate the compatibility of $(U,Th)O_2$ fuel pellets with graphite under simulated TARGET reactor conditions. This type of fuel is a possible alternative to the coated-particle fuel in the present reference design.

The capsule is divided into two regions, and each region contains $(U,Th)O_2$ pellets concentrically positioned within a graphite structure. The two regions have a common purge and are designed to complement each other. The upper region contains pellets that have a uranium-to-thorium ratio of 9.2:90.8, which approximates the TARGET reference fuel composition (5:95), but will not be able to achieve the desired 8 to 10% burnup; the lower region contains pellets that can attain the desired burnup in approximately one year but do not have the reference composition.

The experimental assembly (designated capsule 05-10) was installed in the ORR poolside facility during the reactor shutdown of August 22–29, 1965. The capsule was held in a retracted (low neutron flux) position and at low temperature until we determined that the system for adding CO to the sweep gas was performing satisfactorily. On September 3, after the CO concentration in the sweep gas was adjusted to 230 ppm, the capsule was inserted to a position 2.90 in. from the reactor face. The central fuel temperature attained the design value of 3000°F (1650°C), corresponding to a fuel-graphite interface temperature of 2500°F (1370°C). However, a leak from the sweep-gas system allowed fission products to escape into the secondary containment, and the capsule was retracted. We are attempting to locate and repair the leak so that irradiation can continue.

Fast Gas-Cooled Reactor Fuel Element Irradiation Tests. — A series of capsule irradiation tests is being performed in the ORR poolside facility to support the development of fuel elements for a fast gas-cooled reactor. The capsules are in a predominantly thermal-neutron spectrum, and the tests are intended principally to investigate fuel-cladding interactions. The General Atomic fast gas-cooled reactor design (GCFR) is being used as a reference in the selection of test parameters.

Irradiation of the first capsule, designated P4A-1, was begun and will continue for one ORR cycle. This capsule contains a three-section

fuel element supplied by General Atomic. The upper section is an unfueled, flattened Hastelloy X tube included to provide information to aid in the design of future capsules. The two lower sections contain "fuel-supported" elements consisting of UO_2 pellets encapsulated in 0.375-in.-OD, 0.009-in.-wall Hastelloy X tubing. In the "fuel-supported" element it is intended that the fuel body support the cladding against collapse under external pressure.

The fuel specimens are being irradiated at a heat rating of 20 kw/ft and an external pressure of 800 psia. The maximum cladding temperature is 1400°F, as estimated from temperatures indicated by six thermocouples located in the NaK coolant. Thermal cycles are applied once per day in which the cladding temperature is reduced to approximately 500°F at a maximum rate of 50°F/min by manually repositioning the capsule in the pool-side facility. After eight thermal cycles the fuel cladding was still intact, as indicated by the absence of fission product activity in the primary containment. However, at this point it was necessary to retract the capsule from the high flux region because of a leak that developed in the primary containment system external to the capsule. Attempts are being made to restore the integrity of the primary containment so that the test can proceed.

Coating Large Fuel Particles with Methane. — We systematically studied the deposition of pyrolytic-carbon coatings on 530-μ-diam UC_2 particles in a 1-in.-diam coater. This was an extension of the previous study of the effects of deposition conditions on the properties of coatings on 200-μ-diam particles. Considerations of the fluidized bed volume and expansion of the bed during coating led to selection of a 50-g charge of particles and a total methane flow rate of 4 liters/min, to duplicate as nearly as possible the previous conditions. As before, deposition temperature and methane supply rate relative to initial surface area of the charge were the controllable variables studied. Experiments were run with methane supply rates ranging from 0.06 to 6.0 $cm^3\ min^{-1}\ cm^{-2}$ at temperatures ranging from 1300 to 2000°C in 100°C increments. From 1300 to 1600°C inclusive, an uncooled graphite gas injector was used. Thus the coatings produced in this temperature range were deposited under essentially isothermal conditions. However, from 1700 to 2000°C a water-cooled gas injector was used; therefore, coatings applied in this higher temperature range were deposited in a bed in which an axial temperature gradient existed and in which the nominal coating temperature was an approximate or mean value.

Although 3/16-in.-diam graphite disks were coated with each run, the disks did not seem to circulate along with the particles and, in most cases, had thinner coatings. Coating densities measured on particles were in good general agreement with those of the previous study. The highest density ($>2.2\ g/cm^3$) was achieved at 1300°C with a high methane supply rate, while densities of $2.1\ g/cm^3$ or greater were obtained at 2000°C at intermediate flow rates. A region of minimum density extends from 1300°C with a low methane supply rate to 1600°C at the maximum methane supply rate. The occurrence of high-density regions on either side of a low-density trough suggests that two different deposition mechanisms are involved.

Crystallite sizes and anisotropy factors were not measured, because the coated disk specimens were not considered representative. However, comparison of polished sections under polarized light indicated that the anisotropy of the coatings agreed generally with that found previously.

Deposition efficiencies were calculated from the weight gains of the particles and were as large as 90% at 1800 and 1900°C with intermediate methane supply rates. Slightly lower efficiencies were obtained at 2000°C due to deposition of carbon on the walls of the reaction tube just above the inlet orifice, while a marked decrease in efficiency was observed with decreasing temperature below 1600°C. This latter reduction in efficiency, from values of approximately 70% to as low as 10% at high methane supply rates at 1300°C, is caused by an increase in the fraction of methane that decomposes above the bed.

We conclude that it is possible to deposit the same types of coatings on 530-μ particles as on 200-μ particles. However, deposition temperatures below 1400°C are not practical for large particles if methane is used as the source of carbon, because of low efficiency and excessive sooting in the reaction tube. The greater fluidizing gas flow rate required for large particles makes much higher deposition rates possible (up to 600 μ/hr), but the choice of coating properties is limited at very high deposition rates. The high-density deposits favored for outer coatings are best applied at high temperatures and lower rates (50 to 100 μ/hr).

Postirradiation Examination of Pyrolytic-Carbon-Coated UO_2. — As reported previously, the behavior of coated UO_2 fuel particles during irradiation in in-pile experiments has been excellent. The R/B (ratio of fission-gas release rate to birth rate) values are in the low 10^{-8} range for ^{88}Kr for some particles even at 1600°C and 25 at. % heavy-metal burnup. We have now completed postirradiation examination on capsules A9-1 (particles OR-201), A9-2 (particles OR-298), B9-26 (particles OR-354), B9-27 (particles OR-348), C1-16 (particles OR-343), and C1-17 (particles HB-23). The coatings of some of these particles had built-in gaps, others had porous inner layers, and one (OR-343) had a low-density inner layer that was supposed to shrink to form a gap during irradiation.

The postirradiation appearance of all particles was very good. In the A9-1 and A9-2 tests, there was damage to the inner coating but not to the outer layer, even at over 25 at. % heavy-metal burnup. The particles with buffer layers showed no coating damage, and the OR-343 coating did shrink to provide a gap between the coating and fuel core. These coatings showed essentially no damage.

The oxide cores became more granular, and there were some metallic inclusions at high burnup. However, the oxide cores did not expand during irradiation, as was observed for the carbide cores, and thus exerted no stress on the coating.

Postirradiation Annealing of Coated Fuel Particles. — Postirradiation anneals were carried out on identical triplex pyrolytic-carbon-coated particles (GA-314) with different burnups; the fuel cores were $(Th,U)C_2$. In the B9-21 experiment the particles were irradiated to 0.3 at. % heavy-metal burnup, and in the C1-15 experiment the particles were irradiated to 8.9 at. % heavy-metal burnup. Both groups of particles were irradiated at 1400°C.

The rates of release of ^{140}Ba, ^{144}Ce, ^{137}Cs, and ^{89}Sr during post-irradiation annealing are listed in the table below. The values shown are the average over a 6-hr anneal following an initial 1-hr heating period at each temperature. The ^{89}Sr release rate was higher for the low-burnup specimen at all annealing temperatures. At 2000°C the ^{140}Ba and ^{144}Ce release rates were also higher for low burnup, but the ^{137}Ce release rate was higher at high burnup.

Rates of Fission Product Release from GA-314 Particles
During Postirradiation Annealing

Annealing Temperature (°C)	Capsule	Fission Product Release Rate (%/hr)			
		^{140}Ba	^{144}Ce	^{137}Cs	^{89}Sr
1370	B9-21	0.1	≦0.01	0.01	0.2
	C1-15	0.1	0.04	0.02	0.04
1700	B9-21	0.6	0.04	0.02	2.0
	C1-15	0.5	0.3	0.08	0.4
2000	B9-21	6	5	0.3	7
	C1-15	3	3	6	5

Labyrinth Seals for EGCR Main Blower. — During certain postulated emergency conditions, the EGCR main blowers could be subjected to a vaccum of 9 in. Hg on the suction side. Under such conditions, buffering seal water must be prevented from leaking into the main coolant system, to avoid a steam-graphite reaction in the reactor core. The seals originally supplied with the blowers leaked water at atmospheric operating conditions. Tests were therefore conducted to develop a new seal that would fit into the space of the original labyrinth-seal assembly and meet the no-leakage requirements. A modified Visco-type seal was developed that will operate at 9 in. Hg in the speed range 3500 to 6200 rpm with no leakage. The revised seal was recommended for installation on the EGCR blowers.

REACTOR FUELS AND MATERIALS

Nondestructive Test Development. — We are continuing studies of analytical solutions to electromagnetic induction problems, with special emphasis on eddy-current testing problems. We have revised our original computer program so that we can solve problems with any size and shape of coil and conductor specimen. The solution is flexible enough to allow the conductivity and permeability of the conductor to vary, and we can shape the field with ferrite. The only restrictions are that there be axial symmetry, sinusoidal driving currents, and linear media. At present, we are programming equations to eliminate these last two restrictions.

We can now calculate any physically observable electromagnetic phenomenon, such as induced current, induction heating density, eddy-current forces, and coil impedance. For a number of different coil and conductor configurations we calculated the coil impedances, the variation of electromagnetic field, and the phase and amplitude contours of the generated eddy currents. The expanded calculation method will be applied to a number of eddy-current tests to aid in coil design and provide further insight into eddy-current phenomenon.

Mechanical Properties. — The elevated-temperature ductility of irradiated wrought stainless steels and nickel-base alloys is reduced by helium produced by (n,α) reactions. Many structural components, such as pressure vessels and fuel-element cladding, require welding to fabricate the reactor hardware. Although the joints are usually strong enough, they must also be ductile to withstand thermal stresses. We are investigating the elevated-temperature embrittlement of stainless steel welds and brazed joints. Our initial work indicates that welds are embrittled to approximately the same degree as the wrought alloy. Because of the many variables that influence the irradiation embrittlement of wrought material, we will investigate the importance of welding variables for the ductility of the cast weld metal.

The ductility of a typical braze alloy (Ni-Cr-P) was adversely affected by the helium, but the strength of the brazed joint was not significantly influenced for temperatures below 700°C.

REACTOR COMPONENTS

Reactor Controls Development: Neutron Fluctuation Analysis. — Statistical fluctuations in the current from a neutron-sensitive ionization chamber can furnish information about neutron-emitting systems. We are using such measurements to determine different reactor parameters of interest for reactor control applications.

An experiment was performed at the Critical Facility to measure the subcritical reactivity of the HFIR production core No. 1. An analysis of the data shows that a 10-cent change in subcriticality in the range of minus 2 dollars is readily detectable. Thus, this technique can be a powerful tool to detect differences in future production cores, and it might be a useful test before cores are installed in the reactor.

During the period of the MSRE critical experiments, a series of neutron fluctuation measurements were made to determine the effective delayed-neutron fraction β_{eff}. Analysis of the power spectral density of the neutron fluctuations shows that the β_{eff} for stagnant fuel is greater than that of circulating fuel in the ratio of 1.56 ± 0.18. This is expected, because when the fuel is circulated, delayed neutrons emitted in the primary loop do not participate in the neutron chain reaction, and, consequently, β_{eff} is reduced. The ratio of β_{eff} is in good agreement with that obtained by rod calibration.

Spectral densities of the neutron fluctuation were also measured at the ORR for different reactor power and coolant flow-rate conditions. The

purpose of this investigation was to evaluate the effects of the variation of these parameters on the observed neutron power spectral density. The results show that the amplitude of the neutron power spectrum varies with the square of the reactor power and that the amplitude and frequency shift of the spectra are strongly dependent on the reactor coolant flow. These observations indicate that the reactor transfer function is modified by the reactor temperature and flow. A reactor model was established to describe these effects, and inlet temperature variation appears to be the driving function causing the change of the neutron power spectrum. Since these changes are very large, on-line monitoring of the neutron power spectral density appears promising for evaluating long-range reactor behavior stability.

Reactor Controls Development: Computer Control. — In shimming a reactor, the operator must decide whether he is compensating for normal loss of reactivity — such as burnup, fission product poisoning, or temperature change — rather than for dangerous changes in reactivity — such as might be caused by migrating fuel or poisons. For reactors operating at high flux, the reactivity may change rapidly during transients, and consequently, at the same time, shim rods must be moved rapidly to regain normal operating level. In order to assure safe operation during transient conditions as well as at steady state, an on-line computer is needed with sufficient long-term stability and accuracy to forbid dangerous shimming. A reactor model was established for the HFIR to demonstrate the feasibility of a shim computer. The model was described by a digital computer program, and computation has been performed with a CDC 1604 computer.

POWER REACTOR FUEL PROCESSING

Fluidized-Bed Volatility Pilot Plant (FBVPP). — Design of the FBVPP continues. The FBVPP is to be installed in cells 1, 2, and 3, Building 3019, for study of the processing of zirconium- and stainless-steel-clad UO_2 power reactor fuel elements.

Drawings of the primary reactor, the pyrohydrolyzer, and their primary and secondary filters are being checked prior to approval and release. Most of the drawings for other equipment are in conceptual or preliminary form. Preliminary equipment layout drawings are complete, and structural design of shielding steel and equipment supports has begun.

Preliminary material lists have been prepared prior to ordering nickel plate, sheet, and pipe.

A request for funds was approved, and a request for a directive has been submitted to the AEC. Network diagrams used with the critical-path method of scheduling are now being prepared on the Calcomp plotter, using a magnetic tape generated by the CDC 1604-A.

The compilation of data on corrosion expected in the FBVPP was reviewed again and is in final stages of completion at Battelle Memorial Institute (under subcontract).

Decontamination of the processing system (formerly the Fluoride Volatility Pilot Plant) in cells 1 and 2, Building 3019, was completed. Efforts to remove the pocket of radioactivity remaining near the bottom of the dissolver were ineffective. A more detailed survey showed that the

source of activity was a 60-r/hr section of the HF inlet pipe, indicating that there may have been a salt leak. The pipe section will be removed later for examination.

The inside surface of the dissolver was inspected and photographed through a telescope. There were many small pits and a few wide shallow pits. There were also two parallel cracks, 1/2 in. long and 1/2 in. apart, perpendicular to a girth weld, 3 ft above the distributor plate. A similar crack in the cone, observed after the zirconium program, showed no significant change during the aluminum program.

Equipment dismantling and removal were started.

Grind-Leach Process for Graphite Fuel. — A 15 × 8 in. pulverizer intended for rough crushing of graphite fuels was received and installed. The closest setting of the discharge grating passes material smaller than 1/8 in., which is feed for a newly arrived 8 × 2 in. double-roll crusher. The roll crusher is intended for final reduction of graphite fuel to a size of about 100 μ, which ensures that all coated fuel particles are ruptured.

Processes for Advanced Fuels. — Studies have been initiated on the processing of heavy-water-moderated organic-cooled reactor (HWOCR) fuels. Among the possible HWOCR fuels are SAP- and Zircaloy-clad ThO_2-UO_2 or ThC-UC. SAP is a designation for a sintered Al-Al_2O_3 dispersion. Both shear-leach and chemical decladding processes are potentially applicable to these HWOCR fuels.

Initial tests indicated that significant amounts of SAP cladding would be dissolved in a shear-leach process that included the use of boiling 10 M HNO_3 or 13 M HNO_3–0.05 M HF. About 25% of a SAP specimen that contained about 6% Al_2O_3 was dissolved in a 6-hr digestion with excess boiling 10 M HNO_3. In a similar 6-hr test with boiling 13 M HNO_3–0.05 M HF (F/Al atom ratio 0.07), about 55% of a SAP sample was dissolved, corresponding to an average rate of dissolution of 0.4 mg min^{-1} cm^{-2}.

The reactions of UC and ThC with NH_4F solutions are being studied in connection with the development of Zirflex processes for Zircaloy-clad carbide HWOCR fuels. The feasibility of Zirflex decladding probably will depend chiefly on the behavior of the irradiated carbides. Passivity of the irradiated carbide fuel would be desirable, since the carbide, if hydrolyzed, is converted to a solid fluoride. Not only is the formation of a solid fluoride undesirable from a criticality standpoint, but also the fluoride would increase the corrosivity of the system if nitric acid were used to dissolve the solid fluoride. Reaction of unirradiated UC with 4 M NH_4F at 80°C was complete in less than 5 hr, yielding an off-gas composed of NH_3, H_2, and hydrocarbons (mainly methane), and an insoluble solid that was identified as α-$NH_4F \cdot UF_4$ by x-ray diffractometry. About 20 hr was required for complete reaction of unirradiated ThC with 4 M NH_4F at 80°C. The gaseous products were similar to those obtained with UC, and the solid product was probably $ThF_4 \cdot NH_4F$.

Fluoride Volatility Processing: Dragon Fuel Recovery Method. — Graphite-matrix fuel of the Dragon type contains $(Th,U)C_2$ or (Zr,U)C particles coated with pyrolytic carbon and SiC (triplex coating). Oxidation of the graphite matrix and the outer pyrolytic-carbon coating appears feasible, but the SiC coating is resistant to chemical attack. A suggested

process is fluorination of the oxidation residue followed by dissolution for recovery of thorium and any nonvolatilized uranium. If complete recovery of the uranium by volatilization is desired, separate from thorium recovery by dissolution, the residue would be given alternate pyrohydrolytic and fluorination treatments to remove the uranium occluded in the ThF_4. The ThF_4 residue may be dissolved in refluxing 1 M $Al(NO_3)_3 \cdot 9H_2O$–2 M HNO_3, or it may be pyrohydrolyzed and dissolved in HNO_3-HF. The same oxidation-fluorination process appears practicable for zirconium-uranium carbide fuel, with complete uranium recovery obtained in fluorination by use of the pyrohydrolytic technique.

Several fluorination tests were made with triplex-coated thorium-uranium and zirconium-uranium carbide fuel particles to demonstrate destruction of the SiC and pyrolytic-carbon coatings. Treatment with fluorine at 600°C resulted in complete conversion to thorium and uranium fluorides. In fuel containing a Th/U mole ratio of 10/1, nearly 70 to 80% of the uranium was volatilized. Complete dissolution of the ThF_4 residue was achieved in 1 M $Al(NO_3)_3 \cdot 9H_2O$–2 M HNO_3 solution. In fuel with a Zr/U mole ratio of 7/1, about 50% of the uranium was volatilized. Complete solution of the residue was obtained with 13 M HNO_3–0.05 M HF.

Fluoride Volatility Processing: Chromium Fluoride Studies. – The volatile compound CrO_2F_2 is one of the products expected to form when stainless-steel-based fuel elements are processed by the fluidized-bed method. Experiments were conducted to investigate the way in which CrO_2F_2 interacts with some of the principal phases, for example, NaF, AlF_3, UF_4, and UO_3, in contact with the purge-gas stream. On exposure of these materials to CrO_2F_2 for 0.5 hr at 80°C, NaF appeared to have reacted with CrO_2F_2, producing an unidentified, bright yellow-orange phase. These results indicate that UF_4 was not oxidized to UF_5 by CrO_2F_2, even though the published estimated free energies of formation of the reactants and products had indicated a small driving force (1.6 kcal per equivalent of UF_5) for the oxidation of UF_4 in the reaction

$$CrO_2F_2 + 2UF_4 \rightleftharpoons 2UF_5 + CrO_2 \ .$$

From the relative concentrations of H_2O and HF produced [J. Am. Chem. Soc. 74, 5262 (1952)] at room temperature by the reaction

$$CrO_3 + 2HF \rightleftharpoons CrO_2F_2 + H_2O \ ,$$

we calculated the free energy of formation for CrO_2F_2 to be 193.5 kcal/mole. Based on this free-energy value, CrO_2F_2 could oxidize CrF_4 to CrF_5 (favored by 5.7 kcal per equivalent of CrF_5) and UO_2 to UO_3 (favored by 15 kcal per equivalent of UO_3):

$$CrO_2F_2 + 2CrF_4 \rightleftharpoons 2CrF_5 + CrO_2 \ ,$$

$$5CrO_2F_2 + 4UO_2 \rightleftharpoons 4UO_3 + 3CrO_2 + 2CrF_5 \ .$$

Fluoride Volatility Processing: Studies of Plutonium Retention in Fluidized Beds. – Plutonium retention studies related to the proposed fluidized-bed volatility process for stainless-steel-clad UO_2 fuel were

continued, using the 0.5-in.-OD mini-bed reactors. Two almost parallel experiments with a 5-g bed of Norton RR alumina (120 mesh), which contained 0.5 g of 20% PuO_2–UO_2 and 0.3 g of type 304L stainless steel, showed that pyrohydrolysis is useful in reducing the amount of plutonium and uranium retained in the bed after fluorination. The results were: without pyrohydrolysis, 5.9 and 2.4%, respectively; with pyrohydrolysis, 2.5 and 0.5%. The procedures are summarized below:

With Pyrohydrolysis	Without Pyrohydrolysis
1. HF-catalyzed oxidation of bed: 40-60 vol % HF-O_2 at 650°C; 2 hr	1. Same
2. Fluorinate 1 hr at 450°C Retention: Pu, 6.7%; U, 2.8%	2. Fluorinate 2 hr at 450°C Retention: Pu 5.9%; U, 2.4%
3. Pyrohydrolyze with 73-27 vol % steam-helium at 500°C; 2 hr	3. Omitted
4. Fluorinate 1 hr at 450°C Retention: Pu, 2.5%; U, 0.5%	4. Omitted

In a series of tests in the absence of both UO_2 and stainless steel, the fixation of PuO_2 into alumina (Norton RR, 120 mesh) during fluidization in an inert gas at 650°C reached a maximum value of 2.4% in 2.5 hr. Beds composed of 5 g of alumina and 0.1 g of PuO_2 were fluidized for 0.1, 2.5, 5.0, and 10.0 hr. Subsequent fluorination at 450°C for 1 hr resulted in plutonium retentions of 0.8, 2.4, 1.7, and 2.1% respectively.

In another test, the degree of retention of plutonium and uranium was appreciably influenced by the absence of HF in the oxidation cycle. When treatment with 100% oxygen for 2 hr at 650°C was substituted for a 40-60 vol % mixture of HF-O_2, a subsequent 1-hr fluorination at 450°C reduced the plutonium and uranium retentions, respectively, to 5.1 and 0.1%. Corresponding values in previous tests with 40-60 vol % mixtures of HF-O_2 have consistently ranged from 15 to 20% and from 2 to 5%.

Testing of possible substitutes for alumina has been, so far, limited to crystalline AlF_3 and sol-gel-derived thoria. With AlF_3 the plutonium retention was very similar to that obtained for alumina that had been conditioned with fluorine. A 5-g bed of alumina with 0.3 g of stainless steel and 0.5 g of 20% PuO_2–UO_2 was given a 2-hr treatment with 40% HF–O_2. After a 1-hr fluorination at 450°C, the retention was 8.7%, compared with 7.5 and 9.6% in two tests where preconditioned alumina was used. In a similar test with sol-gel thoria (100- to 150-μ particles), the plutonium retention was about half that obtained for alumina: 7.5% after a 1-hr fluorination at 450°C, compared with about 20% in tests where preconditioning of alumina was not carried out. Preconditioning of thoria has not as yet been attempted.

Fluoride Volatility Processing: Experiments on the Chemical Decladding of Stainless-Steel-Clad UO_2 Fuel in a Fluidized Bed. — The fluidized-bed volatility process for Zircaloy-clad or stainless-steel-clad UO_2 fuel consists in decladding and fuel comminution followed by fluorination to recover the hexafluorides of uranium and plutonium. The need for

making prototype flowsheet tests has led to construction of a 1-in.-OD fluidized-bed reactor and a 2-in. sched-40 nickel pipe reactor, which are equipped with disengaging sections and filters. The filters have a periodic blowback system to prevent plugging of U_3O_8 and other fines. Some initial flowsheet tests of the HF-O_2 decladding step for stainless-steel-clad fuel were made with only uranium present. Plans are being made to install and operate the 1-in.-OD reactor in a glove-box alpha facility for tests with plutonium.

Several exploratory oxidation runs in the 1-in. reactor used 20% HF in oxygen at 600°C for removing the stainless steel cladding. The bed-to-fuel ratio was 1.14. The 20% HF concentration was based on recent favorable results at Brookhaven National Laboratory. Use of 40% HF–60% O_2, the composition giving the highest attack rate, appears to lead to temperature excursions and bed sintering. Bed material in our tests was Norton RR fused alumina, and the fuel consisted of UO_2 plates wrapped in type 302 stainless steel sheet. Oxidation for 4 hr was sufficient for complete disintegration of the stainless steel, but the product was in the form of scale. Hydrogen reduction tests on this stainless steel scale in static boat tests at 600°C resulted in conversion of the Fe_2O_3 product to Fe_3O_4 and possibly some metallic iron; however, there was essentially no comminution.

Further work with reduction, oxidation, or pyrohydrolysis is planned to achieve complete comminution of the stainless steel scale. This is believed important to prevent possible caking or fusion effects in the subsequent volatilization step using fluorine.

Three decladding experiments were run in the 2-in. reactor, and caking was severe in all cases. The tests were made with Alcoa T-61 alumina (as received) as the bed material. The test elements were 3 × 3 assemblies of 0.30-in. type 304 stainless steel tubes containing UO_2 pellets. In two tests, 6-in.-long tubes were wired together with about a 0.060-in. spacing (0.360-in., center to center); in the third test, a 12-in.-long section of a prototype Consolidated Edison element was used. In all tests, wall temperature in the reaction zone was held at 550°C, and the bed fluidized at 1.25 fps with a 40% HF–60% O_2 mixture. Test durations were 4, 2, and 3 hr; the last two runs were shut down by bed plugging. In each case the test elements were about 50% declad, on the basis of weight change. Examination of the reactor showed dense chunks of caked alumina, quite frequently with no evidence of contamination by fuel-element residues. Additional testing of the T-61 alumina in the 2.0-in.-ID reactor at temperature, with a fluidizing gas flow but without a fuel assembly, resulted in caking of the alumina, whether or not HF was present.

THORIUM UTILIZATION PROGRAM

Fueled-Graphite Development: Coated-Particle Development Laboratory. — During this report period the Coated-Particle Development Laboratory safety evaluation has been officially reviewed by the Radiochemical Plant Committee, and permission for operation has been received. Nuclear materials have been introduced into this facility.

The trial coating runs using furnace components suitable for atmospheric pressure conditions were made. Microspheres of sol-gel ThO_2 were successfully coated with pyrolytic carbon. The first run was at 1600°C, and a 60-μ-thick coating with a density of 1.5 g/cm^3 was deposited on 500 g of ThO_2 spheres. The second run was at 1800°C, and a 30-μ-thick coating with a density of 1.65 g/cm^3 was deposited on 1000 g of ThO_2 spheres.

In addition, trial coating runs using the entrained-bed components were made. The coatings produced by this method are currently being evaluated.

Fueled-Graphite Development: Entrained-Bed Coating. — Two coating experiments have been carried out in the 5-in.-ID entrained-bed coating column, at 1625°C and 1700°C, respectively, using a charge of approximately 1.5 kg of 250- to 300-μ-diam sol-gel ThO_2 particles, helium as carrier, and methane as coating gas. The above temperatures were measured on the outside of the coating column. Simultaneously, the gas temperature was monitored at the nozzle exit by a thermocouple, and the temperature about 2 in. above the top of the insert was monitored by a microoptical pyrometer. The gas temperature at the nozzle exit was found to be extremely dependent on the gas flow, and partial substitution of helium by methane increased the temperature drop. Therefore, an increase in gas flow did not result in an equivalent gas velocity increase at the nozzle exit, which is a basic requirement in this design. Adjustment of cooling water flow in the nozzle support did not provide sufficient flexibility. Thus in both runs proper particle conveying could not be achieved; but, despite these complications, coating was attempted to test the remainder of the system.

In the first run, a coating approximately 10 μ thick was applied, and in the second, a coating approximately 20 μ thick. Due to the poor agitation, some particles agglomerated in the vicinity of the nozzle exit. These experiments indicated that the nozzle exit-gas velocity has to be controlled either by temperature or by pressure drop across the nozzle. The second route seems to be simpler and more flexible, and a slight modification has been designated after model studies at room temperature.

Fueled-Graphite Development: Prototype Drum Coater. — Drum-coater components have been assembled in the mockup area, and temporary service has been installed. Preliminary component checking is complete. Testing of loading, unloading, blending, and retention of particles will begin during the next reporting period.

The vacuum chamber which houses the drum coater has been sprayed inside with a coating of copper in an attempt to reduce inductive losses. The chamber and the control panel for the 50-kw motor-generator set are in place, ready for installation of service. A 64-in.-diam flange has been fabricated and was installed on the chamber. This flange mates with the maintenance enclosure.

Process Development Studies. — Tests of sol dispersers for use in sol-gel microsphere preparation were continued. For two rotary dispersers with 0.010-in.-diam orifices exposed on the circumference, the sol-drop diameters at 1.52 fps tangential velocity were the same for three sol molarities (2.0, 3.05, and 3.4 M Th) and two disperser diameters (1.25 and 1.50 in.).

For a shear-nozzle disperser with 0.016-in.-diam orifices, the mean product diameters varied with sol feed rate as follows:

Sol Feed (cm^3/min)	d_{50} for Calcined Product (μ)
4.94	310
9.9	380
15.0	425
20.0	465

A shear nozzle was operated with Paraplex G-62 as the surfactant in the 2-ethyl-1-hexanol; this gives a higher interfacial tension between the 2-ethyl-1-hexanol and the sol than the Span-80 or amine surfactants. The sol-drop diameters and product diameters were appreciably bigger when the Paraplex G-62 was used; sieving of the products to determine mean diameters is not yet complete.

Thorium-Uranium Fuel Cycle Development Facility. — Building construction of this facility is estimated to be 12% complete, compared with a scheduled 18%. Progress on procurement of items for the facility is generally satisfactory, although shield door design is behind schedule.

As to sol-gel equipment design, detailing of the liquid handling equipment and sol dryer was continued. Furnace concepts were further developed prior to the start of final design.

Sintering and Grain Growth of Sol-Gel ThO_2 and $(Th,U)O_2$. — Many differential thermal analysis (DTA) patterns of thoria gel have been obtained during the past month, using zirconium dioxide as an internal standard. To determine whether any reaction occurred between the zirconia and the thoria and alumina also present in the cell, one specimen was used for six successive DTA runs. There was no systematic change in the position or area of the monoclinic → tetragonal peak, suggesting that no significant reaction occurred. It is hoped to confirm this conclusion by x-ray diffraction.

Semiempirical calculations have suggested that for thoria the surface energy should be significantly lower for {111} faces than for {100} faces. This leads to a preferred octahedral habit rather than the previously assumed cubic habit for the thoria crystallites. The x-ray line breadths obtained from (111), (220), and (311) reflections indicate that the crystallites in this particular sample are not perfect octahedra. In addition, there is a distribution of crystallite sizes, and this makes it impossible to choose between cubic and octahedral habits by comparing BET surface area and x-ray crystallite size, as was originally hoped. Electron microscopy will have to be used to settle the question of crystal habit.

Sintering of Sol-Gel Oxide Bodies. — Several similar pellets of ThO_2–8 wt % UO_2 were fabricated to a nominal "green" density of 3.5 g/cm^3. These pellets were used to obtain comparative shrinkages and densities in

various atmospheres and the effect of various atmospheres on the oxygen-to-uranium ratio. The highest densities were achieved when an air atmosphere was used.

Conversion of UO_2 to UC. — Conversion studies were made on 300- to 400-μ-diam fused UO_2 granules containing only 60 ppm carbon; these studies were made to determine the relation between carbon content in the oxide particles and gross porosity which was observed after conversion of sol-gel UO_2 containing 0.6 wt % (approximately 12 mole %) carbon. The fused UO_2 particles were heated with a 1:1 weight ratio of lampblack in a single-fin crucible rotating at 44 rpm. Complete conversion was obtained without significant porosity, reaction bonding, or sintering after heating for 4 hr at 1800°C, the same conditions under which the sol-gel material was converted. The granules, like the microspheres, also retained their integrity and their original shape. These results indicate that an appreciable carbon content in the oxide particles will result in an inferior product after conversion. Further UO_2 conversion studies will be made using low-carbon-content sol-gel microspheres.

Process Demonstration and ^{233}U Distribution. — Installation in Building 3019 of the ^{233}U solution storage facility, with a capacity of 400 kg of ^{233}U, was completed. The five tanks, capable of storing solutions at a concentration of 180 g per liter of ^{233}U, were calibrated three times each, prior to the addition of the borosilicate glass Raschig rings which will be used as a fixed poison. The level of the glass rings will be determined at intervals by inserting an ultrasonic transducer into a closed, water-filled tube located in the center of each tank and measuring the reflection of sound waves from the rings. The instrument is presently undergoing tests to determine the effective signal from the tank walls prior to addition of the rings.

The intermediate-scale (1 kg/day) ^{233}U solvent extraction facility was enclosed with stainless steel panels containing viewing windows and glove ports. Several 8-hr operating periods with "cold" solutions were successfully completed. The system will be operated with ^{233}U when shielding of the feed and waste tanks is completed.

Effect of Firing Temperature on Product Properties of ThO_2 Microspheres. — A systematic exploration of the effects of calcination variables on crush strength, density, and roughness of thoria microspheres is being made. Previous results indicate that the interrelationship of these properties is not straightforward.

A thoria gel was used that was prepared from a standard 2 M ThO_2 sol by forming in 2-ethyl-1-hexanol and steam stripped at 100°C to final dryness. Samples were fired in CO_2 as well as air to determine whether surface area is decreased below that resulting from the use of air.

Temperature has a strong effect on crush strength, density, and roughness below about 1100°C; crush strength and density increase, and roughness decreases. Above 1100°C, temperature has no strong effect on density and roughness; crush strength decreases dramatically. The firing atmosphere had no significant effect.

Gas Release from Sol-Gel ThO_2. — When bulk UO_2 or ThO_2-UO_2 fuel elements are irradiated at high heat ratings, the fuel redistributes itself

by a sublimation-condensation process, forming long, columnar grains. During this process, fission gases or other gases trapped in the lattice or closed pores are released and add to the internal pressure within the capsule. For conventional pressed and sintered oxide pellets, gases other than fission gases are of minor import. The same statement could not be made for oxides made by the sol-gel process because the thermal decomposition of the gel could introduce large quantities of hydrogen, oxygen, nitrogen, or water into the structure. Also, the active surfaces of the fired gels readily adsorb gases, particularly CO_2, and this sorbed gas contributes to the buildup of internal pressure. But, by carefully controlling the atmosphere at critical stages of the sol-gel process, one may control the adsorbed gases. However, removal of gas trapped within the closed pores, if present, would be much more difficult. We have, therefore, conducted an experimental program to determine the quantities of trapped and sorbed gases in sol-gel ThO_2. Fortunately, results showed that the amounts of trapped gases are small.

A modified vacuum-fusion technique was used to determine the amounts of trapped gases. Fluxing agents, BeO and Al_2O_3, were used to lower the melting point of the oxide from 3300 to 1810°C.

Results of the melting experiments were compared with data from the outgassing of fine and coarse powders of sol-gel ThO_2 at 1200°C. The gas release from bulk sol-gel ThO_2 fired in Ar—4% H_2 was negligible when compared with that from fine powders. Air-firing of a duplicate sample had no appreciable effect on the gas release.

The firing atmosphere had a marked effect on the quantity of adsorbed gas for particles in the intermediate size range but only a minor effect on —200 mesh material.

Calculations indicate that 0.2 cm^3 (STP) of fission gases per gram of oxide is produced per atomic percent of burnup. Thus the release of adsorbed gases within a fuel element may have the same effect on the internal pressure of a fuel element as a fuel burnup of 15,000 to 20,000 Mwd/metric ton with 100% fission gas release.

In a continuation of the studies of the amounts of gases evolved from sol-gel thoria at temperatures up to 1000°C, the effect of multiple successive treatments in flowing CO_2-H_2O at 1000°C has been explored. The results for the reversibly adsorbed CO_2 seem to suggest semilogarithmic relationship of adsorption with time for approximately 80% of the potential reduction in gas adsorptive capacity.

The data for the "constituent gas" (that which is evolved on first evacuating the sample after an interrupted treatment) are quite similar but less consistent in their relationship to the cumulative time of treatment. Although the data show considerable scatter, they suggest that after 5 hr of exposure there is relatively little change. Future experiments will examine the effect of exposure length, starting each exposure with a fresh sample of material after conditioning with nitrogen.

Experiments are in progress to study the effect of the CO_2-H_2O treatment on sol-gel material manufactured in different gaseous environments.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — We continued to work on the inspection, maintenance, and modifications required before power operation of the MSRE.

We removed the assembly of graphite and INOR-8 specimens from the core and found that the end of the supporting graphite bar was broken, most likely by the installation of the three-piece assembly. Cursory examination of the specimens and hold-down assembly showed no effect of exposure in the reactor. A look inside the reactor vessel with a periscope disclosed two or three chips of graphite (origin unknown) lying on top of the core, but otherwise all appeared to be in excellent condition. We retrieved the broken pieces of the graphite bar in the sample hole and installed the new one-piece sample assembly. This assembly is suspended from the access plug; it is supported laterally at the top by a bushing in the core outlet screen and at the bottom by the bottom graphite bar, which was undamaged.

The fuel pump gave no sign of trouble in 1700 hr of salt circulation, but we wanted to remove the rotary element for two reasons: as a final rehearsal for remote maintenance and to permit inspection of the internal parts of the pump. This we did. When the rotary element was removed, about 34 in.^{3} of salt which had not drained was in or on the element. This salt was easily contained, and calculations showed that radiation from this amount after power operation would not be prohibitive; thus no modification was required. Metal surfaces which had been in contact with the salt were very clean, but showed no other effect of exposure. There was no sign of wear. The only dimensional change was a slight increase in diameter of the cylindrical part of the pump bowl, which required that a new, larger seal ring be installed on the rotary element. There was evidence of slight oil leakage, but it appeared that very little had gotten to the salt. Some black substance partially covered the upper surface of a small frozen puddle of salt in the volute, and there was more black material at the openings from the bowl to the pump suction. After the inspection, we reinstalled the rotary element and motor.

The control rods were removed for inspection, and we found a badly worn place on the flexible connecting section of one. This led to the discovery that one roller in that thimble was jammed, apparently because the housing was distorted when the roller assembly was welded in. A new roller assembly, designed to get around this problem, was installed and shown to roll freely. The worn part of the rod was replaced, and the rods were reinstalled.

Thermal stresses in the reactor vessel and adjacent piping are of concern because of the loss of high-temperature ductility on irradiation. On the basis of stress analyses of the reactor cell piping, we adjusted the elevation of the fuel pump and the heat exchanger to minimize piping and nozzle stresses during operation.

The on-line digital computer was modified by factory representatives to conform to latest design changes in this model, the BR-340. The computer has not yet passed the acceptance test, which requires seven days of uninterrupted operation. The troubles have not been diagnosed, but do not appear to be in the program.

Concrete-block shielding walls were erected adjacent to the radiator duct, in the fan house, and at the entrance to the coolant cell.

Insofar as possible with the reactor cell open, we tested the device's blocking lines into the cell and where necessary serviced them to reduce leakage to acceptable rates. The tests ranged from the helium block valves to the 30-in. valves in the cell ventilation line.

A large part of the operators' time was spent on the Operating Procedures: reviewing, revising, and editing in preparation for a new issue before power operation.

Metallurgy. — A study was completed on the influence of minor alloying additions on the mechanical properties of INOR-8 welds. The crack susceptibility and some high-temperature properties of INOR-8 were found to be dependent upon the minor alloying additions that were made and on the amount and type of impurities unintentionally retained.

Small changes in the amounts of manganese and silicon used in the melting practices of commercial INOR-8 were found to affect the weldability of this nickel-molybdenum alloy such that proper control could eliminate microfissuring in heavy-section weldments. The use of titanium and aluminum as deoxidizers improved the high-temperature mechanical properties of INOR-8, but the absence of carbon in this alloy drastically reduced the strength and ductility at these temperatures.

The elevated-temperature mechanical properties of aluminum- and titanium-deoxidized INOR-8 were better when silicon was left out of the initial charge, but these properties were worse when no boron was added during melting. Dramatic improvement in the elevated-temperature mechanical properties was observed when deoxidized INOR-8 contained increased amounts of titanium and magnesium or when 2% Cb was added to the alloy.

Chemical Development. — Chemical tests were conducted during the MSRE precritical and zero-power experiments for the prupose of establishing analytical base lines for use in the full-power operating period. Results of these tests not only provided the desired base lines, but also indicated that, with respect to preventing contamination of the fuel salt, current reactor operating procedures have afforded excellent protection of the fuel salt. The MSRE fuel was constituted within the reactor in two steps, first by adding ^{7}LiF-$^{238}UF_4$ to the ^{7}LiF-BeF_2-ZrF_4 (64.75-30.09-5.16 mole %) carrier salt. The mixture produced in this manner was circulated through the fuel circuit for some 250 hr during the PC-2 precritical test. The reactor fuel for the zero-power experiments was produced subsequently by adding small increments of ^{7}LiF-$^{235}UF_4$ into the pump bowl. Final composition of the salt was controlled by the amount of $^{235}UF_4$ required for sustained criticality with one control rod completely inserted.

During the initial fill operations, ^{7}LiF-BeF_2 flush salt was admitted to all parts of the fuel circuit system. On draining the reactor, flush salt remained in the freeze valves as well as residue in the drain tanks. At this point, some 80 lb of salt was unaccounted for by the weigh cell data and is presumed to have remained at these locations. As fuel was constituted at the beginning of the PC-2 test, ^{7}LiF-$^{238}UF_4$ and carrier salt were blended and circulated through the same system circuitry. On terminating the zero-power experiment, the fuel was drained into the storage tank via a single freeze valve. At this stage, therefore, the fuel salt was diluted with a significant amount of flush salt. Its effect was evident in a disparity between nominal and analytical values for uranium concentrations in PC-2 and in differences between nominal and measured ^{238}U enrichment values during the zero-power experiments. If the MSRE fuel circuit salt were diluted with no more than 140 lb of ^{7}LiF-BeF_2 flush salt, the bias between nominal and analytical values for the uranium concentration in both the precritical test, PC-2, and the zero-power experiment would be reduced to zero. We infer, therefore, that approximately

140 lb of $^{7}LiF\text{-}BeF_2$ flush salt diluent remained in the fuel circuitry at the end of the PC-1 test and was responsible for the apparent bias in nominal and analytical values for uranium concentration and ^{235}U enrichment fractions.

An interesting implication of fuel dilution by 140 lb of flush salt is that a revised figure for nominal uranium concentration at criticality becomes available. This figure now becomes 4.453 wt % uranium (of which 1.39 wt % is ^{235}U), lower by 1.013% than the previously accepted value of 4.51 wt % uranium.

Normalized values for iron, chromium, and nickel indicate that no appreciable change in the concentrations of these elements occurred in the approximately 1100-hr test period. This result is in interesting contrast to the changes in the PC-1 test, in which the concentration of chromium in the fuel and coolant circuits increased by 25 and 10 ppm, while iron in the same circuits was decreased by 52 and 50 ppm respectively. The concentration of nickel remained at approximately 5 ppm throughout the test. The free energies of formation of NiF_2, FeF_2, and CrF_2 at 1000°K are −58, −66.5, and −74 kcal per fluorine atom; that is, the fluorides NiF_2 and FeF_2 are unstable with respect to Cr^0 in the Hastelloy N vessel walls. The results of the zero-power tests as well as those from the first prenuclear test indicate that nickel is present in the MSRE salt only as a metallic phase and that iron is also present, predominantly if not entirely in metallic form. We concluded that iron and chromium concentrations varied inversely with time in the PC-1 test but not in the zero-power experiment, primarily because the $^{7}LiF\text{-}UF_4$ enriching salt contained some 2% of the uranium in the trivalent state; that is, U^{3+} in the enriching fluid served to allow the reaction

$$FeF_2 + 2UF_3 \rightleftharpoons Fe^0 + 2UF_4$$

to proceed rather than

$$Cr^0 + FeF_2 \rightleftharpoons CrF_2 + Fe^0 \ .$$

From a chemical standpoint, the operation of the MSRE during the precritical and zero-power experiments was an unqualified success. The salts were maintained in an excellent state of purity during all transfer, fill, and circulating operations. The results of chemical analyses reinforce the long-standing conclusion that pure molten fluorides are completely compatible with nickel-based alloys.

Radiation Effects. — Operational testing of the prototype of the thermal convection, molten-salt loop experiment revealed that gas buildup occurred in the horizontal return line, resulting in loss of flow after about 16 hr of operation with helium as the inert atmosphere. Recovery of flow by evacuation and pressurization was invariably successful, but since constant flow is desired during in-pile operation the autoclave was modified to eliminate this cool horizontal line. Gas transport by the flowing salt and availability of the gas in the cool line due to the solubility difference with temperature were obvious as the source of the difficulty, but not consistent with time-temperature-geometry calculations based on the solubility difference only. However, calculations based on

diffusion-controlled release of the gas under the above driving force are compatible with the observed results. The modifications to the geometry of the experiment allow operation for several hundred hours without flow stoppage from this cause.

Analysis of Radioactive MSRE Fuels. — Relatively few MSRE salt samples have been analyzed since the termination of the zero-power experiment. However, work has continued on the equipment and procedures used in analyzing the samples.

Experience gained during the zero-power experiment indicated a need for the relocation of some of the equipment within the hot cells to reduce the total time required for analyzing the MSRE samples. The changes are in progress.

The negative bias experienced in the coulometric uranium determinations has not been resolved but is still being investigated.

As was stated in the last report, the "P_2O_5" water electrolysis cell was transferred to the experimental setup for the recovery of oxide as water from the molten MSRE fuel solvent. In order to establish the necessary conditions for use of the cell as a readout device, ZrO_2 samples were added to 50 g of MSRE fuel in the hydrofluorinator. Because the cell has a maximum limit on the rate of water electrolysis without breakdown, only 20% of the effluent gas was passed through the cell in the initial runs. It was found that under these conditions the cell was still too easily overloaded. Further runs were made in which the effluent gas stream was split in a ratio of 10:1 and in a ratio of 20:1. After the optimum gas pressures and flow rates had been established, the following results were obtained:

Percentage of Effluent Gas Passed Through Cell	ZrO_2 Taken (mg)	ZrO_2 Found (mg)	% Recovery
10	25.6	26.0	101.6
10	25.8	27.1	104.9
5	72.2	72.6	100.5
5	131.0	134.8	102.9
5	135.0	136.5	101.1
5	139.0	140.0	100.7

At present, analyses are being made on approximately 50-g portions of salt which are being sampled from a 6-kg quantity of the molten MSRE fuel. The sampling of the salt and placement in the hydrofluorinator are being performed in an inert atmosphere. Two runs have been completed, giving the results of 352 and 345 ppm oxide in the fuel.

HIGH FLUX ISOTOPE REACTOR DESIGN AND DEVELOPMENT

Metallurgy. — Metals and Controls, Incorporated, the fuel element fabricator, has now completed qualification of the fabrication procedure for the inner fuel plates and has started production. A new powder metallurgy die has been fabricated for the outer plates, and the fabricator is now in the process of final qualification of this operation. Minor troubles still are being encountered with element assembly and welding.

On September 14 the spare set of control rods for the HFIR was completed and shipped to the reactor site. Experience gained in making the first set was reflected in ability to produce the spare set with dimensional control which was equal to, and in some instances better than, that of the initial set. For example, the 17.832-in.-diam inner control rod cylinder was within ±0.010 in. of its reference diameter in all measurements made along its 6-ft length, a further proof of the merits of the explosive sizing operation.

Component Development and Testing. — The alignment work on the HFIR (phase I) and spare parts (phase II) was completed in July. As a result of this alignment work, it is now possible to align any new upper and lower track assemblies in the core master standard and then install these tracks in the reactor without further adjustment. This has been demonstrated by aligning the spare upper and lower track assemblies (phase II) to a specific position with respect to the vertical center line of the core master standard; installing the phase II track assemblies in the reactor structure; and measuring the width of the annular gaps between the fuel element and the inner control rod cylinder, between the inner control rod cylinder and the four outer control rods, and between the outer control rods and the inside diameter of the removable reflector. These measurements, when compared with measurements taken at the same location on the reactor phase I control rod track, show acceptable agreement between the measured gap widths and the theoretical gap widths. These measurements were made with an eddy-current measuring device and the hydraulic-mockup inner and outer control rods.

The entire reactor core structure and control rod guidance assembly has been subjected to three two-week periods of shakedown operation, and all of the structures and guidance components have performed satisfactorily. All control rod drives, fission chamber drives, and ion chamber drives were installed and were operating during the last two-week period of shakedown operation.

Instrumentation and Controls. — The performance of the counting rate channels during critical tests indicates that the source to be used for normal operation of the reactor is adequate. During the critical tests all counting rate channels behaved as expected. The safety system was sensitized the maximum amount as limited by leakage currents. The resulting trip level, approximately 500 w, will be used throughout the series of critical tests. The reactor power level was slowly increased to confirm that the safety system operated properly. Trip signals were obtained on two channels, which produced a scram.

Installation of the cooling tower temperature-control system was completed. Work is in progress to improve the response time of the flow-sensing instrumentation. The modification, when complete, will provide

flow signals with 63% response times of approximately 250 msec for both safety and control secondary circuit systems. The newly installed control valve does not perform in accordance with the specifications. The valve manufacturer is modifying the valve controller in an effort to correct the difficulty.

The instrumentation for both the gaseous and liquid waste systems has been completed and is now on a routine maintenance program.

The health physics system, including the radiation and contamination alarm system, has been completed and is on a routine maintenance program.

Corrosion and Water Chemistry. — While the corrosion of aluminum in room-temperature water is not severe, a heavily hydrated oxide (bayerite) forms on the surface and causes contacting aluminum surfaces to adhere to each other. This was particularly apparent on the hexagonal heads of the target rods which are clamped together in the target holder; after the first test in the HFIR system, the removal of individual rods from the holder was very difficult. Laboratory tests showed that if the aluminum surfaces are treated in water at 100°C for periods of three to four days, a thin film of α-$Al_2O_3 \cdot H_2O$ (boehmite) forms on the surface. On subsequent exposure to room-temperature water, the boehmite film persists and prevents or greatly retards the formation of bayerite. All of the simulated target rods that have been exposed in the HFIR system since the first run have been pretreated in water at 100°C, and no problems of sticking have been encountered.

During the period of precritical testing, the quality of water in the HFIR primary system has remained satisfactory. With a deionization rate of 180 gpm, the specific resistivity of the water has remained at 0.5 to 1×10^6 ohm-cm. Oxygen was added to the system, and its concentration was monitored with a continuous oxygen analyzer (thallium metal type). While this type of analyzer is satisfactory under the above circumstances, it cannot be used during power operation because it combines radiolytic oxygen and hydrogen before measuring any oxygen left in solution. An oxygen-hydrogen analyzer based on the difference in thermal conductivity between the two gases is available, and plans are being made to install this instrument in place of the thallium-type analyzer.

During precritical circulation tests, an experiment was performed to determine the rate of spontaneous decomposition of hydrogen peroxide in the primary system. At an average water temperature of 40.5°C, the half-life was approximately 6 hr.

Critical Experiments. — During the past several months preparations were being made for the nuclear experiments which were to be conducted in the HFIR. Criticality was achieved for the first time in the facility on August 25, using the critical experiment (HFIRCE-3) fuel elements and control rods. Since that time numerous power distribution and rod calibration tests have been performed. The results to date are in good agreement with what was expected.

So far as we know now, the most significant difference between the HFIR and HFIRCE-3 facilities is associated with the greater amount of water and a difference in coolant channel thicknesses in the control region of the HFIR. In order to estimate shutdown margins for future cases,

before the present experiments were run, it was necessary to estimate the effect of the above differences. Measurements in the HFIR indicate that the shutdown margin for the minimum shutdown case is about 1 dollar greater than had been previously estimated. Since the estimated shutdown margin for this case was essentially zero, the 1-dollar increase constitutes a significant improvement.

Reactor Operations. – The reactor was made critical August 25, 1965, as scheduled. The initial approach to criticality went very smoothly and without incident. Critical runs are being conducted daily on a two-shift schedule to determine under various conditions the power distribution, the shutdown margin, the differential rod worths, and the void coefficients.

All of the engineers and three experienced operators have taken and passed their operator qualification tests. Training of the new operators is continuing; the written portion of their test is scheduled for completion in November.

The initial results of the iodine efficiency tests on the special building hot exhaust charcoal filters were lower than anticipated. Additional tests, conducted to determine the reason for the low efficiency, indicated two problems:

1. the seal between the filter cells and the filter frames was inadequate,
2. the 1/2-in.-thick charcoal beds are not thick enough to obtain the desired decontamination factor.

Plans are in progress to procure 1-in.-thick filters and to provide an absolute seal between the filter cells and the filter frames. It is believed that these modifications will provide the expected decontamination factor for iodine removal.

PHYSICAL RESEARCH

Reactor Operations. — During July and August the ORR operated 75.5% and the LITR 88.5% of the time.

The 240-g fuel elements have been partially burned so that a complete loading of new and partially burned elements may be tested.

The ORR control-room ventilation system was altered so that the room is no longer a part of the reactor building containment, making it possible for the operator to remain at the controls if the building air should be contaminated.

A new experiment facility was placed in service at the LITR in core position 28. A new access port was provided in the south side of the reactor tank.

The BSR accumulated 25.2 Mwhr of energy while being operated intermittently for experiments. The design of the changes required for 2-Mw operation is under way.

Waste Disposal. — A program has been initiated to increase the reliability of the equipment of the central gas disposal facility at stack 3039. The planned modifications include the following:

1. Addition of a second automatically operated steam valve to each of the turbines that drive the emergency cell-ventilation and off-gas blowers. This will reduce the possibility of turbine failures during periods of power outage.

2. Addition of a second automatic damper to the discharge of each electrically driven fan in the cell-ventilation and off-gas systems. This will reduce the possibility of recycling through the electrically driven fans during a power outage when the load is taken on by the steam-driven fans.

3. Simplification of the fan and damper control systems and removal of obsolete parts.

4. Automation of the changeover from the electrically driven blower in the pressurizable off-gas system to the steam-driven emergency standby blower.

Hot-Cell Operations. — Six fueled capsules containing coated fuel particles were disassembled and examined in detail; examination of BeO-graphite compatibility test Mark I was completed. Two UO_2 meltdowns from the Nuclear Safety Pilot Plant were examined and sampled for analysis. One aluminum plate containing Eu_2O_3 was examined; the examination included dimensional and density measurements, defilming, photography, and metallography. Three capsules containing fueled specimens and one containing tensile specimens were disassembled and packaged for shipment for General Electric.

Five ORR poolside boxes, one ORR B-8 experiment, and two monitor canes containing tensile specimens of iron, INOR-8, or stainless steel were disassembled in the ORR hot cells. About 150 tensile specimens were tested in the 3026D cells, and machines in the 3025 cells were operated full time with INOR-8 specimens. Preparations are under way to install four additional creep machines in cell 2 in Building 3025.

PHYSICS AND MATHEMATICS

Charge Spectrometry. — The initial chemical consequence of x irradiation is to produce a hole in an inner shell of one of the atoms. If the molecule contains a heavy atom, the vacancy will most likely occur with this atom and will lead to extensive ionization by means of a series of Auger processes. The highly ionized molecule then violently decomposes.

An investigation of this decomposition has been undertaken by measuring the ion fragments and their recoil energies in a specially designed mass spectrometer. Previous work was done on HI, DI, and CH_3I. The studies have now been extended to C_2H_5I, $C_2H_3D_2I$, and $Pb(CH_3)_4$. These latter studies illustrate, in particular, the effectiveness of electron transfer from all parts of the molecule to the heavy atom, which has undergone extensive multiple ionization. For example, in these studies the hydrocarbon fragments break down nearly completely into carbon and hydrogen ions, while the most abundant iodine and lead ions are respectively doubly and singly charged.

Theoretical Physics. — At the University of Colorado the elastic and inelastic scattering of 18.8-Mev protons from ^{90}Zr has been studied, and angular distributions for the elastic and 17 inelastic groups were measured. At ORNL an optical-model analysis of the elastic scattering was performed. The inelastic scattering was compared with the predictions of the shell model, using a simple two-body interaction between the incident and target nucleons. A Yukawa potential with a range of about 1 fermi and a strength of about 200 Mev gives a good fit to the excitation of the states of the $g_{9/2}^2$ configuration. Excitation of the other states was analyzed in terms of the shell-model configurations expected to contribute. Arguments were derived for the spin-flip strength being small.

The shell model has also been applied to the $^{90}Zr(p,p')$ reaction. The nuclear matrix elements, and particularly the radial form factors, have been evaluated. Interactions of Gaussian and Yukawa form between projectile and target nucleons were used. They were assumed to be central, but spin and isospin dependence was included. The shell-model orbitals were calculated for a potential of the Woods-Saxon shape. The effects of parameter variations and corrections such as may be due to nonlocality of the potentials were studied in some detail. It was shown how the data for the $^{90}Zr(p,p')$ reaction favors a Yukawa interaction with a range of about 1 fermi.

Low-Energy Neutron Time-of-Flight Spectroscopy. — Transmission measurements have been made with the ORR fast-chopper spectrometer on an 8-g sample of ^{180}W enriched to 6.93%. Measurements were made using BF_3 proportional counters at the 45-m flight station with a resolution of 1 to 2% from 0.1 to 100 ev and 50 nsec/m from 100 to 400 ev. The sample was packed to cover four slits, resulting in a sample thickness of 0.0194 atom of W per barn. The transmission data have been analyzed by the area analysis program, assuming a radiation width of 0.060 ev. Parameters have been obtained for five resonances in ^{180}W at the following energies: 15.9, 49.3, 62.7, 75.2, and 87.4 ev.

High-resolution measurements of the gamma rays from thermal neutron capture in the ten isotopes of Sn, the five isotopes of Zr, and four

isotopes of W have been made using lithium-drifted germanium detectors. The detectors had sensitive volumes of 1.9 and 6 cm^2 respectively, and had energy resolutions of ~10 kev at 6 Mev. Measurements have been made also of the gamma rays following the capture of resonance energy neutrons in the isotopes of Sn and Zr. In general, resonance capture in the Sn isotopes results in gamma spectra containing only a few strong lines, such as the transitions to the low-lying $s_{1/2}$ and $d_{3/2}$ states by capture in the even-A isotopes. In a few cases, specific gamma rays were observed with resonance neutrons which were not found with thermal neutrons. The results from all the nuclides are being compared with level diagrams obtained from (d,p) reactions and data on radioactive decay. For example, in the reaction $^{183}W(n,\gamma)^{184}W$ the ground-state transition of 7.411 Mev was observed as well as the primary transitions of 7.300 and 6.509 Mev to the first and second 2^+ states. The gamma rays of energies 0.903 and 0.793 Mev de-exciting the second 2^+ state and 0.111 Mev de-exciting the first 2^+ state were also observed.

High-Voltage Experimental Program: A Study of the $^{89}Y(p,n)^{89}Zr$ Reaction. — We have investigated 90° differential excitation functions and the angular distributions of the neutrons feeding the ground state and the first excited state of ^{89}Zr by the $^{89}Y(p,n)^{89}Zr$ reaction. The incident energy range and steps were so chosen that the 4.8-Mev isobaric analog resonance was studied in detail. The neutrons from the above reaction were detected by a time-of-flight method in conjunction with the pulsed protons, which were obtained from the ORNL 5.5-Mv accelerator. The excitation function of the neutrons feeding the first excited state of ^{89}Zr in the vicinity of the resonance behaves as if there is an interference effect between the resonance and off-resonance reaction mechanisms. Analysis of the neutron angular distributions and of the excitation functions indicates that the statistical compound-nucleus assumptions cannot be used to describe the off-resonance reaction mechanism of the $^{89}Y(p,n)^{89}Zr$ reaction around the lowest lying isobaric analog resonance of p + ^{89}Y.

High-Voltage Experimental Program: A New Scattering Chamber for Gaseous Targets. — A new scattering chamber has been constructed for use with gaseous targets. This chamber was designed with two basic criteria: a high degree of flexibility and the capacity for absolute cross-section measurements of high accuracy. Provision has been made for one movable detector and 11 fixed detectors. The target gas may be separated from the accelerator vacuum system by either a foil or a differential pumping system. A servo-operated valve is used to stabilize the target gas pressure. Measurement of known cross sections for the scattering of protons from hydrogen and krypton indicates that the chamber may be used to measure absolute cross sections to an accuracy within ±1%.

CHEMISTRY

Effects of Radiation on Analytical Methods. — The role of methanol in inhibiting chlorine production in acid chloride solutions upon irradiation with gamma rays was investigated further, and some of the results

are not in agreement with other reported data. Baybarz reported that the depletion of acid and the production of chlorine in 11 M LiCl solution upon irradiation with alpha particles can be arrested by the addition of methanol [J. Inorg. Nucl. Chem. 27, 725 (1965)]. One of the postulates considers methanol to be a scavenger of hydroxyl radicals, in accordance with the following equations:

$$\cdot OH + CH_3OH = H_2O + \cdot CH_2OH \ ,$$

$$\cdot CH_2OH + H = CH_3OH \ ,$$

in which mechanism the methanol is regenerated. Another possibility is

$$\cdot Cl + CH_3OH = HCl + \cdot CH_2OH \ .$$

This reaction replaces any acid that may be destroyed initially. The $\cdot CH_2OH$ radical can then dimerize to give glycol or react with OH radical to give formaldehyde.

When a solution 0.2 M in HCl and 11 M in LiCl is irradiated with gamma rays, chlorine is produced. If methanol is added to the extent of 2 vol %, this chlorine production is arrested for a while, but eventually resumes; meanwhile, the acidity is reduced steadily. Analysis of the gas phase indicates large amounts of methyl chloride. Long irradiations give rise to traces of methane, ethane, and an unidentified gas. Considering the previously reported finding that methanol does not prevent radiolysis of arsenazo III solutions, we tentatively conclude that methanol is not an OH scavenger in gamma-irradiated chloride solutions. We believe further that methanol scavenges the product of the initial radiolysis; thus no net chlorine is found. The following reactions are postulated to take place:

$$Cl^- + H^+ + \cdot OH = \cdot Cl + H_2O \ ,$$

$$CH_3OH + \cdot Cl = CH_3Cl + \cdot OH \ .$$

Separations Chemistry Research: Extraction Equilibria. — In continued study aimed at resolving apparent anomalies in extraction equilibria with alkylamines (ORNL-3647), the deviation from ideality of trioctylamine sulfate (TOAS) proved to be even less than that of the free-base amine (TOA). At 25°C, γ_{TOAS} dropped from 0.985 at 0.022 m to 0.637 at 0.583 m in dry benzene, as compared with γ_{TOA}, 0.957 at 0.021 m to 0.628 at 0.354 m (isopiestic balancing; partial molar volume of TOAS = 857 ml/mole). By elimination, this leaves only a hypothetical large nonideality of the extracted metal or acid complexes to account for the apparent anomalies.

The ability of dimethyl sulfoxide to form complexes with a variety of metal salts in aqueous solution suggested that high-molecular-weight dialkyl sulfoxides, R_2SO, might be useful extraction reagents. At 0.2 M reagent in chloroform, extraction was high for silver and mercury nitrates (extraction coefficient $E_a^o \gg 10$) and for gold chloride ($E_a^o > 1000$) with

di-n-hexyl and di(2-ethylhexyl) sulfoxides. With reagents of still higher molecular weight, these extractions decreased, to $E_a^o < 0.1$ with 0.2 M di-n-tetradecyl sulfoxide.

Separations Chemistry Research: Reverse-Phase Chromatography. — Reverse-phase chromatography columns consisting of dimethyldilaurylammonium chloride in isoamyl acetate on hydrophobic diatomaceous earth are being used to prepare samples of purified specific transfer-ribonucleic acids (t-RNA's). When sodium chloride gradient elution is employed at 37°C and pH 4.5, the phenylalanine-accepting t-RNA and one of the leucine-accepting t-RNA's are recovered essentially free of contamination. Using staged chromatography columns, 20 g of mixed t-RNA's from E. coli was processed in 4-g lots, and approximately 150 mg of purified active phenylalanine-accepting t-RNA and 150 mg of leucine-accepting t-RNA were prepared.

Column chromatography experiments using an inorganic ion exchanger, hydroxylapatite, and phosphate gradient elution have yielded substantial fractionation of t-RNA's. In one experiment, 14 amino-acid-accepting t-RNA's were identified in the column fractions. Experiments are under way to define the parameters controlling the fractionation and to prepare suitable hydroxylapatite.

Physicochemical experiments have been initiated, and the feasibility of measuring the molecular weight of t-RNA's by membrane osmometry has been demonstrated. Preliminary data on crude, mixed t-RNA's gave molecular-weight values from 33,000 to 36,500 in the temperature range 17 to 37°C. The apparent molecular weight seems to increase with decreasing temperature.

Chemistry of Carbides and Nitrides: Uranium and Thorium Carbides. — Substituting D_2O for H_2O had no effect on the hydrolysis of uranium and thorium carbides at 80°C. Within the limits of experimental error, the rates of gas evolution, volumes of gas evolved, and amounts of wax formed were the same with D_2O as with H_2O. Assuming that the response of a deutero-carbon on the gas chromatograph was the same as that of the corresponding hydrocarbon, then D_2O hydrolysis yielded a complex mixture of deuterium and saturated and unsaturated deutero-carbons corresponding to the hydrogen and hydrocarbons found in the H_2O hydrolysis. This indicates that the rate-determining step in the hydrolysis of the uranium and thorium carbides probably does not involve cleavage of the H-O bond in the water, since the reactions show no isotope effect.

Chemical Engineering Research: Stacked-Clone Contactor Development. — Mass-transfer efficiency studies were continued with the two-phase solvent extraction system hexone—1.7 M $Al(NO_3)_3$, 0.2 M acid deficient, with uranyl nitrate as the distributing solute. These studies were made with the 11-clone experimental stainless steel stacked-clone contactor which is being tested with many chemical systems to further evaluate performance and operating criteria. At aqueous-organic flow ratios (A/O) of about 10, mass-transfer efficiencies averaged 69% of the flooding throughput, which was 5.5 liters/min at this flow ratio.

As part of a continuing study to evaluate stacked-clone contactor performance as a function of the physical properties of the liquids, the flow capacity at flooding has been determined for the system mineral

oil—0.01 M HNO_3 at 28°C. The solvent-phase viscosity of 116 centipoises is 20 times the highest dispersed-phase viscosity used in pulsed columns. The observed throughput of 3.9 liters/min (A/O = 3) is roughly comparable to that observed with dilute sodium nitrate—TBP—Amsco systems. These results support the theory that dispersed-phase viscosity has little if any effect on flow capacity at flooding.

Chemical Engineering Research: Macromolecular Biological Separations. — In a continuation of this new activity, three runs were made to recover mixed t-RNA's from E. coli. In the first run, which started August 19, 17.2 kg of E. coli cells was processed, yielding 546 g of nucleic acid precipitates in ethanol. The second run yielded 1075 g of nucleic acid precipitates from 30.5 kg of cells processed.

In both runs the E. coli fermenter was operated with a continuous glucose nutrient feed and draw off, and the cells were concentrated and collected by a Westfalia centrifuge. Periodically, the cells were buffered, phenolated, and centrifuged. The phenol waste was discarded, and the aqueous product treated with ethanol and stored. After about 12 hr of operation, the concentration of cells in the fermenter began to decrease steadily, despite corrective measures, and the runs were stopped.

The cessation of cell growth in the fermenter was attributed to a cell-destroying virus. Prior to the third run, therefore, several possible paths by which the virus could have been introduced into the culture were closed, and a more resistant strain of E. coli (strain w) was chosen for subsequent use.

The third run began August 31 and was continued without interruption until September 3, when it was deliberately stopped. The operation was smooth and uneventful, and 300 kg of cells was processed to yield 17.4 kg of nucleic acid precipitates in ethanol.

The nucleic acid precipitates from runs 2 and 3 were combined and dissolved in sodium acetate solution, and the DNA and the heavier RNA fraction were precipitated in isopropanol, centrifuged out of the solution, and discarded. The lighter RNA fraction was then precipitated by a higher isopropanol concentration and centrifuged out of the solution. A total of 7.9 kg of the lighter RNA fraction was obtained. An analysis of this precipitate gave values for the RNA and DNA concentrations of 22.4 and 0.1% respectively. The remainder of the precipitate is an unknown contaminant, which will presumably be removed in the additional steps required to purify the product.

Mechanisms of Separations Processes. — Standard thermodynamic functions pertaining to the extraction of uranyl nitrate and nitric acid by tributyl phosphate (TBP) are being measured or calculated to place our interpretations of these extraction equilibria on a firmer mathematical foundation.

The vapor pressures of water and nitric acid over three-component solutions containing uranyl nitrate hexahydrate (UNH) were measured by the transpiration technique. Duplicate measurements were made on nitric acid solutions saturated with UNH at 25.0°C, where the initial nitric acid concentrations ranged from 0.4 to 11.35 M. Previously reported data were obtained at lower UNH concentration levels, and thus considerable extrapolation was necessary to calculate activities at or near the saturation limit. The new experimental data should result in more

accurate calculations of activities at intermediate UNH concentrations, since these calculations will be made by interpolation rather than by extrapolation.

Physical Chemistry of Polyelectrolyte Systems. – A study of the effect of cation structure on the behavior of aqueous electrolyte solutions was carried out with the quaternary ammonium halides derived from tetramethylammonium chloride and bromide by substituting either β-hydroxyethyl or benzyl groups, or both, for methyl groups. Osmotic coefficients for these solutions were measured by means of thermal electric osmometry or with the gravimetric isopiestic technique, and mean molal activity coefficients were computed from the Gibbs-Duhem relation. The coefficients for choline chloride and bromide were smaller than those for the corresponding tetramethylammonium salts at all concentrations up to 6 m. The coefficients for trimethylbenzylammonium chloride and bromide were significantly smaller than those for the tetramethylammonium salts, while those for dimethyl-β-hydroxyethylbenzylammonium chloride and bromide were smaller than those for the trimethylbenzylammonium salts at all concentrations. The "water-structure-breaking" action of the β-hydroxyethyl group as compared with a methyl group was taken to be the cause for the lower coefficients of the choline halides relative to the tetramethylammonium salts. The formation of ion pairs, possibly of the "water-structure-enforced" type, appeared to be the dominant factor causing the lower osmotic and activity coefficients observed on substituting a benzyl for a methyl group. These researches are of importance to the understanding of ion exchange selectivity and to the problem of "hydrophobic bonding" in protein and other bio-organic molecule solutions.

High-Temperature Aqueous Solution Chemistry. – The solubility of calcium hydroxide at temperatures from 0 to 350°C was determined both in water and in sodium nitrate solutions of concentrations up to 5 m. In an analogous study, the saturation concentration of calcium was determined in the presence of both calcium hydroxide and calcium carbonate to compare the effect of the additional solid, calcium carbonate. The solubilities in both systems, expressed as the concentration of calcium, varied from 0.030 to 0.00017 m in water and from 0.031 to 0.011 m in 5 m sodium nitrate at temperatures from 0 to 350°C. The overall study was made in order to determine the effect of ionic strength on these solubilities up to very high temperatures and to determine whether the data obeyed an extended Debye-Hückel equation, as those for calcium sulfate did. The solubilities were found to give a reasonably good fit to the extended equation, $\log s = \log s^{o} + S\sqrt{I}/(1 + A\sqrt{I})$, where s is the molal solubility, s^{o} is the extrapolated solubility at zero ionic strength, I is the ionic strength, S is the theoretical Debye-Hückel slope, incorporating also the charges on the several ions, and A is an adjustable parameter. Exploratory studies on magnesium hydroxide and magnesium carbonate were made at temperatures from 100 to 300°C; the measured solubilities (magnesium concentrations) were between 10^{-4} and 10^{-6} m.

Physical Chemistry of Molten Salts: Vapor Pressure Studies. – Measurement of the vapor pressures of the $LiF-BeF_2$ system (ORNL-3845, p. 25) was continued and now includes the composition range 75 to 100 mole % LiF. Over the entire composition range (0 to 100 mole % LiF)

the vapor pressure at constant temperature was found to decrease with increasing LiF concentration in the melt. Isotherms of pressure vs melt composition showed changes of inflection at 67 to 75 mole % LiF, suggesting that the predominant vapor species changes at this melt composition.

The Rodebush-Dixon apparatus, used for measuring vapor pressures, was modified so that vapor pressures in the range 1 to 5 mm Hg could be easily measured.

Physical Chemistry of Molten Salts: UO_2-ZrO_2 System. — In a previous report (ORNL-3568) it was stated that when Li_2BeF_4 melts were saturated with both UO_2 and ZrO_2 at temperatures between 500 and 700°C, the separate pure oxides were found to be the stable phases. This observation was in disagreement with published phase diagrams for the UO_2-ZrO_2 system, which predicted that appreciable solid solution formation should occur. Studies of the phase equilibria for the UO_2-ZrO_2 system have now been extended up to 1200°C.

Equilibrium products were formed within 6 hr by agitating oxide mixtures in the presence of a molten fluoride phase. Below 1115°C, colorless pure ZrO_2 was present. Above this temperature the ZrO_2 was a distinct yellow color because of the dissolution of 0.5 to 1.0 mole % of UO_2. Hence, a reactor system such as the MSRE could be heated to 1115°C before uranium would be precipitated in a stable ZrO_2-rich phase. The solubility of ZrO_2 in UO_2 was found to increase with increasing temperature. At 1115°C it is about 1 mole %. Neither uranium-zirconium oxide compounds nor uranium or zirconium oxyfluoride compounds were observed.

We believe that the differences between the results reported here and those reported previously by others reflect the inability of the other investigators to obtain equilibrium products solely via solid-state reactions. The temperature range for this investigation is more than a thousand degrees below the melting points of the pure oxides; the use of a fused fluoride phase provided a path by which the slowness of the solid-state reactions could be overcome and equilibrium achieved.

Surface Chemistry. — Calorimetric heats of immersion at 25°C of thorium oxide calcined at 1200, 1400, and 1600°C have been measured as a function of outgassing temperature from 100 to 500°C. In contrast with thorium oxide calcined at lower temperatures (650 to 1000°C), the heats of immersion were instantaneous and on a unit surface area basis were independent of calcining temperature. The heat of immersion increased from 520 to 725 ergs/cm^2 with increasing outgassing temperature and was almost identical with that of a different batch of thorium oxide which had been calcined at 1600°, but approximately 25% greater than for a 1200°-calcined sample from a third batch. The slow chemisorption of water vapor on thorium oxide, which was revealed by gravimetric studies, has been confirmed by infrared spectral measurements. In situ gravimetric studies have shown that this slow process occurs on thorium oxide calcined at 650°C or at any temperature up to at least 1200°.

Isotope Chemistry. — The Oxygen-17 Facility has performed well thus far during calendar year 1965. The Water Distillation Cascade remained on stream 100% of the time. The peak concentrations of ^{17}O and ^{18}O in this system have reached 1.8 and 90%, respectively, and are continuing

to rise. Further enrichment of these intermediate products was achieved in the Thermal Diffusion Cascade, where the peak concentrations for ^{17}O and ^{18}O reached 13.6 and 99.9%. Because the demand for 10% ^{17}O was greater than could be supplied, the peak concentration of ^{17}O in this system was restricted near 10% by maximizing the product withdrawal rate.

During this report period several component parts of the thermal diffusion columns were redesigned to minimize inleakage and to reduce filament failures. The configuration of the Thermal Diffusion Cascade, which is a function of the feed composition, was also modified to provide maximum productivity under current operating conditions. Three thermal diffusion columns, not now needed for enrichment of ^{17}O, were diverted to the separation of isotopically pure ^{16}O. This ^{16}O cascade has reached steady state and is currently producing about 5 g/month of oxygen gas in which the total ^{17}O and ^{18}O is less than 40 ppm.

TRANSURANIUM-ELEMENT PROCESSING

Process Development. — Laboratory results to date indicate that hard, stable microspheres of mixed ^{243}Am-^{244}Cm oxides can be prepared by the sol-gel process and that hard fragments of the mixed oxides can be obtained by calcination of the washed hydroxides.

The gradual disintegration of mixed ^{243}Am-^{244}Cm oxide particles previously noted for material that had been calcined at 800°C did not occur when calcination temperatures above 1100°C were used. Calcination at even higher temperatures produced more refractory material. Oxides calcined at 1250°C were extremely difficult to dissolve in either 6 M HNO_3 or HCl at 90°C, while oxides calcined at 1100°C or lower are readily soluble in 2 M HNO_3.

Present experience indicates that ^{243}Am-^{244}Cm mixtures are more amenable to sol preparations than the lanthanides; however, subsequent forming is more difficult because sol concentration is limited to about 100 g/liter by radiation-induced foaming.

Gel particles can be prepared by setting the sol in a drying solvent either in a stirred pot or in a tapered column. However, it has not been possible to prepare uniform microspheres by the stirred-pot method, since the shearing action of the paddle breaks the dilute sol droplets into very small particles of various sizes. Improved uniformity and large particles were obtained when a tapered column was used for sphere forming; however, the drying medium used did not prevent extensive agglomeration of the formed spheres. Efforts to improve forming methods will be continued.

As to the spectroscopy of californium solutions, an unsuccessful attempt was made to determine the solution spectrum of californium with only 7 μg of ^{252}Cf. It is estimated that at least 50 μg of californium will be required to obtain a solution spectrum with the present technique.

All the available californium (7 μg) was carefully purified by a series of ion exchange runs. Upon evaporation of the final chloride solution to approximately 3 μl, a definite chartreuse color could be seen. However, after conversion to a 1 M $HClO_4$ solution, the color had

completely vanished. A spectrophotometric cell (5 cm long, 3 mm in diameter) containing the californium in approximately 150 μl of solution was positioned in a Cary-14 spectrophotometer for this determination. Only a faint indication of absorption was noted in the region of 605 and 595 mμ.

Curium Recovery Facility: Solvent Extraction Processing (Cell IV). – The second ^{242}Cm processing campaign is nearly complete after two months of operation. All 18 targets have been dissolved, and feed solution corresponding to 16 of the targets has been processed through the solvent extraction equipment. The last nine targets contained less curium than the first nine (even after allowing for decay), making it necessary to revise our estimates of product available. A total of 6.5 to 7.0 g of ^{242}Cm product will be recovered.

During the first three runs of the present series, various unsuccessful attempts were made to increase the feed rate or concentration. A series of valve failures (four) have been experienced during this report period. In one instance, a leaking valve permitted ^{242}Cm product solution to seep into the process raffinate catch tank. Following the resolution of the valve difficulties, raffinate losses were only 0.03%, and the feed rate was increased from 0.7 to 0.9 liter/hr without excessive product losses.

During the latter part of the campaign, the salting strength of the scrub was raised from 11.1 to 12.0 M LiCl, and the extractant concentration was increased from 0.6 to 0.7 M amine. These changes were made to increase the distribution coefficient of the curium and improve recovery.

Fission product decontamination remains the same as reported previously. Zirconium and niobium were completely removed, and the ruthenium decontamination factor remained constant (~100), but the decay of ^{103}Ru reduced the activity of the product by a factor of 2. The observed cerium decontamination has been poor, even with a reductant, hydroxylamine hydrochloride, added to the scrub solution.

A major accomplishment of this campaign has been the development of a feed-adjustment procedure that both prevents formation of hydrolyzed zirconium and stabilizes the feed against acid loss for a period of one week. This procedure limits the maximum temperature during adjustment to 120°C rather than the 137°C used in previous runs by use of LiCl as the additive rather than HCl.

With the above feed-adjustment procedure, and with specially treated extractant (passing the extractant through a silica gel–alumina bed), the "crud" problem in the Tramex flowsheet was essentially eliminated.

HFIR Target Fabrication Development. – Fabrication of the 38 simulated HFIR target elements was completed, and they were subsequently treated by steam passivation. During the steam passivation step, the ends of some of the target elements were bent and/or blunted due to improper fixturing. As a result of this damage, the simulated targets had to be returned and repaired.

Some problems were encountered during the fabrication of 32 ^{242}Pu HFIR target elements during the month, but the elements are now essentially complete. The problems stemmed from using tubing which had a smaller inside diameter than was used previously. This smaller tubing was procured to increase the wall thickness of the target rods to help

alleviate the bowing problem of the tube during the collapse step. However, we found that loading the pellets was more difficult because of galling, and the weld contamination problem was greatly increased. After fabricating 12 target elements with the small tubing, we decided to fabricate all of the 32 ^{242}Pu targets with the tubing of the former size. The pellets from the 12 elements that had been fabricated were salvaged, and fabrication with the larger tubing was begun; no major problems have been experienced.

As to target irradiations, metallographic examination of the TRU-3 high-density prototype target is continuing. Although the reaction between the PuO_2 particles and the aluminum matrix is more extensive than in TRU-1, the general appearance is similar. The porous oxide core is surrounded by reaction products and unreacted matrix. Since the burnup level is expected to be near that of the high-heat-rated TRU-2, and the exposure time is longer, the smaller reaction zones indicate that temperature is the primary rate-controlling factor, even at long times and high burnups.

Design and Construction of Processing Plant. — Installation of process equipment by the H. K. Ferguson Company continues nearly on the schedule calling for completion by November 1. They still anticipate meeting it. Installation of most of the in-cell components is completed; cubicle service lines, equipment rack guides, and portions of the cubicle cooling equipment are not yet installed. Piping outside the cell bank, leak testing of the entire piping system, and electrical installation remain as the major incompleted work.

The lump-sum construction contract has not yet been terminated because a few items inside the building, and grading outside the building, have not been finished.

Equipment design is 98% complete. The major items remaining are those added on the recent directive charge — a shielded cave in one laboratory, revamping of the emergency power system, and a 50-ton crane at the unloading dock. ORNL shop fabrication of the remaining cell equipment will be almost 100% complete by November 1.

METALLURGY AND MATERIALS

Physical Properties Studies. — More than 30 thermal conductivity measurements were obtained on a 2-in.-diam single crystal of silicon from 400 to 800°K in the radial heat flow apparatus. The thermal resistivity of silicon can be represented by two straight lines, with a 1.5-fold slope increase occurring near 673°K. It may be significant that this slope change occurrs within the range of Debye temperatures reported for silicon. Below 673°K the temperature dependence of the thermal resistivity is significantly different from that expected for the four-phonon processes thought to occur in this temperature range. To explore this behavior further we are testing polycrystalline silicon specimens. The initial results are in surprisingly good agreement with those for single crystals.

Using the theory of heat conduction in liquids and the known behavior of other rare-earth elements and metallic melts, we estimated the thermal conductivity of liquid promethium to be between 0.1 and

0.25 w cm^{-1} $(°C)^{-1}$ at the melting point (1080°C) and between 0.1 and 0.36 at 2000°C. An equation for the mean value of these limits indicates a positive temperature dependence for the thermal conductivity of the liquid. We also estimated the electrical resistivity and thermal conductivity of solid promethium and the self-diffusion coefficient and atomic volume of liquid promethium. The estimated thermal conductivity of solid promethium had a positive temperature dependence; it varied from 0.18 at 100°C to 0.23 at the melting point.

Direct Observation of Lattice Defects. — By etch-pitting techniques, we are investigating the relative mobility of edge and screw dislocations, the distribution and structure of slip bands, and the interactions between the slip bands and barriers in single-crystal and polycrystalline tungsten under various testing temperatures and applied stresses. We hope to elucidate the role of grain boundaries during plastic deformation and the mechanisms involved in general in the ductile-brittle transition, the temperature below which tungsten becomes too brittle to fabricate. Preliminary results, obtained by etching in an aqueous solution of 15% $K_3Fe(CN)_6$ and 5% NaOH, indicate that the pits are bounded by {110} and {100} crystallographic planes. Therefore, the shapes of the pits are governed by the orientation of the specimen surface, and a computer program has been written to relate the pit shape to the orientation of the polished surface. The accuracy in determining the orientation by use of the pit shape is at least as good as that obtained in standard x-ray techniques. The etch-pitting technique has the advantage that it may be readily applied to fine-grained material. A phenomenon observed during the primary study is that etching fails to distinguish certain grain boundaries, even though pits in the adjacent grains have definitely different shapes. This implies that these grain boundaries possess a very low energy. We are using the etch-pitting technique described above to study orientation differences between these grains in order to determine the nature of the low-energy boundaries.

Physical Ceramic Studies. — Recent study of the creep in compression of polycrystalline thoria revealed that intergranular voids are created by the formation and growth of porelike openings along the boundary. We are studying the creep in much larger grained specimens, so that grain boundary effects can be analyzed in more detail, in an attempt to verify a model proposed to describe the void formation. Instead of ThO_2, we are using stoichiometric UO_2, because its lattice is identical and suitable specimens are available. The grains in the specimens are quite large, having dimensions in the range of 1/4 to 1/2 in., and the boundaries are pore free. The specimens, approximately 0.15 in. square and 0.3 in. high, contain only a few grains and a small range of boundary conditions. Specimens were tested at stresses of 11,000 and 4000 psi at 1500°C. At 11,000 psi the body deformed rapidly, decreasing in length 0.026 in. in 65 min. After compression, a polished face on the specimen displayed numerous slip bands, the grains were displaced with respect to one another, and a void was found between two grains. This void appears to be the result of crack initiation by the intersection of numerous slip bands with the grain boundary. At 4000 psi, less than 0.001 in. deformation was obtained after 115 hr at temperature. Polished faces on the specimen showed no evidence of slip bands, but the grains were displaced

with respect to one another. Examination of the cross section of the specimen revealed the formation of some grain-boundary porosity, even at this low degree of deformation.

Solid Reaction Studies. – The diffusion coefficients of ^{44}Ti in vanadium alloys containing 10, 20, and 30% Ti were determined from 1100°C to within 50° of the melting point of each alloy (1650 to 1750°C). The Arrhenius-type plots of ln D vs 1/T are curved for the 10 and 20% alloys, but the plot for 30% Ti is linear. At a given temperature, increasing the titanium content from 10 to 20% or from 20 to 30% approximately doubles the diffusion coefficient.

CONTROLLED THERMONUCLEAR RESEARCH

The DCX-1 Injection-Accumulation Experiment. – A series of experiments have been completed which have led to the suppression and identification of the microinstability in the DCX-1 for densities up to 3×10^7 cm^{-3}. In one experiment, energy spread was introduced into the stored protons through means of a single cyclotron dee driven stochastically at the fundamental gyrofrequency. These results are in very good agreement with those which used the third harmonic drive technique described previously (ORNL-3845, p. 29). That the stabilized density varies as the energy spread squared and that 30 kev spread will stabilize at a density of 3×10^7 protons/cm^3 is now supported by these two methods. But as before, it is not possible to increase the density of an instability-limited plasma ($n \approx 1 \times 10^8$ cm^{-3}) by the fundamental cyclotron drive. Instead, the drive reduces the density below the instability limit, presumably because of radial losses associated with the electric fields introduced by this technique.

By injecting obliquely to the magnetic field, we have trapped protons with a 4:1 ratio of transverse to longitudinal energy instead of the usual ratio of about 100. No stabilization effect was measured, and we conclude that axial spread is very much less effective than radial spread in stabilizing the plasma, a feature of the negative mass instability.

Further experiments which measured density thresholds at instability onset for equilibrium orbit injection gave the fraction of the total number of stored protons which participate in the instability as 1/40. The measured growth times of the instability, 50 μsec for fundamental and 10 μsec for second harmonic at $n \approx 1 \times 10^6$ cm^{-3}, when interpreted as growth times of the negative mass instability also yield the same participating fraction. The curve of stabilized density as a function of proton energy spread can be fitted by this same theory using 1/40 as the participating fraction. Furthermore, experiments which involve stabilization by injection away from the equilibrium orbit (ORNL-3825, p. 30) can be explained on the basis of a negative mass theory which considers radial oscillations of trapped protons.

This good internal consistency and agreement with theory via several routes give us confidence that we have identified the microinstability.

The negative mass instability requires a negative radial magnetic-field gradient, and the predictions of the theory are sensitive to the value of this gradient. To allow control of the radial gradient from

strongly negative to weakly positive, we have installed a pair of coils between the DCX-1 inboard coils. This may allow us to weaken and even suppress the instability and lend credence to the prediction that positive gradient fields will be free of this menace.

Properties of Plasmas Created by Electron-Cyclotron Heating. — Neutral injection experiments have continued in the EPA facility. In the new 2:1 magnetic mirror field, the density of trapped 20-kev protons obtained by injecting into the electron-cyclotron plasma (ECP) is about an order of magnitude larger than obtainable from neutral gas trapping. Typically, the beam current of 20-kev H^0 is equivalent to 60 ma.

During the period of this report, the cavity and the magnetic-field configuration were modified to produce cold plasma more efficiently with reduced neutral pressure in the center cavity. Outboard mirror coils and outboard cavity sections were added so that the resonant heating zones in these outboard cavities could produce cold plasma to feed the central cavity. The neutral pressure in the central cavity at the surface of the ECP has been reduced to 3×10^{-6} torr, a factor of 3 below that previously obtainable.

The density of 20-kev trapped protons has increased to about 3×10^7 ions/cm^3. The ions decay with e-folding time of approximately 120 μsec when injection is stopped with the ECP still present.

The disadvantage in the present arrangement has been that microwave power is fed only into the central cavity. Insufficient microwave power finds its way into the end cavity to produce cold plasma in large quantities. The waveguide system is now being modified to feed some of the microwave power directly to the end cavities.

Properties of Plasmas Created by Beam-Plasma Interaction. — We assume the neutron source strength of the region about the liner-midplane intersection in burnout V to be a measure of the electron beam-plasma interaction effect. It is necessary that ions, producing D-D target reactions in the liner wall (ground potential), have sufficient energy from the interaction to cross the magnetic field and strike the wall. Three experiments, two with BF_3 counters and one with nuclear track plates, have indicated a neutron source strength in this region of about 10^5 neutrons/sec. We are quite sure that the neutrons we observe are indeed coming from the midplane region and not from the cathode and anticathode and electrodes, because nuclear track plates at the midplane exposed to known sources at the ends were effectively shielded by the coil structure and coil cooling water. A fourth experiment, electrostatic and magnetic analysis of ions escaping the liner through a port in the midplane, gives a flux (3×10^{14} ions cm^{-2} sec^{-1}) and energy range (14 to 170 kev) which can account for the measured source strength.

A second arc has been added to the hot electron blanket experiments. The two arcs are identical but are placed 180° apart. Preliminary results show that either arc heats more efficiently, when operating by itself, without the additional gas load of the second arc. However, simultaneous operation of both arcs more than offsets the adverse effects of additional gas load inherent in two-arc operation. Hot electron densities determined from x-ray intensity measurements show increases in density with two-arc operation by a factor of 4 to 7 over that achieved with a single arc. An increase by a factor of 10 was achieved on one occasion.

Ion Source and Ion Beam Technology. — The new 100-kv ion source test facility was finished the first week in August. The system has a much greater pumping speed (10,000 liters/sec for H_2 on the main manifold and an additional ~5000 liters/sec at the target). It also permits beam studies over a total length of 92 in. and is designed to permit neutralizer studies. Magnetic analysis of beams also is provided for.

Studies have been made on this system with aligned multiple aperture source and extractor geometries using up to 91 holes. These sources were made to produce relatively large amounts of beam in an energy range below 10 kev. A 31-hole source produced 25 ma of H^+ through a 2-in. aperture at 46 in. below the center of the lens.

Efforts to increase the H_2^+ component in the source output have been continued, but no improvement has been obtained.

Theoretical Plasma Physics. — The plasma physics literature is only sparsely populated with detailed comparisons between theory and experiment. However, because the DCX-1 plasma has been diagnosed in much detail, it has been possible to identify one of its modes of behavior (rf electric field signals emitted at harmonics of proton gyrofrequency, polarized perpendicular to the external magnetic field) with the negative-mass instability, NMI, familiar from high-energy accelerators. The small negative outward radial gradient $\partial B/\partial r$ in the magnetic field allows space charge forces to collect protons into charge clusters rotating on a focused circular orbit. For a proton distribution function carefully tailored to represent the DCX-1 (under only somewhat specialized conditions), the NMI theory has now been shown to yield the following properties about as observed: (1) possibility of very low threshold densities; (2) frequencies very near proton gyroharmonics; (3) stabilization by spreading the distribution of proton energies; (4) stabilization by disordering the proton orbits — making them scalloped rather than circular; (5) two methods of stabilization, effective separately, less effective than simple addition would imply. Although simple mirror systems are susceptible to NMI, it is conjectured that projected systems with additional magnetic-field components may be made safe by eliminating focused orbits or at least those on which the average $\partial B/\partial r$ is negative.

Atomic Cross Sections. — Further studies have been made of the gas-cell conversion efficiency to produce energy-dispersed neutral beams. A monoenergetic proton beam was passed through a gas cell composed of insulating electrodes arranged so as to provide a decelerating electric field. Charge exchange collisions of the protons with gas molecules formed neutral atoms with energies characteristic of the equipotential surface on which they were formed. Application of a 40-kv retarding field resulted in a nearly flat energy distribution and a 30% conversion efficiency of incident protons to neutral particles. Exponential decelerating fields increased the beam divergence.

Optimum temperatures have been determined for a silicon surface-barrier detector coupled to a field-effect transistor preamplifier. As the detector system was cooled from room temperature, the peak width (full width at half maximum) for low-energy protons decreased to a minimum of 2.8 kev in the temperature range −100 to −120°C. Further reduction in the temperature increased the width to 3.5 kev at −150°C. Extension of the cooled surface-barrier detector to ions other than protons

resulted in a 4.7-kev peak width for 10- to 30-kev He^+ ions and a 6.7-kev width for 15-kev N^+ ions.

Searches for electron capture by H_3^+ in H_2 gas to form highly excited electronic states have been unsuccessful, implying that any capture collision forms an H_3 molecule with lifetimes less than 10^{-7} sec.

Atomic and Molecular Processes Information Center. — The second annotated bibliography containing approximately 2000 entries has been completed and is in the final editing stage. Attempts are now being made to enter the current literature into the storage system as rapidly as the published papers appear.

Our first bibliography, AMPIC-1, is being used as a data basis to determine the accuracy and completeness of our present concept of computer storage and retrieval. The computer program is being written in a universal language such that little additional effort is required to use any of our available computers.

An exhibit of the activities of the Atomic and Molecular Processes Information Center at the Fourth International Conference on Atomic Collisions in Quebec in August was well received. Since this conference, the number of requests made to the Information Center has more than doubled.

BIOLOGY AND MEDICINE

SOMATIC EFFECTS OF RADIATION

Pathology and Physiology: Observations on Viral, Chemical, and Radiation-Induced Myeloid and Lymphoid Leukemias in RF Mice. — Mice of the RF strain treated with ionizing radiation or radiomimetic alkylating agents (nitrogen mustard, triethylenemelamine, or Myerlan) show a markedly increased incidence of myeloid leukemia, thymic lymphoma, or both, depending on their age, sex, microflora, and physiological state at the time when treated, as well as on the conditions of treatment. Urethane, on the other hand, with or without irradiation, has not affected the incidence of either disease. Myeloid leukemias induced by irradiation have been serially transmitted in adult recipients by cell-free extracts. Such extracts, however, also increase the incidence of thymic lymphomas. Some of the myeloid leukemias are associated with specific chromosomal aberrations in marrow cells. In general, females are less susceptible to the induction of myeloid leukemia than males. This sex difference may be attributable at least in part to the effects of estrogen, since this hormone inhibits viral transmission of the disease. Germ-free mice appear less susceptible to the induction of myeloid leukemia by irradiation than their conventionally reared counterparts, although they are no less susceptible to induction of thymic lymphomas. These observations indicate that a complex interaction among host factors and extraneous agents determines the incidence, latency, hematologic type, clinicopathologic features, and transmissibility of leukemia in mice of the RF strain.

Medium(Low)-Level Long-Term Effects of Radiation: Quantitative Relationships of Intestinal Bacteria in Specifically Contaminated Mice. — To determine intestinal microflora relationships and their effect on host response to irradiation, a colony of RFM/Un_f mice of germ-free origin was multicontaminated with six genera of bacteria. A genetically marked strain of *Escherichia coli*, a *Streptococcus liquifaciens* group G, and a *Staphylococcus albus* were selected for the aerobic flora, while a *Bacteroides sp.*, a *Lactobacillus sp.*, and *Clostridium multifermentans* comprised the anaerobic flora. A quantitatively stable flora was established in the population within 4 to 8 weeks after introduction of the bacteria by means of the feed. The quantitative levels expressed as viable organisms per gram of feces were *Bacteroides*, 8×10^9; *Enterococcus*, 1×10^9; *Lactobacillus*, 2×10^8; *E. coli*, 1×10^8. The *Clostridium* and *Staphylococcus* species dropped to low or undetectable titers. Natural exposure in the offspring of these mice showed similar quantitative levels. Although *E. coli* has remained genetically stable for over a year in vivo, the viable count has decreased in feces of older animals.

Physiology of Blood Platelets: Size Distribution of Blood Platelets After X Irradiation. — The size distribution of blood platelets of male Sprague-Dawley rats exposed to 900 r of x radiation, approximately the $LD_{50/30}$, was determined at intervals after irradiation with a Coulter counter and size distribution plotter to study the effects of irradiation on platelet production and survival. In addition, the numbers of platelets and megakaryocytes were determined. Megakaryocytes declined one-half by 3 days and to 2% of normal by the 5th day. Similarly, platelet counts began to drop by 48 hr, fell to half-normal levels on the 4th day, and to 4% of normal by the 7th day. The time of megakaryocyte and platelet return was variable among experiments. For example, in one experiment megakaryocyte numbers increased markedly by the 16th day and platelets by the 21st day, while in another experiment only moderate increases in megakaryocytes and platelets were seen on the 24th day. No change in mean platelet size has been detected during the first week after exposure of rats to x radiation, but in samples taken between the 14th and 24th days, mean platelet size was larger than that of controls. Since earlier work showed that young platelets are larger than old platelets, the larger platelet size during the period of hemopoietic recovery from acute radiation damage is believed to be a manifestation of a younger-than-normal platelet population. The rate of platelet use as well as production is probably high at this time, thereby maintaining a young population.

RADIATION GENETICS

Human Cytogenetics. — Recent experiments with Chinese hamster cells suggest that the sister chromatid exchanges observed in autoradiographs of chromosomes labeled with tritiated thymidine are in fact interchanges induced by the beta radiation from the radioisotope.

A statistical analysis is being made of an extensive series of human chromosome measurements in an attempt to characterize the human karyotype better. A new printing-measuring instrument, developed for this project, is being evaluated.

COMBATING DETRIMENTAL EFFECTS OF RADIATION

Recovery from Somatic Effects of Radiation: An Influence of Sex on Histocompatibility in F_1 Hybrid Mice. — Bone marrow of B10.D2 mice was transplanted into (B10 × B10.D2)F_1 and reciprocal F_1 hybrid recipients exposed to 700 r of x rays. The growth of the grafted marrow was estimated 5 days later by measuring the uptake of the DNA precursor 5-iododeoxyuridine (labeled with ^{131}I) in regenerating recipient spleens. Marrow from donors of either sex grew well in F_1 females but grew deficiently in both types of F_1 males, which required about four times as many cells as the other recipients did for comparable splenic repopulation. This indicated a resistance of hybrid males not ascribable to their lack of the B10.D2 X chromosome. [(B10 × B10.D2)F_1 ♀ × B10.D2 ♂] backcross progeny segregate in two classes, one resembling B10.D2 mice with respect to sex-chromosome constitution, and the other resembling the F_1 mice for possessing sex chromosomes of different origin. In contrast, [B10.D2 ♀ × (B10.D2 × B10)F_1 ♂] male backcross progeny do not segregate with respect to their sex-chromosome constitution, all of them possessing X and Y chromosomes of different parental origin. The backcross males of both classes segregated for resistance and susceptibility to B10.D2 marrow grafts, whereas the backcross females were all susceptible. The B10.D2 strain is cogenic with strain B10 and was produced by an original cross between B10 and DBA/2 mice; it was of interest, therefore, to establish whether the trait for deficient growth of B10.D2 marrow grafts in male F_1 hybrids was derived from strain DBA/2. Marrow cells of DBA/2 donors grew deficiently in males of the hybrids (C57BL/6 × DBA/2)F_1 and (B10 × B10.D2)F_1, but not in female hybrids nor in (DBA/2 × B10.D2)F_1 mice of both sexes. The findings suggest that resistance of hybrids to B10.D2 or DBA/2 marrow grafts is due to genetic interaction between autosomal alleles of B10 and DBA/2 origin, and that its expression is sex limited.

Mammalian Recovery. — A knowledge of factors which influence or modify damage to cells exposed to ionizing radiation has been instrumental in clarifying radiobiological problems and in providing insights to practical application of protective and therapeutic measures. Although substantial progress has been made with protection, for example, chemical protection, information about methods or agents which enhance repair of radiation damage, particularly of mammalian cells, is fragmentary. The successful advances on this problem have been made almost exclusively from studies with bacteria, higher plants, protozoans, and, more recently, altered mammalian cell lines. Studies have continued in which we have attempted to apply repair measures that have been successful for the lower forms to normal mouse bone marrow cells in culture. The results show that (1) the extent of radiation injury cannot be modified by maintaining cells at any temperature from 0 to 25°C (2° intervals), (2) injury appears to be enhanced beyond 25°C, reaching a maximum at about 38°C, and (3) at 37°C, addition of nucleoproteins or nucleic acid precursors has not influenced the extent of injury. A second series of experiments involved a comparison of the effects of a single x-ray dose with the effects of the same dose split in two equal parts separated by an interval of from 2 to 6 hr. The results show progressive recovery from sublethal damage as the interval between doses increases. The

split-dose effect, first described for altered mammalian cells, applies therefore to normal mammalian cells in culture at 37°C. The effect, however, was not obtained at 2°C.

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Biophysics: Ultraviolet-Induced Free Radicals in DNA. — Studies were made on the induction of free radicals in DNA strands irradiated at dry-ice temperature and analyzed at liquid-nitrogen temperature by electron spin resonance techniques. Particular attention was given to the radical pattern which Pershan et al. [Physics 1, 163 (1964)] have determined to arise from a hydrogen addition product to thymine. Very crude action spectra suggest that the thymine radical was induced primarily by wavelengths in the range 3000 to 3500 A. Overall spectral shape, spacing of lines, their relative intensities, and saturation of the thymine signal as reported by Pershan et al. were confirmed.

Biophysics: Preliminary Results of DL-α-Alanine as a Biological Dosimeter for High-Intensity Radiation Sources. — The use of the radiation-induced electron spin resonance (ESR) in powdered DL-α-alanine as a solid-state dosimeter is based on elemental similarity to biological tissue, stability, and sensitivity to radiation damage. The use of small (10 to 500 mg) readily handled dosimeter packages permits studies of the dose distributions in phantoms or mice.

The ESR derivative signal height is directly proportional to the exposure, or dose, of ionizing radiation. Although any amino acid might be used for this purpose, the five spectral lines of alanine provide an internal check for base-line drift and signal distortion. The measured exposure, relative to Fricke solution dosimetry, yields a linear ESR signal response to values over 300 kr, although comparison with samples exposed to about 140 Mr indicates a saturation value at 2 to 5 Mr, at exposure rates of about 9.3 kr/min.

Bradshaw et al. reported a linear ESR signal response to exposures in the 100-r to 100-kr range at rates of about 1 kr/min [Radiation Res. 17, 11 (1962)].

Examination of the ESR signal of alanine under varied conditions of field modulation and microwave power indicates that there may be more than one free-radical component present. Each major line has been partially split, with typical values of 3.8 to 4.2 gauss at the center line and 5.8 to 10.8 gauss at the outermost line. Further studies at several different exposure rates with single crystals, now being grown, rather than the polycrystalline material, should enhance this splitting.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Basic Instrumentation: Elimination of Undesirable Undershoot in the Operation and Testing of Nuclear Pulse Amplifiers. — The techniques of modern network theory were applied to some of the problems of pulse shaping in nuclear pulse amplifiers. A technique referred to as pole-zero cancellation was evolved which permits a system designer to specify the pulse

decay time at the amplitude-limiting section of the system and the pulse shape at the final output as completely independent variables. The dead time of the amplifier system can be reduced by orders of magnitude without degrading the signal-to-noise ratio. In addition, the usual charge-injection system was examined, and a simple linear lumped-element network was evolved which makes possible an improved evaluation of the amplifier response to detector charge pulses.

Basic Instrumentation: Study of Field-Effect Transistor Characteristics. — The characteristics of a new low-noise junction field-effect transistor (2N3823) are being investigated. The measured noise line width of a charge-sensitive preamplifier with the device in the input stage was 1 kev full width at half maximum for silicon detectors at room temperature and 0.7 kev at the optimum temperature for zero simulated detector capacitance and a DL-RC filter. Preliminary measurements show that the optimum case temperature of the device is about −120°C.

Basic Instrumentation: Irradiation of Germanium Detectors. — Some preliminary work was done to investigate the possibility of producing large-depletion-depth germanium detectors by gamma-ray compensation. Gamma-ray irradiation of germanium creates electrically active acceptor-type defects. A high degree of compensation may eventually be attained. Two germanium diodes, one made of n-type base material, the other of p-type base material, were irradiated in a ^{60}Co source to 2×10^{15} r/cm^2. No significant change in capacitance (i.e., depletion depth) was observed, but the photovoltaic current created by the irradiation decreased significantly. This effect is interpreted as due to a reduction of the lifetime of the minority charge carriers, caused by the lattice defects introduced through irradiation.

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