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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JANUARY 1962

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OAK RIDGE NATIONAL LABORATORY

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

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

JANUARY 1962

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

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROGRAM

Pebble-Bed Reactor Experiment. – Designs of various components of the pressure envelope for the primary coolant system of the PBRE were analyzed for structural adequacy. The design of the cylindrical pressure vessel with torispherical heads was found to be satisfactory. The vessel has an inside diameter of 9 ft 1 in. and an overall height of 18 ft 4 in. and is to be fabricated of A212, grade B, carbon steel. The design pressure and temperature are 625 psi and 650°F respectively. Both the cylindrical portion and the heads are 2.5 in. thick, except in the nozzle attachment regions, where local thickness increases are required for reinforcement.

The pressure-vessel liner, which is to be made of type 304 stainless steel, is a cylindrical shell with a head in the form of a conical section capped by a spherical upper portion. Since the liner must withstand an external pressure of 3 psi at 1250°F, a 1/2-in. thickness was chosen to provide an adequate structure after a corrosion allowance of 1/8 in.

GCR-3. – Based on an economic study which indicated that the reduction in capital cost to be achieved by increasing the core power density in a gas-cooled reactor from 2 to 7 w/cm^3 would reduce the power cost by about 1 mill/kwhr and that any advantages gained by increasing the power densities beyond this level were small, calculations are being made of the feasibility of attaining core power densities of 6 to 8 w/cm^3 in large helium-cooled reactors of the EGCR type. The fuel element design chosen for this study is a seven-element cluster of 1.0-in.-OD stainless-steel-clad UO_2 pellets. The surface of the element is assumed to have low-height circumferential ribs which serve as turbulence promoters. Results of preliminary calculations have shown that the heat transfer coefficient is high enough so that the heat removal problem is internal, rather than external, to the fuel element. Further, it is evident that a much better understanding is needed of the contact resistance and fission-gas-release data.

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EGCR Top and Bottom Dummy Flow Tests. – Additional flow tests on the bottom dummy were performed for a fixed orifice 0.190 in. in diameter. Based on the orifice tests to date, the pressure differential vs flow of air through the fixed orifice can be correlated by the equation

$$W = 1080 C d_o P_1 \left\{ \frac{M}{T_1} \left[\left(\frac{P_2}{P_1} \right)^{2/k} - \left(\frac{P_2}{P_1} \right)^{(k+1)/k} \right] \right\}^{1/2} ,$$

where

W = gas flow rate, lb/hr,
C = flow coefficient,
d_o = fixed orifice diameter, in.,
P_1 = upstream static pressure, psia,
P_2 = downstream static pressure, psia,
T_1 = upstream absolute temperature, °R,
M = molecular weight of gas,
$k = c_p/c_v$ (for air) = 1.400.

The flow coefficient appears to be a function of the orifice diameter; therefore other tests with different orifice diameters are being performed to bracket the expected EGCR conditions.

GCR-ORR Loop No. 2. – The second ORNL-designed compressor was installed, and flanged joints are being leak-checked. The flexible tubing joint in the gas line that connects to the buffer gas and cleanup systems between the north cell wall and the movable rack was installed. Fabrication of the 18-in. plug components is essentially complete, and assembly has been started. The dummy fuel element material has been delivered, and fabrication has been started. The operating manual for the loop is being prepared.

Tests of Compressor with Gas-Lubricated Bearings. – Tests are being run on an experimental compressor with gas-lubricated bearings that was obtained from Bristol Siddeley Engines Limited, England. The compressor bearings will operate with a test loop pressure as low as 1 psia without indications of the bearings touching. The compressor has been operated intermittently for 750 hr and has successfully survived 20 starts and stops.

Materials Investigations. – Instantaneous collapse tests of type 304 stainless steel at 1200°F have yielded results that can be correlated with stress-strain tensile properties. Inelastic collapse pressures for perfect tubes can be determined readily by a new graphical method of solution. The effects of geometric imperfections (ovality and wall-thickness variation) on the critical pressure for collapse have also been related to the tensile properties. Ovality has been shown to cause a severe loss in stability. Wall-thickness variations within standard commercial tolerances, however, have only a minor effect and can be neglected. Thus, by knowing the expected minimum properties of a material and the expected maximum geometric imperfections, tubes can be designed to resist a given external pressure.

A study of the data accumulated in a recent test of stainless steel in helium containing CO and CO_2 in which the gases were carburizing and oxidizing has shown that carburization does not increase systematically

as the P^2_{CO}-to-P_{CO_2} ratio is increased. Enough evidence has been accumulated from various tests covering a wide range of gas ratios to indicate a maximum in the carburizing tendencies at a value of the P^2_{CO}-to-P_{CO_2} ratio of approximately 0.25. The oxidation of type 304 stainless steel in this environment ($CO_2/CO = 0.1$) should result in oxides that are stable in an atmosphere composed of 90% CO. X-ray data, however, show that Fe_3O_4 was the principal oxide formed, although it was not expected according to the published literature. It is tentatively assumed that localized gas segregation near the metal surfaces was responsible for these observations.

Solid-solution fuel particles of UO_2-ThO_2 suitable for incorporation into a BeO matrix are being studied. The weight-loss behavior of mechanical mixtures of UO_2 and ThO_2 heated in air to 1450 and 1700°C was examined in tests in which the UO_2 was added as equivalent U_3O_8, since it was anticipated that the reaction between UO_2 and ThO_2 in air would form a single-phase solid solution with an approximate composition of $UO_{2.5-2.9}$-ThO_2. The results showed that ThO_2 tended to stabilize the UO_2 with respect to oxidation in air and that the solid solution formed readily at 1450°C.

Fabrication studies are being carried out on beryllide compositions with possible application as bulk fuel for a gas-cooled converter or breeder reactor. Compositions with thorium-to-uranium ratios of 20 to 30 are being studied by using mixed beryllides and dispersions of ThO_2-UO_2 in $ThBe_{13}$. Shrinkage determinations on the $(Th_{19}U)O_2$ that will be dispersed in the $ThBe_{13}$ matrix were started. Low-calcined (500°C) sol-gel-processed $(Th,U)O_2$ was difficult to sinter in vacuum because of the large amount of adsorbed gas in the material. All sintering of the loose powder was performed in an open muffle furnace. The density of the as-received $(Th_{19}U)O_2$ particles was 6.25 g/cm^3 (61.0% of the theoretical density based on a calculated density of 10.17 g/cm^3). When the particles were sintered at 1000 and 1450°C, the respective density increases were to 9.38 and 9.96 g/cm^3 (92.2 and 98.0% of theoretical). These data show that the as-received dispersoid powder has a large shrinkage capacity.

Sintering studies on the dispersion of $(Th_{19}U)O_2$ in $ThBe_{13}$ have met with only limited success. Large cracks developed when 40 wt % as-received $(Th_{19}U)O_2$ in a $ThBe_{13}$ matrix was fired to 1550°C. These cracks were presumed to be caused by the evolution of the large amount of gases adsorbed in the as-received material. Experiments with $(Th_{19}U)O_2$ precalcined to 1000 and 1450°C resulted in a body having a number of microcracks believed to be caused by differential thermal contraction between the dispersoid and the matrix. The microstructure of the core containing the as-received dispersoid particles showed a void around each particle; however, some microcracks were also present.

Irradiation Program. — Profile and diametral measurements were made on ORR-irradiated capsule 01-4, which consisted of an instrumented stainless steel capsule containing UO_2 pellets. The element had bowed about 0.015 in., and there were bulges on the diameter of about 0.012 in. The bulges were similar to those which might be obtained from an axial twist of the tube. A similar capsule (08B-4) bowed 0.010 in., but there was no evidence of diametral changes other than slight surface depressions. The 18-in.-long fuel element specimen irradiated in GCR-ORR loop No. 1 was also measured, and bowing of about 0.010 in. was found. The maximum bowing occurred in the central one-third of the element.

Capsule E-1A, which consisted of UO_2 pellets clad with stainless steel and which was irradiated in the ETR, was found to have severe cladding failures. Two axial cracks, almost connected end to end and having a combined length of about 2.5 in., extended along the central portion of the capsule. The maximum crack width was about 0.035 in. Fission products were found in a sample of the NaK heat transfer bath, and NaK was found throughout the interior of the capsule. Two transverse sections showed central voids about 0.090 in. in diameter in the originally solid UO_2 fuel pellets. Around these central voids were columnar grains that extended out to about midradius. Radiochemical analyses indicated a higher fuel burnup than expected: 5200 Mwd per metric ton of uranium, as compared with an estimated value of 3000. The higher value is in fair agreement with neutron-flux measurements and supports other indications that the interior fuel temperature and the heat flux were much higher than estimated prior to and during irradiation.

Final decladding of the BeO capsules irradiated in ETR experiment ORNL-41-6 was completed in the hot cells. Preliminary observation of these, the largest specimens irradiated to date, shows general cracking but no deterioration to powder. All the BeO used in this experiment has been recovered, and samples are ready for analysis.

THORIUM UTILIZATION PROGRAM

Planning and Evaluation Studies. — Fuel-cost optimization of a large, one-region, one-fluid molten-salt converter reactor was continued. Increasing the diameter of the core from 15 ft to 20 ft led to lower fuel inventories and thus to lower fuel costs. The estimation of epithermal cross sections of fission products was improved and resulted in lower estimates of neutron losses in partially thermal reactors. Substantial errors in previously obtained estimates of neutron losses to Pa^{233} were detected and corrected. The fuel-cycle cost obtained for a partially optimized 1000-Mw (electrical) molten-salt converter reactor was 1.0 mill/kwhr at an average conversion ratio of 0.8; the associated specific power was 0.8 Mw (thermal) per kilogram of fissile fuel, while the average power density in the molten salt was 50 w/cc.

The treatment of fission products in the equilibrium reactor code was extended so that 115 nuclides are linked together through equations which consider transmutation and decay chains. It was found that the secondary products resulting from decay and transmutation processes increased the calculated fission product poisoning by about 20% above that associated with no secondary products, at a fuel exposure of one fission per fissile atom present.

Testing of Thoria-Pellet and -Suspension Fuels. — The development and fabrication of a loop for insertion into beam hole HN-1 of the ORR was completed. In this second in-pile slurry loop an attempt will be made to irradiate pure thoria to a fission dose with inbred U^{233} equal to or exceeding the 7×10^{16} fissions per gram of solids of the first in-pile slurry loop experiment. Several improvements in the loop are expected to increase substantially the effective irradiation rate of the slurry.

Engineering of Thorium Reactor Systems. — Friction factors and heat transfer coefficients measured at temperatures to 250°C for an 0.8-μ, 650°C-fired slurry at 500 g of ThO_2 per liter both indicated that a departure from the turbulent-flow correlation occurred at friction factors about two-thirds of the transition friction factor for Newtonian fluids. Although it can be said that the flow was not laminar at velocities below the critical velocity because of the good heat transfer, it was not possible to deduce whether the transition was due to variable viscosity or a changing velocity profile.

An experimental room-temperature loop was modified by the addition of a 100-L/D calming section, static pressure taps, and a traversing impact tube for measurement of velocity profiles and friction factors. Water calibration tests were completed, and the system was prepared for slurry addition.

Homogeneous Reactor Experiment No. 2. — The HRE-2 fuel was removed from the reactor system and was delivered to the Fission Product Pilot Plant, where it is being processed for U^{235} recovery.

Tools for cutting up the core vessel are being moved to the site.

MOLTEN-SALT REACTOR PROGRAM

MSRE Design. — Final revisions on all component drawings were completed, and approved shop drawings were issued for fabrication. Pump-mount and heat-exchanger-mount drawings were issued for approval. Layout drawings of the drain-tank cell were issued for comment. Drain-tank-mounting drawings are 75% complete. Final layout of heater and thermocouple plugs in the reactor is 50% complete.

The electrical one-line diagram was completed, with final disposition of emergency and standby power; wiring diagrams for all cell heating are 30% complete. Layout of heaters on the coolant circuit was completed. A mockup of typical primary-piping support is 75% complete.

The control rod with drive mockup is 90% complete.

Designs were completed for the thermal shield, radiator-door mechanism, and component cooling; the designs are being modified as a result of comments on the drawings.

A pressure-suppression system — to keep the containment pressure below 40 psig during the maximum credible accident — was designed, and the cost is being estimated.

MSRE Procurement and Construction. — Orders were placed for formed heads and special headers and fittings for the MSRE components. Welders in the UCNC shops have passed their tests and are now qualified to start fabrication of MSRE components. Dished heads for the pump stationary elements were fabricated by Lukens Steel Company, and preliminary inspection, before heat treating, indicated that they are satisfactory.

The contractor making the alterations to Building 7503 has qualified his welders to perform the code welding required in the drain-tank cell and the 24-ft-diam containment vessel, and work is progressing in these areas.

Chemical Development. — The freezing behavior throughout the complete range of mixtures of MSRE fuel and coolant has now been explored in sufficient detail to fully confirm previous conclusions that no high-melting combination or adverse segregation of UF_4 occurs. Dilution of the fuel, $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (70-23-5-1-1 mole %, mp 441°C), with coolant, $LiF-BeF_2$ (66-34 mole %, mp 450°C), increased the liquidus temperature in a virtually linear manner on a mole basis; at 1 mole of coolant in 25 moles of fuel, the first crystals to freeze changed from $6LiF \cdot BeF_2 \cdot ZrF_4$, as for the fuel, to Li_2BeF_4, the primary phase of the coolant.

Representative figures for the effective solubility of fuel in water are as follows: 0.0025 m U^{4+}, 0.37 m Li^+, 0.003 m Th^{4+}, 0.065 m Zr^{4+}, and 0.12 m Be^{2+} at 25°C, or 0.015, 0.57, 0.0081, 0.18, and 0.20, respectively, at 90°C. For the coolant the apparent saturation at both 25 and 90°C was reached with 0.57 m Li^+ and 0.45 m Be^{2+} in solution. All these values are expected to vary in practice due to selective leaching and to different conditions of dissolution. The amount of uranium found in the water phase at 90°C, 3.5 g/liter, probably represents the effect of oxidation by air in addition to solubility and shows that the water for quenching fuel spills should be poisoned.

Irradiation Tests. — The fourth irradiation experiment, ORNL-MTR-47-4, has been designed to determine if CF_4 will exist in the cover gas over fissioning MSRE fuel when the graphite is submerged. The cover gas from the previous irradiation experiment where the fuel was held in graphite "boats" was found to contain CF_4. The experiment also should further demonstrate the compatibility of the fuel—graphite—INOR-8 system under thermal conditions at least as severe as those expected during MSRE operation.

The four capsules designed to fulfill these objectives are INOR-8 cans, each 1 in. in diameter by 2.25 in. long, containing CGB-grade graphite cores, 1/2 in. in diameter by 1 in. long. The top of the graphite is submerged 0.45 in. below the free surface of the fuel. The cover-gas volume over the salt is ~3 cc. These capsules will be contained in a sodium bath maintained at ~1000°F. Three capsules will be filled with fuel containing 0.70 mole % UF_4, and one will have fuel with 1.47 mole %. The design values for mean interface temperatures between graphite and fuel and between INOR-8 and fuel for the two types of capsules are, respectively, 1350 and 1150°F, and 1600 and 1170°F. Natural convection in the sodium is expected to produce a range of ±75°F for the metal temperatures and ±15°F for the graphite surface temperatures. However, because of the necessity to submerge the graphite, the metal of the end cap in contact with the salt will operate at nearly the graphite surface temperature.

Two capsules designed to study the effect of power density on the formation of CF_4 also are to be placed in this experiment. These capsules will contain fuel in cylindrical graphite crucibles and should provide a basis for comparison with the previous experiments to evaluate the effectiveness of graphite submersion in preventing the net generation of CF_4. Each crucible, 1/2 in. in inside diameter and 1-3/4 in. long, will be contained in an INOR-8 can 5/8 in. in diameter and 2.25 in. long.

Out-of-pile tests with dummy capsules have been utilized in the development of a hot-filling technique and apparatus which is now ready for

testing the in-pile capsules. The results from three types of thermal-cycling tests on dummy capsules indicate that the in-pile capsules during irradiation should survive the thermal cycling anticipated from MTR shutdown and startup. It is possible that some CF_4 may be released to the cover gas during shutdown through cracks in the frozen salt.

The capsule design was completed, and work is progressing on the fabrication of the in-pile capsules and the supporting equipment. The target date for insertion at the MTR is March 12; the scheduled irradiation period is 12 weeks.

Radiation Effects. – After 0.3 Mwhr of exposure (~10% burnup) the helium cover gas above MSRE fuel in graphite boats (operated at temperatures above 800°C, test 47-3) contained as much as 10% CF_4 in spite of a pronounced thermodynamic incompatibility between CF_4 and both the fuel and the INOR-8 capsule wall. This amount of CF_4, less than 2 cc at STP, corresponded to a negligible consumption of graphite but represented a calculated loss of fluoride anions from the fuel sufficient to convert about 9% of the UF_4 to UF_3. Two similar capsules, except that the graphite boats had been partially prepermeated with fuel, produced only 10% as much CF_4 from a larger total amount of fuel, perhaps because the graphite-fuel interface was hotter and thus the back reaction was facilitated. To date, efforts to detect or identify reduced species in the fuel samples have been unsuccessful.

Reactor Processing. – Incorporation of the H_2-HF treatment for oxide removal from fuel or flush salt into the fluorination system was given a preliminary examination. The equipment planned for the fuel-storage cell will require only minor modifications and additions. The 400°C sodium fluoride trap for niobium removal during fluorination will serve a similar purpose during the H_2-HF treatment, when considerable NbF_5 should be evolved. Corrosivity of condensed HF-H_2O azeotrope will require that the temperature be kept above 112°C, instead of the 60°C normally maintained to prevent UF_6 condensation, until the off-gas is neutralized. Neutralization will require a caustic scrubber. This scrubber could replace the activated-charcoal trap for iodine removal during fluorination. If analysis of the off-gas is required as a check on oxide removal, it may be necessary to condense the excess HF and sample for water content before neutralization and disposal to the waste system.

STUDIES AND EVALUATIONS

Reactor Evaluation Studies. – The fabrication cost equations developed by Stein *et al.* (Y-1368) have been used to calculate fuel element fabrication costs as a function of cladding material, fuel-element design, and fissile material. Of particular interest was the fabrication cost of recycle fuels relative to that of enriched uranium fuels for the following reactor types: Dresden, Yankee, Candu, GCR-2, and Hallam. The need of glove boxes during parts of the fabrication process increased fuel costs by about $9/kg for Dresden-type elements and about $21/kg for Yankee-type elements (direct fabrication without the need of glove boxes was considered the standard method). Remote fabrication with the need of glove boxes and

heavy shielding increased fabrication costs by about $56/kg for Dresden-type elements and about $134/kg for Yankee-type elements. The costs obtained were generally lower than those used in previously reported fuel value studies; this was due to the combined effects associated with different ground rules (large production rates and continuous processes were considered in Y-1368) and with a more thorough evaluation of costs.

Nuclear fuel costs have been calculated for civil defense reactors operating at 100, 300, and 500 Mw (electrical); the reactors considered were of the Yankee, Dresden, GCR-2, Candu, Hallam, and Bodega Bay type. Of the reactor types studied, the lowest fuel costs were associated with the advanced boiling-water reactor of the proposed Bodega Bay type; at 330 and 100 Mw (electrical), respectively, the fuel costs were 2.3 and 2.5 mills/kwhr at 4-3/4% fuel inventory charge and 2.7 and 2.9 mills/kwhr at 12% inventory charge.

ADVANCED REACTOR DEVELOPMENT

Advanced Test Reactor Corrosion Program

To expedite the collection of data on the effect of heat flux on the corrosion of aluminum, a second test facility, including stainless steel pump loop, power source, and instrumentation, was constructed. Tests in the new facility showed that oxide accumulated on the aluminum specimen at rates lower than those for the same type of specimens exposed under identical conditions in the old test facility (the one used to conduct the HFIR tests). Examination of the specimens from the new facility revealed a thin layer of an iron-rich deposit on top of the boehmite. Such a deposit was never observed on the specimens from the old facility.

Thus it seemed possible that some component removed by the water from the new stainless steel was deposited on the boehmite and that this film was responsible for the lower rate of oxide formation in the new loop. To investigate this possibility, the new loop without an aluminum specimen was operated for four days with deionized water at 275°C to develop a thin corrosion-product film on all stainless steel surfaces, and a bed of new stainless steel turnings was placed in the old loop. Identical tests were then run in the two loops. Results of these tests showed that the rate of oxide accumulation on the specimen exposed in the new loop after pretreatment was greater than it had been and that the specimen exposed in the old loop with the stainless steel turnings present was significantly lower than it had previously been. Examination of the specimens is incomplete at this time, and other tests are in progress to verify the above observation.

High Flux Isotope Reactor Design and Development

Physics. — Recent calculations made in connection with the HFIR critical experiments show considerably better agreement between the experimental and calculated power distribution than previously reported. The differences in the two calculations resulted from a change in the dimensions of

the fuel annuli, the latest dimensions being more representative of the critical experiment. By comparison with the experimental values, the calculated values of local power density in the inner annulus are now −4% at the inside edge, +6% (maximum) at about midposition, and −3% at the outer edge. In the outer annulus the corresponding values are −5% at the inside edge, +2% (maximum) about one-quarter of the way across the annulus, and −30% at the outer edge. The comparatively large difference at the outer edge is largely attributed to the inability of the one-dimensional calculation to take into consideration the localized window-peaking effect that resulted from the particular position of the control plates. This will be investigated further with a two-dimensional calculation.

The most significant dimensional change made in the above calculated cores was in the thickness of the water-aluminum annulus between the two fuel annuli. The power distribution in the inner annulus appears to be very sensitive to this dimension, and the effects are being investigated in greater detail.

The reactivity worth of the fuel in the critical experiment has been estimated by withdrawing the control rods to the normal fully withdrawn position, by adding a solution of boron to the coolant water in the fuel region to maintain criticality, and then by calculating the worth of the boron. The result was $\Delta k = 0.117$. Through the use of this value and fuel-cycle calculations it was estimated that under the most adverse conditions (most poisonous conditions in target and reflector) the fuel cycle would be about 6 days. Under the most favorable conditions the cycle would be about 13 days. Conditions that corresponded to the previously reported 14-day cycle resulted in an 11-day cycle.

A possibly significant unknown in the above fuel-cycle calculations is the high-cross-section impurity content of the aluminum and beryllium used in the critical experiment. By use of concentrations found in vendors' specifications (usually upper limits), it was estimated that boron and cadmium burnup would result in a one- to two-day increase in the above estimated fuel cycles.

In view of the apparent shorter-than-desired fuel cycles it was decided to investigate the possibility of adding sufficient fuel for a five-day extension. This will take about 1.4 kg of additional fuel, making a total loading of 9.4 kg.

To compensate for the reactivity worth of the added fuel, more burnable poison will be added; and since the use of more boron will further jeopardize the fuel cycle, it is proposed to use a mixture of boron and cadmium in the contoured filler pieces of the inner-annulus fuel plates. (This is in contrast to the previously specified uniform distribution of boron in both filler and meat.) This should result in the maximum worth and greatest burnup of the burnable poison and in the flattest fuel distribution. Preliminary calculations indicate that this type of core will have a 20% flatter fuel distribution in the inner annulus and will have a 6% smaller peak thermal flux in the island at time zero. However, rapid burnup of the cadmium will quickly diminish the 6% deficiency and later in the cycle should result in an even greater peak flux than achieved in the 8.0-kg core.

Metallurgy. – Fabrication of an outer-annulus element containing composite fuel plates for use in the HFIR core mockup is in progress. Of 120 additional fuel plates processed through a revised manufacturing schedule, 84% were found to be completely blister-free. Excellent control of the fuel plate thickness (±0.2 mil) was realized, which should promote improved dimensional control when these plates are being curved to the involute shape.

Welding studies are in progress to establish parameters for an all-welded element comprised of two concentric tubes between which curved fuel plates are positioned.

Design. – The hot-spot analysis procedure has been expanded to include the effects of fuel plate deflections and oxide buildup on calculated burnout power level. In analyzing the hot-spot conditions it is assumed that the hot spot occurs at the core outlet, at the point of maximum pressure-induced deflection. All the hot-spot factors were assumed to apply simultaneously at the hot spot, including heat-flux-increase factors resulting from the presence of a 1/8-in.-diam unbonded area. Recommended heat-transfer-coefficient and burnout-heat-flux correlations were used (about 95% of the experimental data above the correlation curve). The results indicate that for reactor operation at 900 psi with a 73.5-psi core pressure drop the calculated burnout power level at the start of a fuel cycle is 165 Mw for a core with coolant-channel thickness tolerances of ±6 mils average and ±10 mils maximum local thickness variation. After 15 days of operation at 100 Mw, the buildup of oxide film causes a reduction in calculated burnout power level to 150 Mw. The end-of-cycle local heat-flux burnout margin is 1.84, assuming a 1/8-in.-diam blister at the hot spot. With no unbonded area at the hot spot, the local heat-flux burnout margin is 2.5.

A core with local coolant-channel thickness tolerances of ±6 mils average and ±10 mils maximum is believed to be an attainable goal. On the basis of these calculations and in view of the conservative assumptions, uncertainty factors, and heat transfer correlations employed, these tolerances appear to be satisfactory.

NUCLEAR TECHNOLOGY AND GENERAL SUPPORT

Basic Reactor Physics. – The inception of a precise measurement of the value of α, the ratio of the capture cross section to the fission cross section, for U^{233}, U^{235}, and Pu^{239}, over a wide range of energies was noted previously. The design of the experiment and the associated equipment were discussed in ORNL-3193 (p 177).

A preliminary run, demonstrating the practicality of the experiment and testing the performance of the equipment, was completed. However, since the U^{235}-coated plates being used in the special fission chamber are known to be inadequate because of coating inhomogeneities, the data now being analyzed are not expected to be particularly good. In addition, this run utilized only 1-μsec neutron pulses. Later runs will employ 8-μsec pulses, as well as improved plates for the fission chamber.

Basic Reactor Shielding. — The model IV gamma-ray spectrometer of the Bulk Shielding Facility is being used in measurement of the gamma-ray spectrum of the stainless steel core of the Bulk Shielding Reactor II. Pulse-height spectra are being obtained at distances of 1, 5, 10, 15, and 20 cm, normal to the reactor. Later measurements will include spectra at various angles from the normal to the core.

An IBM 7090 code has been written to process the spectral data being obtained. This code takes the information from a 256-channel pulse-height analyzer (in punched-tape form), examines it for consistency, and reduces the data from its original form of counts per channel per second vs channel number to counts per Mev per second vs energy (Mev), if given appropriate calibration curves derived from known monoenergetic sources.

Nondestructive Test Development. — A program is in progress to develop techniques for the measurement of the fuel density in cylindrical fuel rods. At present, the preliminary studies are restricted to density determinations of vibratory-compacted fuel rods.

A principal part of the technique is the measurement of the attenuation of gamma rays transmitted through the specimen. The gamma rays supplied from 1 curie of Co^{60} are collimated to a 1/8-in.-diam beam through 10 in. of lead. The detection and recording system consists of an NaI(Tl) crystal, a photomultiplier tube, a high-voltage supply, and a modified Brown recorder with a 21-mv suppression circuit. The rods are driven longitudinally through the gamma-ray beam, and the variations of attenuation are recorded as a function of rod length.

The rod-cladding materials to be studied include aluminum- and zirconium-base alloys and austenitic stainless steel. Rod sizes will vary from 1/4 to 3/4 in. in outside diameter, with wall thicknesses in the range from 0.005 to 0.040 in. The types of fuel to be investigated are ThO_2, UO_2, UC, and UN, with compaction resulting in densities 80 to 95% of theoretical.

With the limited number of fuel rods thus far examined, the sensitivity of the measurements for a given density change is less than 1%. The calibration will be more accurate after density standards have been fabricated.

This system should prove applicable to the evaluation of fuel rods manufactured by vibratory compaction, swaging, or other methods which are proposed for the BNL kilorod, the transuranium, and other fuel-cycle programs.

Mechanical Properties Research. — Previous examinations of type 304 stainless steel specimens exposed to flowing CO_2 at elevated temperatures showed that carburization had occurred. In order to allow more detailed studies of this reaction, an experimental facility has been set up which utilizes CO_2 in which the carbon atoms are tagged as C^{14}. A pump to circulate the CO_2 was developed and has been described in detail (ORNL TM-75). The facility has operated successfully for approximately 6000 hr of test time. A suitable autoradiographic procedure for determining the distribution of carbon has been developed, although refinements are being made constantly.

Several significant observations have been made concerning the reaction of CO_2 with type 304 stainless steel:

1. Carburization has been observed to occur during exposure to flowing CO_2 (pressure ≅ 300 mm Hg) over the range 1350 to 1700°F.

2. In specimens exposed to CO_2 for about 50 hr at 1350°F, large carbon gradients were observed between the oxide-metal interface and the center of the specimens. Only very slight gradients were observed in specimens exposed for 50 hr at 1500°F and above.

3. Slight carbon gradients have been observed between grain boundaries and bulk grains.

These studies have been expanded to include the carburization of beryllium in CO_2 atmospheres. Work in this area is being hampered by the reactivity of the autoradiographic emulsion developing solutions with the beryllium. Protective coatings of 0.00025-in.-thick Mylar sheet are being used to protect the beryllium.

Several beryllium samples have been exposed to $C^{14}O_2$ over the range 1100 to 1500°F and are ready for examination.

Zirconium Metallurgy. — Zirconium-niobium alloys containing 1 to 5 wt % Nb and 0.1 to 0.5 wt % ternary addition of other elements are of considerable interest for steam and water service at temperatures too high for Zircaloy-2 because of corrosion resistance requirements or strength requirements at operating temperature, or both. In recent studies, electrical resistivity and hardness measurements on a series of zirconium-niobium alloys have confirmed the solubility curve of niobium in α-zirconium (5 to 6 wt % Nb maximum at 610°C) reported by Rodgers and Adkins, in contrast to the more recent data reported by the Denver Research Institute and Chalk River of less than 0.5 and 0.1 wt % Nb. Microstructural examination of the specimens has confirmed the electrical resistivity and hardness data except for specimens heat treated in the region between 600 and 650°C. In this temperature range, the microstructures of the specimens present an anomaly which has not yet been resolved.

Solid Reaction Studies. — Preliminary experiments have been completed on determining the diffusion coefficients of Nb^{95} in pressed and sintered UO_2 pellets. The experiments were performed by depositing a very thin layer of isotope onto an optically flat surface of the UO_2, subjecting the specimen to a diffusion heat treatment, removing several thin sections parallel to the optically flat surface, determining section thicknesses by use of an electronic comparator, counting the Nb^{95} activity in each section removed, and relating the resulting penetration profiles to the diffusion coefficient by use of the appropriate solution to Fick's second law. Results indicate that at temperatures near 1500°C the bulk diffusion coefficients of Nb^{95} in UO_2 are about four orders of magnitude larger than reported values for diffusion of uranium in UO_2. Future experiments will be performed to expand the temperature range for diffusion of Nb^{95} in UO_2 and to determine diffusion parameters for other cations in UO_2.

Materials Compatibility Studies. — In studies of the Nb-O-K and (Nb–1% Zr)-O-K systems, significant pickup of oxygen in Nb–1% Zr alloy was noted after exposure to potassium containing 400, 1400, and 2400 ppm O_2 (butyl bromide analysis) at 1500°F for 500 hr. Under similar conditions only minor oxygen changes were noted in niobium. No significant weight changes or metallographic evidence of corrosion was found in either Nb or Nb–1% Zr. Re-examination of an earlier test using Nb–1% Zr specimens

contaminated with 150 to 3600 ppm O_2 and exposed to potassium at 1500°F has led to the calculation of a distribution coefficient

$$K_{1500°F} = \frac{C_{\text{oxygen in Nb-1\% Zr}}}{C_{\text{oxygen in K}}}$$

of approximately 3 to 4 for oxygen in this system.

Previous studies have demonstrated that the presence of small amounts of oxygen in niobium causes a decrease in its corrosion resistance to lithium. Studies of this system have continued, and the following is a summary of the results that have been obtained thus far:

1. Corrosion of niobium by lithium increases with increasing oxygen concentration in the solid metal.
2. At 1500°F, the critical oxygen concentration for corrosion to occur is 500 ppm.
3. Oxygen concentration of the lithium does not affect the corrosion resistance of uncontaminated niobium.
4. Both grain-boundary and transgranular attack occur.
5. Grain-boundary attack is favored, but as grain size increases transgranular penetration increases.
6. Transgranular penetration occurs along a crystallographic plane, tentatively identified as (110).
7. Rate of attack is very rapid; heavy attack has been observed after 0.1 hr at 1500°F and after 0.5 hr at 400°F (melting point of lithium is 356°F).
8. Morphology of corrosion product and total depth of penetration are functions of the test temperature.
9. In addition to lithium penetration, transfer of oxygen from the solid metal to the liquid metal has been observed.

Fuel Element Development. — Liquid-phase sintering is being studied as a means for solving the severe problems associated with UC fabrication. Processes have been found for substantially lowering the fabrication temperature (by use of liquid-phase sintering) while concomitantly giving structures free of the eutectic constituent (all-UC products). One technique for accomplishing this employs the vaporization principle. With UAl_2 as the liquid-promoting agent, aluminum is volatilized while the uranium is combined with carbon:

$$12UC_{(1+x)} + UAl_2 \rightarrow (12 - y)UC + (1 + y)\text{liq}(U,Al,C) \rightarrow 2Al\uparrow + 13UC \ .$$

Compared with a sintering temperature of 2000°C for achieving acceptable bulk density in unaided fabrication of UC, densities as high as 94% of theoretical have been achieved with liquid-phase sintering at temperatures as low as 1350°C. As little as 3% of the original aluminum remains after fabrication.

Reactor Controls Development. — Experimental investigation of multiple servo systems (described in ORNL CF-61-1-82) was extended to continuous systems as well as the on-off controllers previously studied (ORNL-3174). In this approach, a multiplicity of identical servo systems (at

least three) have their outputs added algebraically. Failure of any one servo will not interrupt the process, since the others will always be able to override it. When one of three continuous servos in use fails, an error equal to half the error needed to produce maximum (saturated) output will be produced in the controlled variable. Whether this error is significant depends on the servo gain; higher gain gives less error. On the other hand, the allowable gain is limited by stability considerations. A model of a reactor with three controllers was studied by using an analog computer, and the maximum acceptable gain was determined for a variety of reactor and controller parameters. For the examples considered, the maximum error in reactor power was less than 5%, which is acceptable for most plants.

The effect of a negative temperature coefficient of reactivity is to increase the allowable servo gain. In the example studied, a temperature coefficient as large as 2×10^{-4} $\delta k/°F$ changed the allowable gain, and thus the maximum error, by less than a factor of 2.

Power Reactor Fuel Processing: Corrosion. – Unwelded Corronel 230 exposed to boiling 20 M HNO_3 for 384 hr was corroded at rates of 7.3, 0.88, and 0.62 mils/month in the vapor, interface, and solution phases respectively. Vapor-zone specimens appeared to have undergone intergranular attack. In boiling 10 M HNO_3–0.5 M NaF–0.75 M $Al(NO_3)_3$, unwelded Corronel 230 was corroded at rates of 0.82, 0.78, and 0.81 mil/month in vapor, interface, and solution, respectively, in 240 hr exposure. Corresponding rates for welded titanium 45A were 0.04, 0.20, and 0.34 mil/month. Unwelded Corronel 230 was corroded at rates of 3.70, 12.3, and 25.2 mils/month in 48 hr exposure to vapor, interface, and solution, respectively, of refluxing 1 M HF. These data will serve as a base line for corrosion tests in flowing HF-H_2O_2 solutions.

In 24 hr, corrosion of Haynes alloy No. 25 in chlorine increased from about 6 mils/month at 500°C to 1290 mils/month at 730°C; corresponding rates for Nichrome V were 2.4 and 650 mils/month.

Power Reactor Fuel Processing: Solvent Extraction Studies. – In studies on flowsheets of the Purex and Interim-23 type for removing Th^{228} and U^{233} containing ~100 ppm of U^{232}, uranium was decontaminated from thorium by a factor of ~100 and met U^{233} product specifications. In the tests, synthetic feed solutions were extracted with (1) 30% tributyl phosphate in Amsco and scrubbed with aluminum nitrate or ammonium nitrate containing fluoride and (2) 2.5% TBP and scrubbed with either a static or a countercurrent aluminum nitrate solution. Uranium holdup in the static scrub was, however, too high for practical application.

The soluble organic materials resulting from leaching of unirradiated UO_2-graphite Pebble-Bed Reactor fuel with 90% HNO_3 had no apparent deleterious effect on uranium extraction with TBP, decontamination, or stripping. Fission products had been added to the 90% HNO_3 dissolvent prior to the leaching. Excess acid was removed by evaporation, and the uranium was extracted with 5% TBP in Amsco and stripped with 0.01 N HNO_3. Uranium loss to the aqueous raffinate was 0.3%, and to the stripped solvent, <0.01%. The gross decontamination factor was 6000.

A column 27 cm long by 8 mm in diameter containing 80- to 100-mesh unfired Vycor glass removed 96.6% of the protactinium and 0.01% of the thorium from a solution containing 116 g of Th per liter, 6.2 N HNO_3, and

3.76×10^6 counts min^{-1} ml^{-1} of Pa^{233}. A total of 250 ml of this solution was passed through the column at an average flow rate of 1.08 ml/min; protactinium adsorption was constant throughout the run. The protactinium was eluted with 0.5 M oxalic acid, 80% in the first 25 ml of eluant at a flow rate of 0.5 ml/min, and only 4% in the next 25 ml. Qualitative tests indicated that the elution can be improved by increasing the residence time of the eluate in the column. Preliminary experiments gave no indication of the ultimate capacity of the unfired Vycor for protactinium. The maximum adsorption measured was 6 μg of Pa^{233} (120 mc) per gram of glass.

Power Reactor Fuel Processing: Graphite-Base and Uranium Carbide Fuels. — Combustion of UO_2-graphite fuels containing iron as an impurity produces an ash from which uranium cannot be completely leached with boiling nitric acid. The amount of uranium lost to the acid-insoluble residue decreased from 2 to 0.1% as the Fe/U mole ratio in the fuel decreased from 2.4 to 0.09.

Type GBF graphite reacted slowly with boiling 90% HNO_3, only 60 to 70% being oxidized in 200 to 500 hr. The chief products were CO_2 and CO, although 3 to 6% of the carbon was converted to water-soluble organic acids, mainly (60 to 80%) mellitic acid. No evidence for nitration of the soluble organic products was found.

Specimens of $UC_{1.5}$, $UC_{1.6}$, and $UC_{1.7}$ that had not been heat-treated yielded less methane on hydrolysis than did UC. Greater quantities of water-soluble organic species and about the same amounts of C_2 to C_8 hydrocarbons were also observed with increasing carbon concentration in the specimens.

Power Reactor Fuel Processing: Darex Process. — In further development work on countercurrent stripping of chloride from Darex dissolver solutions with N_2O_4-NO_2 equilibrium mixtures at 45 to 55°C in a 2-in.-diam column 6 ft high packed with 1/4-in. Berl saddles, chloride was removed to < 350 ppm as compared with ~3000 ppm in a 3-ft-high column with 500 to 600% excess NO_2. Extrapolation of the data indicates that chloride can probably be removed to < 350 ppm with 200% excess NO_2 in a column 12 to 24 ft high.

In 1-liter laboratory-scale runs with a batch dissolver and a continuous, titanium-ring-packed, chloride stripper column, Darex dissolution of APPR fuel yielded 1.7 wt % gelatinous precipitate, predominantly SiO_2, which clung to vessel walls and readily plugged the stripper column. The addition of 0.01 to 0.04 M fluoboric acid to the dissolvent solubilized the silica when 24 g of stainless steel per liter was dissolved. At higher fuel loadings, 24 to 60 g of stainless steel per liter, SiO_2 solubility decreased sharply, even though the F/Si mole ratio was constant, but as little as 0.01 M HBF_4 produced a granular, rapidly settling precipitate. About 87% of this material passed through the system; the remainder, which was held up in the stripper column, was easily washed out with hot dilute HNO_3. About 11% of the fluoride appeared in the overhead condensate (0.015 M F) when the dissolvent contained 0.02 M HBF_4.

Fluoride Volatility Pilot Plant. — In two runs, TU-9 and -10, 40-kg charges of nonirradiated fully enriched zirconium-uranium alloy fuel were processed with HF at flow rates of 150 and 180 g/min respectively. Dissolution rates for the two runs were essentially equal, 2.9 and 2.2 kg/hr

for 90 and 100% completion respectively. Corresponding HF utilization efficiencies were 16.6 and 21.1%, for total dissolution times of 20.0 and 18.8 hr. Nonrecoverable uranium losses were 1.36 and 0.89 g.

Although initial uranium concentrations in the fluorinator feed salt were identical, fluorination of the TU-9 salt to < 10 ppm U required only 100 min at a fluorine flow rate of 6 std liters/min compared with 123 min at 6 and 13 to 18 std liters/min in run TU-10. Temperature-zoned absorber performance was satisfactory in both runs. Waste-salt recoveries for both runs were 100 ± 5%. A complete uranium balance was not made due to system holdup. At the end of the normal dissolution, 4 hr of additional HF flow at 40 g/min was used to solubilize residual metallics, and visual examination of the vessel after run TU-9 showed essentially clean surfaces.

Run TU-9 was completed without incident. Operational problems in run TU-10 included a bad compressor bearing, a leak in the HF flow control valve, lower-than-normal heated-duct temperatures, and a UF_6 product line plug. The dissolver off-gas system performed well during both runs at recycle HF flow rates of approximately 350 g/min. A cursory visual inspection of the system after run TU-10 showed no massive deposits or accumulations of off-gas solids. The off-gas filter, which showed no pressure-drop increase during either run, has now been in service for six consecutive runs without the Yorkmesh cartridge having been cleaned or discharged.

PHYSICAL RESEARCH PROGRAM

Reactor Operations. — Operating time for the ORR during the quarter ending December 31, 1961, was 74.6%.

In order to provide a high flux in the experiment facility in the north side of the ORR, fuel will be loaded against this side during the February shutdown. A new hydraulic-tube facility will be installed in position E-8 and a boiling detector in position E-9 during this shutdown. The boiling detector is being investigated as a possible safety instrument to give protection against a local cooling deficiency.

Waste Disposal. — The release of particulate radioactivity from the stacks has been further decreased by installation of a filter in a main duct. Part of the monitoring instrumentation for liquid process waste is in operation and is being connected to the central control room.

Several designs are being considered to replace trench No. 6, which developed a leak. The alternatives being considered are a replacement trench, a series of 3-ft-diam seepage holes, and an evaporator to reduce the volume of the intermediate-level waste to a point where trench No. 5 could handle the entire output.

Hot-Cell Operations. — Operations in the hot cells have been routine. Two irradiated APPR fuel elements were received from Fort Belvoir. One unit containing europium suppressors will be disassembled for detailed examination next spring. The other, a welded-type fuel element, was given close visual inspection and is being returned to Fort Belvoir. Several GCR capsules containing samples of UO_2, UC_2 fuel particles coated with pyrolytic carbon, and UC_2 dispersed in graphite were disassembled and examined. Decontamination of cell A was completed following removal of the SRE disassembly equipment.

PHYSICS AND MATHEMATICS RESEARCH

Charge Spectrometry: Determination of Fragment Ions from Decomposition of $(CH_3Xe^{130})^+$. — With the use of a specially designed mass spectrometer, a determination has been made of the fragment ions resulting from the decomposition of $(CH_3Xe^{130})^+$, following the radioactive decay of CH_3I^{130}. By comparing these results with those obtained from a companion study of CH_3I^{131}, it was possible to ascertain the role played by recoil energy, because the recoil from the decay of I^{130} is considerably larger than that from I^{131}. The only notable difference found on comparing the results of the 15 fragments investigated in the study on CH_3I^{130} with the results from CH_3I^{131} is that the intensity of CH_3^+ is 20 times greater in the case of CH_3I^{130}, with a corresponding decrease in the intensity of the parent ion $(CH_3Xe^{130})^+$. This suggests that the principal role of recoil energy in the decay of I^{130} is to sever the C-Xe bond. The excitation necessary to account for the remaining fragments comes from the sudden change in nuclear charge following β^- decay and from multiple Auger processes following internal conversion. From measured values of the more highly charged xenon ions, the percentage of internal conversion

in the decay of I^{130} has been estimated to be 1-1/2%, in agreement with nuclear-chemical data.

Charge Spectrometry: Ratio of Internal Bremsstrahlung to Beta Emission of He^6. – A small, thin-walled, Lucite cylinder was used as an He^6 source volume. The internal bremsstrahlung quanta were detected with an NaI crystal, and the betas were detected with a plastic scintillator. In order to evaluate the relative solid angles subtended by the two counters, measurements were also made with Ne^{23} as the source gas. The precision of the geometry factor thus depends on a knowledge of the branching ratios of Ne^{23}. A first estimate of the internal bremsstrahlung-to-beta ratio is (2.1 ± 0.2)%, which is to be compared with a value of 0.4% obtained by numerical evaluation of the theory as developed by Lewis and Ford [Phys. Rev. 107, 756 (1957)].

Charge Spectrometry: Shape of Internal Bremsstrahlung Spectrum of He^6. – A large Lucite source volume was used for the investigation of the shape of the internal bremsstrahlung spectrum of He^6. The detectors were an NaI crystal and a plastic scintillator, the latter serving as a monitor of variations in the source strength. Several types of runs were made to evaluate corrections for external bremsstrahlung. In order to evaluate effects from this extended source, energy calibrations were made of the internal-bremsstrahlung detector by taking spectra of various gamma-emitting sources, first at the end of the source volume near the NaI crystal and then at the far end; one run was also made with a gaseous source, Ar^{41}.

Charge Spectrometry: Half-Life Determination of He^6. – The determination of the half-life of He^6 was undertaken as one means of examining the source gas for possible contaminants. Preliminary runs indicated agreement with spectral observations that an upper limit could be set of about 0.5% for gamma(N^{16})/internal bremsstrahlung(He^6). Data were recorded with a movie camera which took pictures of a scaler as it accumulated counts from the beta detector while the gas in the source volume decayed, after being isolated from the rest of the system. By combining 30 runs, a good statistical accuracy is expected for a determination of $T_{1/2}$ covering a decay period of about eight half-lives.

Neutron Diffraction. – Neutron diffraction studies have been made by means of the ORNL magnet diffractometer on single-crystal specimens of holmium in order to investigate the mechanism of magnetization in this substance. At each of two temperatures, 64 and 4.2°K, magnetic fields up to 22.3 kilo-oersteds were applied to the specimens at angles of 0, 30, 60, and 90° to one or both of the principal directions a_1 and b_1 in the basal plane, and the distribution of magnetic reflections in the vicinity of important nuclear reflections was studied. At 4.2°K the conical arrangement of moments, which is characteristic of holmium in the virgin state, was transformed in low fields to a ferromagnet in which the moments are parallel to a b-direction. The a-direction is a hard direction for magnetization. At 64°K the application of a magnetic field produced two or three distinct magnetic configurations, depending upon the strength and direction of the field.

High-Voltage Experimental Program: The 3-Mv Pulsed Van de Graaff. — The Van de Graaff, now in routine operation after major alterations, is being used for two experiments: to obtain data for η (U^{235}) at 30 kev; to obtain 30-kev neutron capture gamma-ray spectra for Cu, Cu^{63}, Pt, W, and V.

High-Voltage Experimental Program: Heavy Even-Even Nuclei Bombarded by Fast Neutrons. — It can be shown that the differential cross section for formation of compound nuclei in states ($KI\pi$) may be expressed as follows:

$$\sigma(\theta)_{K\pi} = \frac{\lambda\!\!\!^{-2}}{8}(-1)^{-K-1/2} \sum_{xI\ell} (-1)^{x/2}(2I + 1)T_{\ell I}C^{IIx}_{-KK0}Z(\ell I\ell I,\tfrac{1}{2}x)P_x(\cos\theta) .$$

$$\text{Define } W(KI) = \frac{(-1)^{-K-1/2}}{2} \sum_{x} (-1)^{x/2}C^{IIx}_{-KK0}Z(\ell I\ell I,\tfrac{1}{2}x)P_x(\cos\theta) .$$

With this definition the normalization is such that $\int_{-1}^{+1} W(KI)d(\cos\theta) = 1$, and the above cross section may be simply written:

$$\sigma(\theta)_{K\pi} = \frac{\lambda\!\!\!^{-2}}{4} \sum_{\ell I} (2I + 1)T_{\ell I}W(KI) ,$$

where I is the angular momentum of the compound nucleus, and K is the component of I along the major axis of symmetry. For even parity, π, only even values of ℓ are allowed, and the sum over I extends over all possible values consistent with the angular momentum of the system. The cross section then refers to the probability of formation of a given state for an assumed value of K, and the angle refers to the spatial distribution of the major axis relative to the neutron beam axis. To illustrate the simplicity and utility of this expression, three bands of particular interest in relation to fission-fragment angular distributions are:

$$\sigma_{1/2^+}(\theta) = \frac{\lambda\!\!\!^{-2}}{2}\left[T_0W\left(\tfrac{1}{2}\,\tfrac{1}{2}\right) + 2T_2^-W\left(\tfrac{1}{2}\,\tfrac{3}{2}\right) + 3T_2^+W\left(\tfrac{1}{2}\,\tfrac{5}{2}\right) \ldots\right] ,$$

$$\sigma_{1/2^-}(\theta) = \frac{\lambda\!\!\!^{-2}}{2}\left[T_1^-W\left(\tfrac{1}{2}\,\tfrac{1}{2}\right) + 2T_1^+W\left(\tfrac{1}{2}\,\tfrac{3}{2}\right) + 3T_3^-W\left(\tfrac{1}{2}\,\tfrac{5}{2}\right) + 4T_3^+W\left(\tfrac{1}{2}\,\tfrac{7}{2}\right) \ldots\right] ,$$

$$\sigma_{3/2^-}(\theta) = \frac{\lambda\!\!\!^{-2}}{2}\left[2T_1^+W\left(\tfrac{3}{2}\,\tfrac{3}{2}\right) + 3T_3^-W\left(\tfrac{3}{2}\,\tfrac{5}{2}\right) + 4T_3^+W\left(\tfrac{3}{2}\,\tfrac{7}{2}\right) \ldots\right] .$$

Others may be written by inspection. The coefficients of the W(KI) represent directly the strengths of the (KI) levels, provided that no two bands of like parity can compete. When such competition does exist, the coefficients indicate the combined strengths of the levels with like ($I\pi$). The reaction cross section is

$$\sigma_r = 2\pi\left[\sigma_{1/2^+}(\theta) + \sigma_{1/2^-}(\theta)\right] .$$

These relations are being used in analyses of fission-fragment angular distributions; a table of the W(KI) for K and I up to 7/2 as functions of θ has been prepared.

CHEMISTRY RESEARCH

Effects of Radiation on Analytical Methods. – Various studies have been continued for the purpose of detecting and evaluating the effects of gamma radiation on methods of chemical analysis, with particular consideration of methods involving the use of ethylenediaminetetraacetate (EDTA). Spectral studies were made which revealed that the infrared absorption spectra of gamma-irradiated solutions of EDTA differ significantly from the spectrum of nonirradiated solutions. These differences are being analyzed and evaluated.

The rate of change of EDTA concentration as a function of irradiation time and intensity and the pH dependence of the sensitivity of EDTA to gamma irradiation were also investigated. Gamma radiation was observed to decrease the chelating power of an EDTA solution. The rate of degradation, based on the loss of chelating capacity, was found at all concentration levels to be approximately 1 picomole (10^{-12} mole) ml^{-1} r^{-1} and essentially independent of the H^+ concentration over the pH range from 1 to 11. The slope of curves obtained in the amperometric titration of EDTA with VO^{2+} was observed, however, to be dependent on the H^+ concentration of the EDTA solution at the time it was irradiated. For solutions irradiated at a low pH, the initial current was much greater and the end point less well defined than for EDTA solutions irradiated at higher pH values. Low pH favors the formation of reducible ionic species; whether these species are derived from degradation of EDTA or from other sources is, at present, unknown.

It was also established experimentally that, in general, gamma radiation at a level of about 10^5 r is required for destroying the chromophoric groups of various dyes used as end-point indicators in titrations with EDTA.

Mechanisms of Separations Processes. – Simulated ORNL low-activity-level waste, containing 3.3 ppm of Ca^{2+}, 10^{-2} M NaOH (pH 11.8), and tracer Sr^{89}-Sr^{90}-Y^{90}, was decontaminated from the strontium in a 6-in.-ID countercurrent foam column. Decontamination factors, which were 500 to 5000 with aqueous flow rates of 8 to 18 gal ft^{-2} hr^{-1} and (3.8 or 7.6) $\times 10^{-4}$ M sodium dodecylbenzene sulfonate surfactant, decreased with increasing flow rate, which should be maximized, and with increasing surfactant concentration, which should be minimized. Analytical and graphical methods were derived for correlating decontamination factors, single-stage separation factors, aqueous throughput, and volume reduction in a countercurrent foam separation column and are being used to calculate optimum decontaminating conditions.

The heights of transfer units for either isotopic exchange or stripping of strontium were determined in the above column for 7×10^{-4} M sodium dodecylbenzene sulfonate, 10^{-3} M NaOH, about 10^{-6} M $Sr(OH)_2$ with Sr^{89} tracer, and liquid feed rates of 20 to 40 gal ft^{-2} hr^{-1}. Values based on

the liquid phase were 3 to 4 in. for 18 in. of foam in the column, when air was introduced through a porous stainless steel gas sparger, and 1 to 3 in. for 24 in. of foam and 1 to 2 in. for 4 to 11 in. of foam, with a spinnerette sparger consisting of seventeen hundred ninety-four 50-μ-diam holes. The difference is due to foam condensation and channeling with the sintered sparger caused by relatively irregular bubble size. Strontium concentration variations from top to bottom in the liquid and foam phases were factors of 1.6 to 8 in the isotopic exchange runs, compared with 250 to 500 in the stripping runs.

In extension to acid concentrations > 5 M in an earlier study on the extraction of nitric acid by tributyl phosphate—diluent solutions, acid activities measured at 25°C by a transpiration technique in the range 5 to 15 M showed good precision.

Volatility Studies. — Laboratory studies indicated that pure fluorine in place of gaseous HF is not feasible for the dissolution of zirconium-uranium alloy fuel in fused salts. In 36-32-32 mole % $LiF-NaF-ZrF_4$ at 500°C, Zircaloy-2 dissolution rates were 0.5 to 1.5 mils/hr with high-purity fluorine in a 1-in.-diam reactor at a flow rate of 100 ml/min. However, when 30% HF was used with the F_2, or the salt was presaturated with HF, dissolution rates were 10 to 11 mils/hr. The addition of ~1000 ppm $FeCl_3$ at 500, 600, and 700°C, respectively, increased the rates with no HF and with high-purity fluorine to 5, 6.5, and 8.5 mils/hr. Attempts to promote the direct gas-phase reaction between fluorine and metal failed, even at 700°C. Ignition of Zircaloy-2 with fluorine resulted in some melting only when the metal sample was incompletely submerged in the fused salt.

The addition of gaseous HCl to the HF did not increase the dissolution of Zircaloy-2 fuel in fused 36-32-32 mole % $LiF-NaF-ZrF_4$ at 500°C. In individual tests in a 1-in.-diam reactor with 100, 100, and 0 ml/min of HF mixed, respectively, with 0, 100, and 100 ml/min of HCl, the rates were 10.5, 11, and 2 mils/hr. These data indicate that the high reactivity of HCl for zirconium alloy (Zircex-type process) is hindered by conversion of the volatile $ZrCl_4$ to the nonvolatile ZrF_4.

Processing of graphite—10% UO_2 fuels by burning them with fluorine in fused salt does not appear attractive on the basis of one test. A small explosion occurred in a small amount of residue left after a run in which the dissolution rate in 36-32-32 mole % $LiF-NaF-ZrF_4$ salt at 500°C was 48 mils/hr with fluorine at a flow rate of 100 ml/min. This dissolution rate includes much material that either disintegrated or reformed as sooty material on the side of the reactor.

In further studies zirconium oxide was dissolved in fused salt by HF at rates about twice as great as for zirconium metal under similar conditions. Preliminary chemical analyses of the film formed on partially dissolved oxide indicated that an intermediate oxyfluoride may be formed.

Solvent Extraction Research. — In radioactive cell tests of the chemical flowsheet proposed (ORNL-3211) for final-cycle purification of plutonium from Purex 1BP solution by tertiary amine nitrate extraction, operation was excellent. Plutonium recovery was also excellent, but gamma decontamination factors were 100 to 500, at least 100-fold lower than needed, and than obtained in tests with tracer Zr-Nb and with dissolver-solution mixed fission products. The results strongly indicate that the

Purex extraction product contained a significant concentration of more extractable Zr-Nb species, and feed pretreatment will therefore be required. The flooding capacity of pulsed columns (aqueous-to-organic ratio = 4.1) was found to be > 2400 gal ft^{-2} hr^{-1} for a compound extraction-scrub column with sieve plates (0.125-in.-diam holes, 10% free area), operated with the organic phase continuous at 30 cycles/min. The stripping capacity (organic-to-aqueous ratio = 5, aqueous continuous) was 540 gal ft^{-2} hr^{-1} for the sieve-plate column and 440 gal ft^{-2} hr^{-1} for a nozzle-plate column at 35 cycles/min.

Tracer strontium extraction from 4 M $NaNO_3$ by di(2-ethylhexyl)phosphoric acid and its sodium salt in benzene indicated that $E_{Sr} = [Sr]_{org}/[Sr]_{aq} \propto [H^+]^{-2}$ up to 15% sodium form of the reagent and that E_{Sr} varies as the total reagent concentration at all ratios of sodium form to total reagent. Equilibrium curves (log $[Sr]_{org}$ vs log $[Sr]_{aq}$) with the ratio of sodium form to total reagent constant at levels from 0.1 to 1.0 had slopes of 1 at low $[Sr]_{org}$. The highest organic/Sr ratio varied from 2.4 to 1.6. The data indicate that, at low total strontium and pH, strontium is extracted, releasing two H^+ ions per Sr^{2+} ion and forming a mononuclear compound in the organic phase, and that the organic phase either presents an average of two exchange sites per molecular unit to the strontium or extracts it into a highly polymerized species which remains polymerized.

In further testing of alkylbenzenes for possible use as diluents for organophosphorus extractants, benzenes substituted at two points (diethyl- and diisopropylbenzene and tetrahydronaphthalene) were less stable on exposure to 2 M HNO_3 than the corresponding monoalkylbenzenes. Iso chain branching in the monoalkylbenzenes appeared preferable to normal, secondary, or tertiary.

Chemistry of Technetium. — Three different chloride species of Re(VI) were spectrophotometrically observed when Cl^- was added in small increments to Re(VI) in concentrated H_2SO_4. At low Cl/Re ratios a mixture of the species was obtained, but at high Cl/Re ratios (approx 75) only the rhenium(VI) chloride complex with the highest chloride content was obtained, with an absorption maximum at 4360 A. Analysis of the spectrophotometric data obtained on solutions prepared by addition of excess Cl^- (HCl) to H_2SO_4 solutions (Cl/Re ~ 75) containing rhenium in average oxidation states varying from Re(VI) to pure Re(V) reveals that the Cl^- causes the Re(V) to disproportionate into rhenium(VI) chloride complex and Re(IV): $[Re(VI)\cdot xCl][Re(IV)]/[Re(V)]^2 = 5.6 \pm 3$.

Similarly, three different bromide species of Re(VI) were observed. A study of the effect of Br^- on the disproportionation of Re(V) was not possible in this case because the Br^- causes reduction of Re(VII) in concentrated H_2SO_4. Unlike the chloride complex, the rhenium(VI) bromide complex is formed slowly, the rate of formation from Re(V) being first order with respect to HBr, with an apparent bimolecular rate constant of 4.9 liters $mole^{-1}$ min^{-1}. The complex containing the highest bromide content formed at high Br/Re ratios gives an absorption spectrum with maxima at 5525 and 6475 A. Both the chloride and bromide complexes with the highest halide content are extractable from the H_2SO_4 solution with CCl_4, $CHCl_3$, or n-C_6H_{14}; the species corresponding to lower halide content do not extract.

A violet rhenium species, observed by others, in 6 M H_2SO_4 with a broad absorption maximum at 5450 A and tentatively assumed to be some combination of Re(V) and Re(VI), was obtained by dilution of a concentrated H_2SO_4 solution of Re(V) with H_2O to give a 6 M solution and allowing it to stand for three to four days. Data obtained from a spectrophotometric titration of the violet species with $Ce(SO_4)_2$ are consistent with the hypothesis that an Re(V)·Re(VI) complex is formed which has a molar absorbancy of approximately 15,000 at 5450 A and a dissociation equilibrium constant of approximately 10^{-3}. Most of the rhenium (85 to 90%) is present in the solutions as Re(VII), the violet species representing only a small portion of the total rhenium.

Reactor Neutron Cross Sections. — Microgram quantities of enriched Sm^{150} were irradiated with cadmium filters in the ORR to an integrated flux of 5×10^{19} neutrons/cm^2 (monitored with Co^{59}). The Sm^{151} formed was determined mass spectrographically. A preliminary evaluation of the data yielded a thermal cross section σ_{th} = 120 barns and a resonance integral I = 230 barns for the reaction $Sm^{150}(n,\gamma)Sm^{151}$.

Nuclear Chemistry: Characterization of Short-Lived Fission Products. — Long-lived W^{188} was prepared by successive (n,γ) reactions on enriched W^{186} which was irradiated for three weeks in the LITR. The decay, followed for over three years on a beta proportional counter, showed a half-life of 75.0 days. This is just the value of the W^{185} half-life, and since Lindner has measured the value for W^{188} as 65 days, it must be concluded that the larger fraction of the activity results from the pure β^- emitter W^{185}. However, a value of about 62 days for W^{188} was determined from the gamma-ray decay and is in agreement with the measurement of Lindner.

Fermi analysis of the beta-ray spectrum showed a component at 425 kev which is due to W^{185}, and another at 346 kev which is assigned to W^{188}.

With tungsten sources from which the Re^{188} daughter had been freshly removed, gamma rays at 60, 125, 156, 226, 255, and 286 kev were observed, along with what appeared to be multiple peaks in the regions of 100 and 200 kev. Except for the peaks at 125 and 156 kev and most of the intensity at 60 kev, these gamma rays are assigned to the decay of W^{188}, in agreement with the current work of Burson et al.

A survey of the pertinent literature was made to ascertain the status of data on U^{233} fission product yields. The various experimental determinations were evaluated, and the most recent mass spectrometric results were used as a basis for deriving a set of preferred yields. These yields were compared with values reported in two other recent compilations, and for yields > 1% the three sets agreed with each other to an average precision of < 5%. It was concluded that recent measurements have somewhat improved the reliability of U^{233} fission yield data but that some additional experimental work is needed.

High-Temperature Aqueous Solution Chemistry: Effect of Hydrostatic Pressure on Liquid-Liquid Immiscibility. The System UO_2SO_4-H_2O. — By use of a visual method developed previously (ORNL-3174, p 26), the effect of hydrostatic pressure on the temperature of liquid-liquid immiscibility was determined for 0.14, 0.28, 0.58, 1.22, and 2.60 m stoichiometric UO_2SO_4

solutions in H_2O solvent. Plots of temperature against pressure revealed, at pressures up to 300 atm, and in the region of temperature between 280 and 320°C, linear relationships. The coefficient, $\Delta t/\Delta p$, was approximately +0.080°C/atm for all five concentrations; this was in agreement with previous results on 1.4 m UO_2SO_4.

High-Temperature Aqueous Solution Chemistry: Electrical Conductance at High Temperature and Pressure. – Electrical conductance studies were continued (ORNL-3174, p 26). The conductances of a 0.0005 m K_2SO_4 solution (H_2O solvent) were determined at 25.5, 343, 450, 537, 647, and 746°C at pressures of 1 to 3500 bars. Similarly, conductances of a 0.00218 m K_2SO_4 solution (H_2O solvent) were determined at 259, 375, 520, and 619°C over the same range of pressure. Some comparative, specific conductances of these solutions, obtained by cross-plotting the data, are given in the following table:

Specific Conductances (ohm^{-1} cm^{-1})

Temperature (°C)	K_2SO_4 Concentration at 1000 bars		K_2SO_4 Concentration at 2000 bars		K_2SO_4 Concentration at 3000 bars	
	0.0005 m	0.00218 m	0.0005 m	0.00218 m	0.0005 m	0.00218 m
25		0.667		0.693		0.701
30	0.18					
250	0.726		0.746		0.760	
350	0.700	2.69	0.734	2.88	0.752	2.97
400	0.671	2.46	0.720	2.78	0.743	2.88
500	0.536	1.67	0.677	2.52	0.706	2.73
600	0.260	0.547	0.610	2.16	0.662	2.52
700	0.058	0.190	0.524	1.63	0.605	2.34
800	0.029	0.130	0.418	0.893	0.547	2.07

High-Temperature Aqueous Solution Chemistry: Development of High-Pressure Vessel for Determining Solubilities at Extremes of Temperature and Pressure. – A small high-pressure vessel incorporating a Teflon gasket was developed for use at temperatures up to 500°C and at pressures up to 1000 bars [Rev. Sci. Instr. 32, 1060 (1961)]. The Teflon permitted easy sealing and opening of the vessel. Eight of these vessels were constructed and were used for determination of solubilities of metal sulfate in H_2O at extremes of pressure and temperature.

Chemical Separation of Isotopes. – Single-stage separation factors for both carbon and oxygen were measured for the isotopic exchange between CO_2 gas and a 4.6 M solution of $(CH_3)_2NCOOH$ in benzene. The phases were equilibrated at 23.5°C, and two separate experiments were performed with contacting periods of 9 and 92 hr to ensure the attainment of isotopic equilibrium. The average separation factors are:

$$\frac{(O^{18}/O^{16})_{gas}}{(O^{18}/O^{16})_{liquid}} = 1.0064 , \qquad \frac{(C^{12}/C^{13})_{gas}}{(C^{12}/C^{13})_{liquid}} = 1.0060 .$$

Chemical Applications of Nuclear Explosions. — Hydrogen consumption for the reduction of $MgSO_4$ decreased from 2.8 to 2.6 moles per mole of $MgSO_4$ as the temperature was increased from 734 to 784°C, but for the reduction of $CaSO_4$ the hydrogen consumption was constant at 4.0 moles per mole of $CaSO_4$ from 885 to 915°C. The quantity of SO_2 produced during $MgSO_4$ reduction increased by a factor of 1.5 as the temperature increased from 734 to 784°C. Kinetic studies showed the reduction of $MgSO_4$ and of $CaSO_4$ by H_2 to be autocatalytic.

Samples of NaCl and $MgSO_4$ which had been shocked with high explosive to a few kilobars at Frankford Arsenal were analyzed for chemical decomposition. Determination of the pH and the hydroxyl content of water solutions of the samples showed only 0.01% decomposition of NaCl and no detectable decomposition of $MgSO_4$.

METALLURGY AND MATERIALS RESEARCH

Ceramics Research: Sintering Studies. — The role of plastic deformation in the formation of a weld neck between sintering particles of calcium fluoride is being investigated by means of an etch-pit technique to show dislocations. The plastic deformation of a crystal requires generation and motion of dislocations; therefore a high dislocation concentration may be indicative of plastic flow. Small single crystals of CaF_2 were placed on a cleaved surface of a larger single crystal and the specimens were heat-treated to cause a small degree of sintering. After the small crystals were removed, the larger crystal was etched in sulfamic acid to reveal dislocations. Preliminary experiments have shown a massive concentration of dislocations in the immediate area of neck formation, implying that the mechanism of sintering involves plastic flow, although not necessarily to the exclusion of other processes.

Physical Property Studies. — Thermal-conductivity measurements on 93.4%-dense UO_2 in the radial heat flow apparatus have been extended to 873°C, where a value of 0.0332 w cm^{-1} $(°C)^{-1}$ was obtained (corrected to theoretical density). Spurious emfs in the temperature control circuitry have made operation difficult at the higher temperatures. If this trouble can be removed, measurements can be extended to 1400°C.

The quenching apparatus for measuring thermal diffusivity to 1600°C has been assembled, and a modified specimen support is being tested. Fluidized-bed studies with Al_2O_3 have shown the need for uniform distribution of the incoming gas in the bed.

The total hemispherical emittance of three specimens of an Nb—1% Zr alloy has been measured from 300 to 1200°C. All three specimens oxidized, even at chamber pressures of 6×10^{-6} mm Hg, and this caused unusually high results. Heat loss by the specimen thermocouples is being evaluated on a platinum specimen, and results indicate that the three 5-mil thermocouples remove 10% of the total input power at 90°C and 3% at 830°C.

Conduction losses from the test section have not been measured, but do offer a means of measuring the specimen thermal conductivity if precise specimen dimensions can be determined.

CONTROLLED THERMONUCLEAR RESEARCH

DCX-1 Facility. – Installation of the new high-vacuum liner was completed. After bakeouts at 375°C the base pressure (ion gage) obtained with diffusion pumps, but without Zeolite, titanium, or liquid-nitrogen cooling, was 4×10^{-7} torr. This figure represents an improvement by a factor of 2 over that experienced with the previous liner. Zeolite was added to the inner vacuum region; and additional bakeouts reduced the pressure, under otherwise the same conditions, to 2.5×10^{-7} torr. Cooling the liner and the shields in the intermediate region with liquid nitrogen and depositing titanium in both regions have produced a pressure of 4×10^{-9} torr. Proton containment times of about 20 sec were measured with an injected H_2^+ current of 0.2 ma. Higher currents were not injected at this pressure, but under somewhat different conditions, containment times of 10 sec were measured at 5×10^{-9} torr with 3.0-ma H_2^+. By way of comparison, the lowest pressure obtained with the previous liner was 3.5×10^{-9} torr, at which pressure the containment times were 11 sec with about 2.5-ma H_2^+.

It is not yet clear to what extent the Zeolite has contributed to the improvements in vacuum conditions. These may be primarily the result of the revised geometry of the new liner and the prolonged high-temperature bakeouts.

The Varian mass spectrometer was installed, and data are being accumulated on the composition of the residual gas in the liner region at various phases of the vacuum manipulations.

More studies have been made of the responses of the neutral-particle detector signals during periods of plasma accumulation (buildup) and decay. At base pressure a transition from nearly exponential to nearly linear buildups has been observed to accompany an increase in H_2^+ current from 1 to 5 ma. The linear nature of the buildups at the higher current could gradually be changed to the exponential by raising the pressure slightly. In all cases the decay curves have been nearly exponential.

High-Current Injection. – The principal activity during the last two months was the installation of a beam injector in the DCX-2. Preliminary tests are in progress. Modest current beams have been injected. The source accelerator tube and lens are those used previously in the accelerator-tube test facility.

The first tests of an external epoxy skirt for use with the accelerator tube have been successful. A new tube using this arrangement is being planned.

Theory. – An interpretation has been given to the change of modes from low density to high density observed in the electron cyclotron heating experiment when the neutral pressure surpasses a critical value. Below the critical pressure, the electron leakage rate determines the density. Above the critical pressure, ions begin to be made faster than they

can escape by drifting out of the plasma. Then the ion leakage rate determines the density, which grows until the accompanying electrons sufficiently "burn out" the neutrals to balance ion loss and production. A similar process is thought to account for a rapid growth of secondary plasma in the DCX-1 when the pressure exceeds a critical value of the same magnitude as that observed in the cyclotron heating experiment.

Radio-Frequency Heating. — Recent experiments have been aimed at determining the geometry of the hot electron gas in the mirror geometry device (PTF) mirror field. A differential magnetic Hall probe bridge has been developed which enables measurements of a 100-milligauss/cm diamagnetic gradient in the presence of a 6400-gauss dc mirror field. Recent data obtained with this bridge is in the process of being reduced and will be used by the Magnetics Group for calculation of plasma shape.

Observations on the energetic x rays emanating from the microwave heated plasma indicate a burst character where $\sim 10^7$ photons of the order of 1 Mev are emitted in 0.25 μsec. The amount of hot plasma involved in a burst is relatively small, and the burst rate is correlated with heating-source modulation in a way not yet fully investigated.

The folded cusp facility (ELMO) is near the preliminary testing stage, and the application of 13 cm of heating power to this magnetic field configuration should be possible within the next six weeks. The PTF will then be converted to a much higher power microwave source (10.8 kMc; 5 kw) and rf source (2.7 Mc; 10 kw). Modifications are being included for defining the plasma geometry by means of neutral injection into the desired resonance heating zone. The injection system being fabricated is a type of Laval nozzle.

BIOLOGY AND MEDICINE PROGRAM

SOMATIC EFFECTS OF RADIATION

Pathology and Physiology: Genetic Control of Protein Synthesis. — Strain-specific differences in electrophoretic mobility, solubility, and crystallographic properties of hemoglobin have been used to identify the origin (donor or recipient) of erythrocytes in irradiated mice injected with homologous hematopoietic cells. By these methods, the relative quantities (±5%) of each of two or three types of erythrocytes in mixtures can be determined from a single assay with 0.1 ml of blood. Furthermore, it has been shown that hemoglobin synthesized by transplanted hematopoietic cells retains its strain-specific physicochemical properties unaltered by growth in an irradiated foreign host. Correlation between hemoglobin and serotype, even in chimeras showing graft regression, has indicated that the strain-specific hemoglobin and serotype were both either of donor or of recipient type. The properties of electrophoretic mobility, solubility, and crystalline structure among mouse hemoglobins are controlled by alleles at two loci, Hb and Sol, which segregate independently. Analysis of tryptic hydrolysates of the α and β chain of mouse hemoglobins has revealed that alleles at Hb dictate the molecular composition of the β chain. Although less well documented, the α chain is presumably controlled by alleles at Sol. Hybrid hemoglobin molecules, possessing one

chain (α or β) typical of the donor and the other chain typical of the recipient, have not been found; thus hemoglobin synthesized in erythrocytes of radiation chimeras is presumably controlled by the hematopoietic cell of origin.

RADIATION GENETICS

Mutagen Mechanisms: Bacterial Metabolism. — The possibility that the radioresistance of *Escherichia coli* that develops in glucose-containing media may be attributed to in situ production of a chemical protector has not been eliminated. With the assumption that such a protective agent would be of the sulfhydryl type, attempts were made to destroy such an agent by preirradiation treatment with iodoacetic acid. Incubation of resistant cells with 2×10^{-4} M iodoacetic acid results in a drastic sensitization of the cells. Similar sensitization is also produced in sensitive cells. Unirradiated cells show no toxicity of iodoacetate at concentrations that produce maximum sensitization of irradiated cells. A prerequisite for sensitization is the presence of the compound in the cell suspension during irradiation, since removal of iodoacetate by washing destroys sensitization. Although all the results obtained are consistent with the idea that irradiation produces a toxic by-product of iodoacetate, such a product must be of short half-life, since it cannot be detected in the obvious control experiments with irradiated cells or irradiated iodoacetate.

Mutagen Mechanisms: Bacterial Genetics. — It has been established that radiation response of stationary phase cultures of *Escherichia coli* K12 is under control of a group of genes. Several of these are on the proline-T_6-histidine segment of the linkage map. In recent investigations it has become apparent that these genes do not control the radiation response of logarithmic phase cultures of the same strains. The recombinants derived from crosses of AB312 (♂) and AB331 (♀) are being analyzed in an attempt to understand the genetic basis for the radiation response of logarithmic phase cultures.

Genetic and Cytogenetic Effects of Radiation. — In past experiments, attempts have been made to estimate forward-mutation rates in the ad-3 region of *Neurospora* following x irradiation of wild-type strain 74A. With the "direct method," recovery of all the purple mutants induced is possible where the mutational event leads to a complete (nonleaky) or an incomplete (leaky) adenine requirement. However, the only class of mutants that can be analyzed in experiments with a wild-type strain is that which is homozygous-viable. Demonstration of a class of mutants that behave like recessive lethals in such experiments is difficult, and there seems to be no way to obtain an accurate estimate of what fraction such mutants might constitute of the total number induced. However, with the use of the heterokaryon between an ad-3A-ad-3B double mutant (hist-2 ad-3A ad-3B nic-2 al-2$^+$; inos; pan-2$^+$) and "wild type" (hist-2$^+$ ad-3A$^+$ ad-3B$^+$ nic-2$^+$ al-2; inos$^+$; pan-2$^+$), a method is being developed that will allow such an estimation by permitting recovery and analysis of both the viable mutations and the recessive lethal mutations. Preliminary data from such

experiments have already shown that previous estimates of the forward-mutation rates in the ad-3 region are too low, since a substantial proportion of the mutants recovered with this technique are not viable and behave as recessive lethal mutations.

COMBATING DETRIMENTAL EFFECTS OF RADIATION

Mammalian Recovery: General. — Previous work by other investigators has shown that cells from mouse lymph nodes can be transplanted to an irradiated recipient mouse, where they can function immunologically either in homograft reactions or in production of circulating antibodies. The capacity of transplanted lymph-node cells to differentiate into erythrocytes and granulocytes is being studied in irradiated recipients. Cell doses of 50 to 200 million per mouse have been given intravenously in three experiments, and Fe^{59} uptake has been used as a measure of erythropoiesis. Iron uptake, even at a dose of 200 million cells, was only slightly greater in experimental animals than in irradiated, untreated controls. No granulopoiesis and only rare foci of erythropoiesis were seen in the spleen red pulp of the experimental mice in one experiment, whereas no erythropoiesis or granulopoiesis was seen in any tissues of radiation controls. In contrast to tissues of controls ten days after irradiation, the transfused mice had germinal centers and lymphocytes in lymph nodes and spleen white pulp. Many Ab-forming cells and some plasma cells were seen, and they are thought to be derived from donor lymph-node cells.

Reinvestigation of the chromosomes of seven of the eight persons involved in the Y-12 criticality accident has been completed. The samples were taken 3-1/2 years after the accident. The men still show aberrations of both chromosome number and chromosome form in their peripheral leukocytes, although with a somewhat lower frequency than that observed in 2-1/2-year samples.

Radiation Immunology. — Studies on the kinetics of antibody production by secondarily stimulated spleen cells were completed. Semilogarithmic plots of circulating antibody titer vs time result in curves which are typical of cellular multiplication (e.g., bacterial cultures). By increasing either the amount of stimulating antigen or the number of spleen cells (cultured in the isologous, irradiated animal), the period of adjustment (lag phase) is decreased. The slope of the exponential increase in antibody is the same regardless of the amount of antigen or the number of cultured spleen cells. The maximum amount of antibody produced (reflected by the retardation phase of antibody production) is determined by the amount of antigen supplied, but the decrease in rate of antibody production is not merely a matter of antigen exhaustion. With these results for reference, it is now possible to conduct a series of experiments which will provide clearer understanding of the mechanism of antibody production, particularly of the nature of the adjustment and retardation phases.

When mouse spleen cells are transferred into in vivo culture at intervals after initial antigen stimulus, typical antibody production is obtained. When, however, equivalent numbers of cells are transferred

after secondary antigen stimulation of the intact donor animal, the results are quite different. During the period 1 hr to 2 days after secondary antigen, there is a sharp loss (> 90%) in the ability of the cells to produce antibody in culture. Between 3 and 5 days there is partial restoration of antibody-producing capacity. Beyond 5 days there is another sharp decrease in antibody produced — in all cases by the same number of transferred cells. Experiments in progress will test the hypothesis that loss of antibody formation is due to migration of antibody-producing cells from the spleen upon challenge with a second injection of antigen.

It has been previously established that during primary and secondary antibody production, spleen cells divide with an average generation time of 12 hr, whereas in the nonstimulated spleen the average generation time is 24 hr. Also, the inhibitory effect of x irradiation upon antibody production has been studied extensively. The effect of x irradiation on spleen-cell generation in relation to secondary antibody response has been the subject of a recent investigation. When spleen cells are labeled (H^3-thymidine) and x-irradiated (500 r), there is a progressive decrease of labeled cells during the subsequent 48 hr. Thus these cells either do not divide or do so at a very slow rate. Antigen injection just after irradiation has no effect on division rate during this 48-hr period. During the first 4 days after irradiation of the primary immunized animal, there is little or no spleen-cell division. Capacity to divide begins to return shortly thereafter, and the generation time is about 24 hr. On the other hand, division capability is restored by 2 days when secondary antigen is administered immediately after irradiation. Under these conditions, the dividing cells have an average generation time of about 12 hr.

MOLECULAR- AND CELLULAR-LEVEL STUDIES

Cell Growth and Reproduction. — When the lens of the adult newt is removed operatively, it is regenerated by the dorsal margin of the iris tissue. That RNA synthesis may be enhanced during this classical example of cell-type transformation has been suggested by an earlier cytochemical study of K. Takata [Experientia 8, 217 (1952)]. An autoradiographic study of this phenomenon has been conducted with the adult *Triturus viridescens* as the material. Very soon after removal of the lens, and before the onset of regeneration, a strong increase in uptake of tritiated cytidine in the RNA of the nucleus of the iris cells has been demonstrated. The dorsal margin of the iris, where the regeneration occurs, seems to incorporate more isotope than the ventral margin. But both parts of the iris show a significant increase in the incorporation, compared with the corresponding parts of the iris in the undisturbed eye of the same individual. The results suggest not only the importance of RNA synthesis as an essential step of the morphogenetic event but also involvement of nuclear activity in determination of the differentiation type.

Biophysics: Electron Spin Resonance. — Studies have been made of electron spin resonances produced by ultraviolet (UV) irradiation of amino acids and proteins in the spectrometer cavity with the materials at almost liquid-nitrogen temperature. Taken at face value, the data show three results: (1) for equal ultraviolet exposures, tryptophan is the most

sensitive amino acid; (2) proteins show more resonances than the sum of their amino acid constituents, which suggests breakage of peptide or disulfide bonds in the proteins; and (3) for equal absorbed doses, far fewer resonances are produced by UV than by ionizing radiation, which suggests that excitations of less than 5 ev are insignificant in the production of free radicals by ionizing radiation.

Biophysics: Nuclear Magnetic Resonance. – Studies of the amount of bound and unbound water in a variety of intact and ground plant seeds by nuclear magnetic resonance (NMR) techniques have shown this approach to be ideally suited for such determinations. The technique will be of considerable value when used in conjunction with electron spin resonance studies of irradiated seeds where a knowledge of water content is of great importance. The method appears applicable in the range between 1 and 20% water content. Below 2 or 3% by weight the shape of the NMR absorption band indicates that the water is not in the liquid state and is possibly loosely chemically bound. More recent work has shown the method to be applicable also to such seed components as pure embryos alone as well as to the intact seed.

Biophysics: Ultraviolet Studies. – Studies on starved cells of *Amoeba proteus* have been done by ultraviolet (UV) irradiation of (1) whole cells, using a germicidal lamp; (2) nuclei of cells flattened to a thickness of 20 μ, using a broad band of far UV (2300 to 3400 A) in a single beam 25 μ in diameter; (3) cytoplasm of such cells, using four nonoverlapping microbeams 100 μ in diameter; and (4) cytoplasm of such cells, using the four microbeams, one of which includes the nucleus. The cytoplasm of amoebae has been found to be surprisingly sensitive to UV irradiation. Preliminary results indicate that death results from cytoplasmic irradiation at doses (per unit area) only about three times the lethal dose for nuclear irradiation. There appear to be differences in the ratio of death to division delay of cytoplasmic vs nuclear irradiation, but these have not yet been clearly resolved.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND INSTRUMENTATION

Basic Instrumentation. – An improved technique for producing p^+-i-n^+ diode detectors was developed. A thin (~20-μ) dead layer having a low (~1 to 2 ohms/square) sheet resistance was formed on the front of the p^+-i-n^+ junction diode, which had been made by the lithium drift process. The n^+ layer was formed by diffusing phosphorus into p-type silicon and then diffusing lithium through the phosphorus-doped layer. The concentration of lithium was controlled such that most of the lithium subsequently drifted into the silicon, leaving a thin dead layer. The rate at which the lithium drifted into the silicon was found to depend directly on the power dissipated in the diode. By means of an automatic control system, the lithium drift operation proceeded at power dissipations in excess of 50 w.

A detector prepared by this technique was tested before and after being stored for three months. Monoenergetic 1-Mev beta particles and a 50-v bias were applied to the diode. The pulse spectral shape did not change, indicating that the detector did not deteriorate measurably.

Several 20-mm^2 silicon surface barriers with protected edges (amine-free epoxy) and 0.5-mm-wide guard rings were tested. At 1000 v, reverse leakage currents were 1.0×10^{-7} to 2.0×10^{-7} amp. For 0.976-Mev k-conversion electrons from Bi^{207}, the resolution was 13.7 kev (full width at half maximum) at 1200 v applied bias (~1-mm barrier depth) and was less than 20 kev at 2000 v bias.

ISOTOPE DEVELOPMENT PROGRAM

Application of Radioisotopes in Analytical Chemistry. — Laboratory work was started on the development of a radiometric method for the determination of fluoride. In principle, the method involves the addition of an appropriate radioisotope to form a stable metal fluoride complex which is subsequently extracted. The radioactivity of the extract, which is then determined, is a measure of the concentration of fluoride. Various solvents for carrying out the extraction are being studied, as well as possible radioreagents such as Sb^{124}, Sb^{125}, Nb^{95}, and Ta^{182} for labeling the fluoride complex.

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