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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

FEBRUARY 1961

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

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

February 1961

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

SPECIAL NUCLEAR MATERIALS PROGRAM

Dissolver Solution Analyses: Uranium. – The "secondary" controlled-potential coulometric titration of uranium (with iron as the intermediate) was studied further. Precision estimates indicate that uranium can be determined by this technique with a relative standard deviation of 0.4% when 2.5 mg of uranium is titrated; the deviation is 0.2% or less when 10 mg or more is titrated. Uranium was determined by this technique in the presence of as much as 600 mg of type 304 stainless steel without a prior chemical separation. Molybdenum is a serious interference. Attempts to separate uranium from molybdenum by precipitation of ammonium diuranate have been unsuccessful.

In order to coulometrically titrate microgram quantities of uranium, the sensitivity of the ORNL mark III coulometric titrator was increased. It was found that uranium in the range of 500 to 1000 μg could be determined with a relative standard deviation of 0.8%. However, an extraction of 500 μg of uranium with 5% triisooctylamine in xylene followed by the coulometric titration did not yield satisfactory results. It was established that 1000 μg was the smallest quantity for a successful triisooctylamine extraction and subsequent titration of uranium.

A polarographic method for the direct determination of U(IV) in samples containing U(VI) as well as various other ions is being developed. It has been found that U(IV) ion demonstrates a well-defined anodic wave at an $E_{1/2}$ of −0.20 v vs the S.C.E. in a supporting electrolyte composed of a solution of $Na_5P_3O_{10}$ whose pH has been adjusted to approximately 9.0. The relationship of the diffusion current vs concentration of U(IV) has been found to be linear in the range of 50 to 200 μg of U(IV) per milliliter. The effect of pH and possible interfering ions has been studied, and a procedure for the dissolution and subsequent handling of samples to prevent air oxidation of the U(IV) present has been delineated. Further studies are being made for determining the feasibility of polarographically estimating directly both the U(IV) and U(VI) content of samples (such as U_3O_8) in the same medium. The data obtained to date indicate that both species can be determined directly in the same sample aliquot to within ±3%.

Dissolver Solution Analyses: Plutonium. – Exploratory work was done on the separation of plutonium by anion exchange from a sulfate medium.

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A distribution coefficient of approximately 400 was found when Pu(IV) in 0.05 M H_2SO_4 was equilibrated with Dowex 1-X4 resin, but increasing the acidity causes an abrupt decrease in the distribution coefficient. Consequently, this separation does not look promising for use in dissolver solution analysis.

REACTOR DEVELOPMENT PROGRAM

GAS-COOLED REACTOR PROJECT

Operation of the EGCR Without Experimental Loops. — An analysis is being made of the effect on EGCR operation of deferment of completion of the experimental loops. For this analysis it was assumed that there would be no stainless steel through-tubes, the small loop positions in the core would be filled with EGCR fuel elements, and the large loop positions in the reflector would be filled with graphite. The analysis has progressed to the point where the following conditions may be fixed:

1. It is unnecessary to load the initial core with two zones of different enrichment.
2. The enrichment of the first core should be 2.46 wt % U^{235}.
3. No half-strength control rods will be required at startup.

The basic philosophy of operation will be the same as that recommended with the loops present. Selected control rods near the center of the core will be fully inserted, and operation of the remaining rods as a bank will be used to provide continuous control. The first refueling will be required after about 30 months of operation, and the feed enrichment required for a 10,000 Mwd/metric ton equilibrium cycle will be ~2.25 wt % U^{235}. The enrichment of the initial loading has been set rather high in order to obtain a high average exposure for the first core, and an average exposure of about 7000 Mwd/metric ton should be realized. With the above combination of enrichments, an important result is the elimination of the severe hot-channel factor previously associated with loading the feed fuel elements into the initial core. This was the major factor in the previous recommendation of two-zone loading when the loops were present.

Performance of Beryllium-Clad 19-Rod Fuel Elements. — A study was made of the performance of a fuel element consisting of 19 beryllium-clad rods with and without fins. The basic configuration has nineteen 0.5-in.-OD rods arranged on an equilateral triangular pitch in a graphite sleeve. The sleeve has a hexagonal hole with rounded corners. The finned rods have six longitudinal fins per rod (0.5 in. OD) with a constant fin thickness of 0.040 in. and variable fin height.

The total power which could be taken out of a channel with an exit gas temperature of 1050°F and a nominal maximum surface temperature of 1150°F with the total available EGCR pressure drop was determined for fin heights of 0.100 and 0.145 in. Relative to the average power per channel with a seven-rod stainless-steel-clad EGCR fuel element, a fin height of 0.100 in. and a clearance between tips of fins on adjacent rods of 0.050 in. gave a value of 1.342 for a coolant flow rate of 2350 lb/hr, and a

fin height of 0.145 in. and the tips of the fins in contact gave a value of 1.713 for a coolant flow rate of 3000 lb/hr.

Dimensional Studies of EGCR Fuel Element. – Tests have been conducted to determine the effect of the lower temperature of a thermal cycle on the axial elongation of the EGCR fuel element. In these tests the lower temperature was varied from 572 to 1184°F, and the upper temperature of the cycle was held at 1500°F. The data indicate that the fuel elements should not undergo plastic deformation in the axial direction unless the lower temperature limit of thermal cycling is below 1000°F.

Studies of Graphite Outgassing. – Specimens 1-1/4 in. in diameter and 1 in. long obtained from section No. 2 of the EGCR graphite have been degassed in the temperature range 30 to 1800°C. The results indicate that the gas content of the outer portion of the bar is more variable than that of the central portion. Values ranging from 14 to 48 cm^3 of gas per 100 cm^3 of graphite were obtained for the outer portion, whereas a spread of only 44 to 55 cm^3 was found for the central region. The bulk of the graphite in the 6-in. slab examined has a gas content of 40 to 50 cm^3 per 100 cm^3 of graphite. The CO-to-H_2 ratio in the evolved gas was greater than 1 for all 11 specimens studied.

Specimens 1-1/2 in. in diameter and 2 in. long machined from the central portion of section No. 2 of the EGCR graphite were degassed from room temperature to 1000°C using external resistance heating. A total volume of ~18 cm^3 of gas per 100 cm^3 of graphite was measured. This is somewhat greater than the ~14 cm^3 found earlier for specimens cut from a region closer to the surface. The graphite from the central portion of the bar evolved a surprisingly large amount of SO_2 at 600°C. One specimen evolved 0.9 cm^3 of SO_2 per 100 cm^3 of graphite, while 0.6 cm^3 was found for another. The volume of gas evolved plotted against the logarithm of time gave a linear relationship at both 300 and 600°C. Similar treatment of the data obtained at 1000°C gave a line with a slope of 3.1 for the time interval 150 to 3000 min and another line having a slope of 5.2 for the time interval from 4000 to 10,000 min.

Irradiation Experiment Examination. – High-temperature measurements of the pressures in four EGCR prototype capsules irradiated in the ORR have been completed. The pressures in capsules 04-1, 06-1, 07-1, and 08-1 showed a linear relationship with temperature. The estimated burnups for these capsules were respectively 1600, 1900, 1350, and 1200 Mwd/metric ton. It appears that there is no significant pressure contribution from cesium at temperatures up to 1740°F.

Aliquots from dissolved fuel pellets irradiated in ORR capsules have been analyzed for Cs^{137}, Ce^{144}, and total uranium content. The anomalous results of the Cs^{137} analyses are being checked. It appears that some cesium may have plated out on the inner surface of the cladding.

Profile and outside-diameter measurements were made on ETR-irradiated stainless-steel-clad capsule E-9 at 90° intervals around the capsule by use of a double-point dial indicator. There was no change in diameter as a result of irradiation.

Two siliconized silicon carbide–coated graphite capsules containing pellets of UC_2 dispersed in graphite were irradiated in the closed-cycle facility in the B9 loop of the ORR at 1800°F in a thermal neutron flux of

1×10^{14} neutrons cm^{-2} sec^{-1}. After 170 hr of irradiation, an increase in the activity of the cooling gas was noted, and the test was terminated. The capsules will be removed from the reactor during the next shutdown and will be examined.

Instantaneous Fission-Gas-Release Experiments. – Additional measurements of xenon and iodine release from UO_2 were obtained by allowing the fuel to remain at constant flux and temperature until an equilibrium release rate was established. The fuel was then withdrawn from the reactor, and the iodine release rates were determined by measuring the continued xenon production. The amount of gaseous xenon released from the fuel during the cooling burst is indicated below:

Equilibrium Temperature (°C)	Total Atoms in Fuel		Atoms Released in Cooling Burst (%)	
	Xe^{133}	Xe^{135}	Xe^{133}	Xe^{135}
890	6.0×10^{16}	2.6×10^{15}	0.023	0.022
1010	6.6×10^{16}	2.7×10^{15}	0.048	0.065
1100	4.4×10^{11}	2.4×10^{15}	0.57	0.53

The amount of iodine released from the UO_2 at equilibrium was as follows:

Temperature (°C)	Amount of Isotope Released as Iodine (%)	
	I^{133}	I^{135}
720	100	100
890	46	83
1010	48	75
1100	28	46

The total amount of xenon emerging from the system (both from the UO_2 and the deposited iodine) was highly temperature dependent, as indicated below:

Temperature (°C)	Production (atoms/sec)		Release (atoms/sec)	
	Xe^{133}	Xe^{135}	Xe^{133}	Xe^{135}
720	1.43×10^{11}	13.6×10^{10}	2.5×10^{8}	6.8×10^{7}
890	0.914×10^{11}	8.7×10^{10}	6.0×10^{8}	2.3×10^{8}
1010	1.02×10^{11}	9.8×10^{10}	1.5×10^{9}	6.7×10^{8}
1100	1.02×10^{11}	9.8×10^{10}	3.7×10^{10}	9.9×10^{9}

Equilibrium was not attained at 1100°C. The measurements at 1100°C were unusual in that gas-release equilibrium was not established within the usual five days. Moreover, short-lived isotopes (which reach equilibrium quickly) showed a release rate that increased with time at the same rate as the release of longer-lived isotopes. This leads to the conclusion that some change in the release mechanism occurs, such as slow grain growth or grain movement. The test was terminated after 7 days when the reactor was shut down for refueling. The gas release rates given above for 1100°C are those at the end of the test. The fuel is now being operated at a lower temperature in order to determine whether the change in the gas-release mechanism was permanent.

Fabrication of Fueled BeO. — The fabrication of crack-free specimens of BeO containing 30 vol % UO_2 introduced as 100- to 150-μ particles was found to be reproducible. The highest density obtained was 92% of theoretical.

Microscopic cracking of BeO specimens containing fuel introduced as 150- to 250-μ particles was reduced by adjusting the heating rate during the initial sintering stage so that 1 hr was required to reach 1000°C. The highest density obtained in these specimens was 93% of theoretical. The microstructure of the specimens showed that a large portion of the fuel grains had fractured; this effect is not understood. Voids around the fuel particles indicated that the shrinkage had not occurred in the manner calculated. It appears that BeO containing fuel does not shrink to the same extent, based on green density, as unfueled BeO. Attempts are being made to solve this problem by increasing the preshrinkage of the fuel.

Fission-Gas Retention in BeO-Base Fuels. — Qualitative agreement was found between the amount of grain growth in sintered specimens of unirradiated BeO containing 20 vol % UO_2 and the amount of Xe^{133} released during irradiation of duplicate specimens. In 30 hr at 1800°C, there were marked increases in grain sizes of both the UO_2 and the BeO phases, and all the Xe^{133} in the irradiated sample had been released. It is concluded that the mechanism of release is associated with grain growth, as well as with xenon diffusion.

Adsorbers for Removing Fission Product Gases from Helium. — An initial iodine-retention test of Whetlerite charcoal, type ASC, 12/30 mesh, was terminated after 216 hr of operation at 325°C. No evidence of iodine release from the 0.25-in.-deep adsorber was noted during the test, and analytical data indicate that the charcoal retained 99.99+% of the iodine initially present. Whetlerite charcoal is designed for use in gas masks and is produced by impregnating Pittsburgh BPL charcoal with salts of silver, copper, and chromium. A test was also made on Pittsburgh BPL charcoal, 12/30 mesh, to differentiate the effect of these salts on iodine retention. After operation at 325°C for 168 hr, a 0.25-in.-deep bed of BPL charcoal was found to have retained only 30.2% of the iodine initially present. It appears therefore that the superior iodine-retention performance of Whetlerite charcoal is due to the presence of the metal salts. A second test of Whetlerite charcoal for 168 hr at 325°C again produced 99.99+% iodine retention.

Measurement of High Temperatures. — Tests have been started on a group of nickel-chromium alloys to determine whether they are suitable thermocouple materials for use in a stagnant helium atmosphere. The following types of thermocouples were prepared from bright wire: (1) Chromel-A and alloy 875, (2) Chromel-AA and alloy 875, (3) special Chromel-P and special Alumel, and (4) regular Chromel-P and regular Alumel. After six days of exposure at 1000°C, only the special Chromel-P and the regular Chromel-P showed significant drifts. The drift of both was −1.4 mv at 35°C.

THERMAL-BREEDER REACTOR PROGRAM

Homogeneous Reactor Program

Homogeneous Reactor Test. — Operation of the HRT with the new batch of fuel (added in January) was continued until February 8. The reactor ran at 3 to 5 Mw for 82 hr and it was concluded from the chemical analyses of the fuel that the fuel was stable. The reactor behavior was consistent with this conclusion, but interpretation of the temperature data was partially obscured by effects of a bypass which developed in the fuel letdown heat exchanger between the feed and letdown portions and by erratic operation of the feed pump.

The reactor was shut down on February 8 after fission product activity was detected in the reactor cell sumps. Smearing and monitoring techniques localized the source of the leak, which was found to have originated in a fitting in the feed line from one of the fuel dump tanks. The line will be equipped so that it can be isolated by freeze plugs during future operations. During this shutdown, the letdown heat exchanger, the fuel feed pump heads, and three high-pressure system valves will be replaced also.

Good progress is being made in the maintenance work, and operation should be resumed in March.

Thorium Blanket Studies. — In the preparation of thoria-gel spheres > 1/8 in. in diameter, the occurrence of cracking on drying and firing was shown to be related to the size of floc particles deposited during evaporation of sol. Thixotropy in the sol and strength in the gel were increased as floc sizes were decreased. Steam-denitrated thorium oxide containing initially 2.1% nitrate and 3.2% H_2O was agitated with water at 100°C; the nitrate content of the aqueous phase increased from an immediate value of 0.07 M to a maximum of 0.12 M and the acid deficiency from 0.05 to 1.10 M in 40 hr. The reaction was similar but slower at 26°C. With the above changes, thixotropy was increased to a maximum and floc size to a minimum at about midrange, while further hydrolysis increased floc size and converted the thixotropic sol to a free-flowing one.

Chemical Research for Aqueous Homogeneous Reactors. — In the five-component HRT fuel system, UO_3-CuO-NiO-SO_3-H_2O, the compositions of solutions saturated simultaneously with the three solid phases $UO_3 \cdot H_2O$, $CuO \cdot 3UO_3$, and $NiO \cdot 3UO_3$ were determined at 300, 325, and 350°C. In these solutions the UO_3:SO_3 ratio at 300°C increased from 0.55 to 0.95 as the sulfate concentration was increased from 0.002 m to 0.06 m; the CuO:SO_3 and

$NiO:SO_3$ ratios were 0.009 ± 0.003 and 0.024 ± 0.004, respectively, over this sulfate concentration range. When the temperature was raised, the $UO_3:SO_3$ ratio was decreased; at 0.01 m sulfate the ratios were 0.75, 0.72, and 0.69 at 300, 325, and 350°C respectively. At 350°C and approximately 0.016 m sulfate an isothermal invariant point was found at which the three solids were simultaneously in equilibrium with two liquid phases; the light phase had a $UO_3:SO_3$ ratio of 0.75; the $CuO:SO_3$ and $NiO:SO_3$ ratios were very low.

In the three-component system $CuO-SO_3-D_2O$, a subsystem of the five-component HRT fuel system, the solubility of $3CuO \cdot SO_3 \cdot 2D_2O$ was determined at 300, 325, and 350°C. The $CuO:SO_3$ ratios in the saturated solutions were 0.50, 0.60, and 0.75, respectively, in solutions which were 0.010, 0.10, and 1.0 m in sulfate. These ratios were nearly independent of temperature in this range. At 350°C and approximately 0.1 m sulfate an isothermal invariant point was located at which $3CuO \cdot SO_3 \cdot 2D_2O$ and two liquid phases were in equilibrium, with a $CuO:SO_3$ ratio of 0.58 in the light phase. At higher sulfate concentrations the solid phase disappeared, leaving only the two liquid phases.

In the system $UO_3-SO_3-H_2O$, isothermal invariant points were located at which one solid phase, $UO_3 \cdot H_2O$, and two liquid phases were present in equilibrium; the light-liquid-phase invariant compositions had $UO_3:SO_3$ ratios of 1.06, 0.93, and 0.79 with sulfate concentrations of 0.125, 0.048, and 0.019 m at 300, 325, and 350°C respectively. The solubility of $UO_3 \cdot H_2O$ in $H_2SO_4-H_2O$ was determined at these temperatures over a range of sulfate concentrations from 0.015 m to those at the invariant points; the $UO_3:SO_3$ ratios were near 0.78 in 0.015 m sulfate at all three temperatures but showed a small decrease with increasing temperature.

The temperature dependence of the iron-catalyzed decomposition of peroxide in dilute sulfuric acid was studied over the range 140 to 250°C. An interesting anomaly was noted in the range 190 to 210°C, where the observed rate decreased with increasing temperature. The phenomenon has been interpreted as the inactivation of dissolved iron via a hydrolytic process.

Slurry Circulation Experiments. — The 200B loop test section for measuring friction factors in turbulent flow was installed and was calibrated with cold water. The observed friction factors were close to those predicted from standard correlations.

Capillary-tube viscometer measurements were made with LO-41 thoria slurry at three concentrations and four temperatures. It was noted that the ratio of coefficient of rigidity to water viscosity was approximately independent of temperature for a given slurry concentration. A marked "slip" effect was noted, particularly at the highest concentrations and at moderate rates of shear. At high rates of shear the flocs appear to be broken up so strongly that the settling unit (and consequently the slip effect) is quite small.

The 200Z slurry pump, which contains alumina radial bearings operating in water, has operated for 8770 hr.

Engineering Development. — The HRT mockup was shut down as a result of failure of the suspension spring of the float level controller. A run in progress — circulation of uranyl sulfate solution at 260°C with an excess H_2SO_4 level of 0.045 m — had indicated no abnormal corrosive attack.

Testing of the oxygen compressor for use in recycling contaminated gas from atmospheric pressure to 2500 psi was discontinued because of the impending close-out of the program.

The full-scale model of the HRT core was operated with the thermocouple and the corrosion-specimen holders. Excessive flow bypassing from inlet to outlet was observed on the basis of salt injection measurements. In the case of the thermocouple holder, the bypassing was reduced to a marginally acceptable level of 30% with the use of a flow restrictor at the core entrance.

The flow model was converted into a reactor simulator by injecting salt at a number of points in the vessel (simulating heat generation), and the signal from a conductivity probe at the core outlet was fed into an analog computer programmed to calculate and record Δk and power on the basis of the HRT temperature coefficient, delayed neutrons, and power level. The records, obtained at a number of simulated core power levels between 0.4 and 2.5 Mw, bear a striking resemblance to the period and power traces of the HRT. A statistical comparison of the two different sets of data is being made.

The spare HRT circulating pump was prepared for a hot run-in test.

Design. – Flow tests of the proposed thermocouple and corrosion-specimen holders for insertion into the HRT core indicated the need for an improved flow pattern, and modified designs were therefore investigated. It appeared practical to improve the flow by attaching the corrosion specimen holder to the lower screens (rather than to the core access flange) and by adding a flow restrictor at the entrance to the core. Methods were devised for placing thermocouples at any of a limited number of desirable locations in the core or in the outlet line.

A conceptual design was completed for a flow nozzle which might be inserted into the core entrance of the HRT. This nozzle would probably improve the core-wall cooling and could possibly reduce the magnitude of power fluctuations observed during reactor operation.

MOLTEN-SALT REACTOR PROJECT

MSRE Design. – Drawings for the reactor, drain tanks, heat exchanger, and radiator were completed. As soon as the drawings have been checked, they will be issued for final approval.

Electrical-heater circuits for the primary cell have been determined and have been sized as far as load centers, conductors, and control devices are concerned.

The building alteration design is proceeding satisfactorily.

The conceptual layout was completed for a set of remote-maintenance tools for pump removal. The tools are now being detailed.

A method was developed for providing sampling of the core graphite without removing the pump. The method involves a 3-in. penetration of the reactor-vessel top head by a sched-40 pipe. Detailing of the graphite-sampling scheme is in progress.

Component Development. – The operation of long-term forced-circulation corrosion test loops was concluded. During the program 25 loops were operated an aggregate of 289,500 hr at test conditions. The program has demonstrated the ability of INOR-8 to contain molten fluorides.

Two natural-circulation loops, testing the compatibility of MSRE fuel salt, molybdenum, graphite, and INOR-8, remained in operation.

Residence times were measured in the lower head of the one-fifth-scale core model by following the decay of electrolyte concentration with a conductivity probe. Calculations based on the measurements indicate that the highest temperature in the lower head will be less than 20°F above the mixed mean temperature of fluid in the head.

A mockup of the sampler-enricher drive mechanism, latch, sample capsule, connecting tube, latch stop, and capsule guide was constructed, and testing was started.

Testing of a resistance-heated freeze valve in a 1-1/2-in. salt line was concluded after 100 trouble-free freeze-thaw cycles. Testing of a Calrod-heated valve, which has a more convenient electrical circuit, proceeded successfully up to a total of 80 freeze-thaw cycles.

Remote Maintenance. — A portable-shield mockup is being constructed for installation on the manipulator bridge of the Remote-Maintenance Demonstration Facility. Tooling suited to semidirect work will be developed and will be tested in use with this shield.

Preliminary tests indicated that two types of manifolded, remotely operated disconnects are feasible for MSRE-type operations. In one type of joint, up to eight cones in one block of the assembly fitted into surfaces machined into a second block; the assembly was held together by a clamp operated with a single bolt. In a second type of joint, concentric Flexitallic gaskets were used to provide sealing.

Pump Development. — The Inconel centrifugal pump (PKP type) has now circulated $LiF-BeF_2-ThF_4-UF_4$ (65-30-4-1 mole %) at 1225°F for 3530 hr. Equipment modifications were completed for the back-diffusion experiment utilizing Kr^{85}. The pump with a molten-salt-lubricated journal bearing has now circulated $LiF-BeF_2-UF_4$ (62-37-1 mole %) at 1225°F for 7250 hr.

The xenon-removal (stripper) configuration was fabricated and was installed in the water test stand for the MSRE primary pump. Initial testing at a 70-gpm bypass flow indicated the need for installing baffles in the pump tank to control behavior of the pump-tank liquid. The time required to reduce the concentration of dissolved CO_2 by a factor of 2 will be measured for this stripper configuration at 70- and 50-gpm bypass flow rates.

Thermal-stress calculations for the primary pump were checked, and the calculation methods were verified. The stresses, based on an approximate temperature distribution, appear to be compatible with the strength of INOR-8 at the given temperature. Calculations are being made for verifying the assumed temperature distribution.

Specimen bars poured from the same heat as the first INOR-8 impeller or volute casting will be used at ORNL to determine the weldability of the cast material. Alternative methods of joining the cast volute to the wrought pump tank are being considered.

Chemistry. — Intensive efforts to obtain reliable analyses of trace concentrations of oxides in fuel melts are beginning to give quantitative information regarding several aspects of MSRE fuel behavior. Careful monitoring of oxide and sulfide levels during fuel purification with H_2 and HF shows that the undesired anions are removed from solution at a fast

rate, even though precipitates are very intractable. The use of H_2 in admixture with HF effectively suppresses corrosion of nickel and copper containers. The fuel, $LiF-BeF_2-ZrF_4-ThF_4-UF_4$ (70-23-5-1-1 mole %), has an average gram molecular weight of 43.5 and a liquid molar volume of about 17.5 cc. When 40 moles of melt are treated with 4 moles of HF (H_2: HF, 1:1) at a rate approaching 0.02 mole of HF per minute, the dissolved oxide falls from 2000 to 200 ppm, and the total sulfur from 400 to 40 ppm. Oxide analyses lower than 100 ppm were not encountered even after prolonged treatment.

The effectiveness of ZrF_4 as an oxide scavenger to protect against precipitation of UO_2 has been examined in $LiF-BeF_2-ZrF_4-UF_4$ (60-35.5-4-0.5 mole %) by means of incremental addition of BeO as an intentional contaminant. The BeO appears to react metathetically with ZrF_4 to precipitate ZrO_2 until the Zr^{4+}/U^{4+} ion concentration ratio in the liquid reaches 1.5; then the UO_2 and ZrO_2 coprecipitate and the residual concentration ratio remains constant. This implies that ZrO_2 is 1.5 times as soluble as UO_2; an apparent oxide concentration of 500 ppm associated with solution saturated with both oxides provides an upper limit for the solubility products, but further confirmation of the validity of the oxide analysis is being sought.

Metallurgy. – An evaluation is being made of the crack susceptibility of various heats of INOR-8 as a result of welding. Cracking and/or microfissuring has been observed in test welds made with various combinations of base material and filler material. In an attempt to evaluate the seriousness of the problem, several test weldments have been prepared, a large number of metallographic samples are being examined, and other weldments are being prepared. In addition, hot-ductility test samples are being machined from several different heats of material for testing and evaluation at Rensselaer Polytechnic Institute. Complete chemical analyses on the various heats are also being obtained.

As a result of the various tests conducted to date, it appears that weld-metal cracking can be essentially eliminated by adequate purification and deoxidation of the melt during initial casting. The addition of 2% Cb to the basic INOR-8 composition appears to provide an added insurance against weld-metal cracking.

The influence of melting practice and composition upon microfissuring in the heat-affected zone is also under study.

In-Pile Tests. – Assembly work is continuing on the third MSRE graphite-fuel capsule (ORNL-MTR-47-3). Fuel fabrication is complete, and samples are being analyzed. A preliminary test designed to allow observation of the fuel melting and freezing in a quartz tube was unsatisfactory insofar as visibility was concerned. It was demonstrated, however, that the fuel can melt and freeze without causing structural damage to the graphite.

Final assembly of the in-pile capsule should begin shortly.

Reactor Analysis. – Estimates were made of the dose rates outside the MSRE reactor cell and drain-tank cell with currently proposed shield thicknesses of 7 ft and 6 ft of ordinary concrete respectively. Above the reactor cell the dose rate during operation was estimated to be 90 mr/hr. Additional shielding will be added above the reactor and drain tank cells to reduce the radiation level to less than 1 mr/hr. The dose rate above the drain-tank cell, immediately after draining the reactor core, was estimated to be 62 mr/hr; 2 hr after draining the core this dose

rate was 0.47 mr/hr, and the integrated dose over a long period of time amounted to less than 25 mr.

Preliminary nuclear calculations were completed for an MSRE core having the fuel separated from the graphite by INOR-8 tubes. For a core with 8 vol % fuel salt, insertion of 30-mil-thick, 2-in.-diam INOR-8 tubes increased the critical circulating-system inventory by about 70%; the percentage of fissions caused by thermal neutrons decreased from 92% to about 88%; the average number of fission neutrons released per neutron absorbed in the fuel was reduced from 2.02 to 2.00; and the temperature coefficient of reactivity due to expansion of the core salt was reduced by about 1%.

Heat Transfer and Physical Properties. — Measurements on the enthalpy of the coolant mixture $LiF-BeF_2$ (66-34 mole %) have been partially completed. The preliminary data indicate a heat capacity of approximately 0.53 Btu lb^{-1} $(°F)^{-1}$ for the liquid between 400 and 650°C. The dependence of the heat capacity on the temperature over this range will be established as additional data are accumulated.

Assembly of the apparatus for determining the density and surface tension of the MSRE fuel mixture is essentially complete. Initial testing with water showed some leaks in the pressure-measuring system. Studies with the salt mixture will be started after correction of this difficulty.

Fuel Processing. — The compound precipitated by addition of liquid SbF_5 to a solution of LiF in anhydrous hydrogen fluoride was not stoichiometric, typically having an Li/Sb ratio of 1.4. Some of the compound was recrystallized from anhydrous HF by dissolving it in a large volume (solubility about 0.9 g of Li per liter) and slowly evaporating the solvent. Well-formed crystals > 1 mm on an edge were obtained, in contrast to the fine powder that was dissolved. Chemical analysis indicated that this is the 1:1 $LiF-SbF_5$ compound, and x-ray diffraction examination showed that the crystals were monoclinic, with cell dimensions of 5.42, 5.18, and 7.50 A and an angle of 93.6°. The theoretical density is 3.83 g/cc. Properties for this compound have not been reported previously.

The $LiF-SbF_5$ compound, when heated in an inert atmosphere in a copper or nickel container, decomposed in the range 400 to 650°C, as indicated by the appearance of a white smoke when the vapor contacted moist air. The volatile material was SbF_3, however, instead of SbF_5. Apparently the compound reacted with the container to form NiF_2 or CuF_2 and SbF_3, which then volatilized. Decomposition temperatures were lower in copper containers than in nickel. In nickel the compound was not completely decomposed after being heated to 700 to 750°C, and it may be stable to higher temperatures if reaction with the container does not take place.

The dissolution rate of $LiF-BeF_2$ (63-37 mole %) in stirred boiling 90% HF averaged about 50 ml/hr for the first 10 min, and then dropped to around 10 ml/hr. A surface layer, believed to be BeF_2, formed as the dissolution proceeded. The rate was lower when the temperature was below boiling, and stirring had little effect on the rate when the solution was boiling.

ANP PROGRAM

Shielding Research. — The new Tower Shielding Reactor (TSR-II) was operated up to a power of 100 kw. The ratio of neutrons to gamma rays in rems at 75 ft from the bare reactor was greater than 25 to 1.

The TSR-II was then placed in the Pratt and Whitney reactor shield mockup in order to check the shield design calculations. This will be done by determining whether the dose in the Boeing crew shield with the reactor operating at 100 kw agrees with the calculated value. This experiment will be followed by a comprehensive examination of the shielding characteristics of the mockup.

Columbium Alloy Research. — Two types of stainless-steel-clad columbium tubing were studied. One specimen consisted of type 446 stainless steel cladding over columbium. A flash plating of copper was applied to the outer surface of the columbium and the inner surface of the stainless steel to promote bonding. The other specimen consisted of three layers: type 304 stainless steel over copper over columbium. Both specimens were heat-treated for 2 hr at 1850°F after the drawing operation. Metallographic examinations showed that in both types of tubing there was good bonding at the steel-copper interface and poor bonding at the columbium-copper interface. In order to further evaluate the bonds, samples of the tubes are being heated to temperatures up to 1800°F, and other samples are being thermally cycled between 1700°F and room temperature. Machining of the tubing was also started to prepare specimens for experimental fabrication of tube-to-header joints.

The aging response of Cb—1% Zr alloy specimens from heat S8FW, one of the heats used in the original aging studies, was determined after 2 hr of solution annealing at 1600°C and at 1800°C. The aging response at 1500°F after annealing at 1800°C was greater and more rapid than after annealing at 1600°C. This result would be expected if the higher temperature anneal caused greater solubility of the material responsible for aging. If it is assumed that oxygen causes the aging, as was postulated previously, it can be concluded that a temperature higher than 1600°C is required to dissolve all the oxygen in the alloy. The oxygen content of the alloy after heat treatment has not yet been determined; it was ~200 ppm prior to annealing.

A study of weld aging of Cb—1% Zr alloy as a function of postwelding heat treatment was nearly completed. The aging of welds in specimens of heat S4KW was verified by an aging treatment of 100 hr at 1500°F. Then welds made in specimens of this heat were annealed at temperatures ranging from 1700 to 2200°F in 100°F increments for 1, 2, 3, 4, and 5 hr, and representative annealed specimens were given an aging treatment of 100 hr at 1500°F. The Rockwell B hardness of all specimens was measured and bend tests were made. Complete metallographic surveys of the unaged annealed welds were conducted, and a survey of the annealed and aged welds is in progress. The data obtained thus far indicate that the most effective annealing temperature is in the range 2000 to 2200°F and that the annealing time should range from 3 hr at the lower temperature to 1 hr at the higher temperature. Also, the annealing treatment apparently prevents any further aging, with attendant embrittlement, from taking place. The metallographic study has shown that a precipitate, which is presumed to be the same material as that which causes embrittlement, appears after effective annealing as an extremely heavy coherent network throughout the matrix. The amount of this precipitate definitely increases with time and temperature. It is postulated that aging no longer occurs after annealing since the annealing causes the precipitate to assume a form and distribution which prevents it from acting as a strengthening agent.

A second in-pile tube-burst experiment was carried out in the ORR on an assembly of eight specimens prepared from Cb–1% Zr alloy supplied by Pratt and Whitney. The failure times on the eight specimens indicated that at 1800°F there is no more than a 10% reduction in rupture strength due to irradiation. The times to failure were all less than 55 hr, and the neutron flux (> 1 Mev) was approximately 5×10^{13} neutrons cm^{-2} sec^{-1}.

Liquid-Metal Technology. – Construction of a Haynes alloy No. 25 loop for circulating boiling and condensing potassium was completed. A run scheduled for 3000 hr was begun with the boiler temperature at 1600°F.

Preliminary data were obtained on the thermal conductivity of molten lithium (99.98% purity) by using an axial-heat-flow, comparison-type apparatus with compensating guard heating. The values obtained for the conductivity, based on heat flows calculated from the stainless steel heat meter, are shown below:

Temperature (°F)	Thermal Conductivity [Btu hr^{-1} ft^{-1} $(°F)^{-1}$]
650	27.5
750	27.8
890	27.2
1075	28.7
1490	28.0

Beryllium Oxide Research. – Inspection of BeO samples from the last three irradiation experiments in the ETR revealed serious deterioration in some specimens. One experimental assembly contained three 1-in.-long, 0.8-in.-diam specimens of hot-pressed BeO (density 2.9 g/cm^3) that were cut from large hexagonal blocks originally made for the ARE. The central temperature was approximately 1000°C, and the outer wall temperature was approximately 850°C. These specimens received a dose of approximately 5×10^{20} fast neutrons/cm^2 (> 1 Mev). The examination showed that these three specimens had some cracks but that they were not falling apart. This experimental assembly also contained six other specimens of BeO with diameters ranging from 0.8 to 0.4 in. that were at lower temperatures and received less irradiation; they showed no cracks. In addition, three 0.8-in.-diam specimens were irradiated to a dose of 5×10^{20} fast neutrons/cm^2 (> 1 Mev) at a central temperature of 120°C with a temperature gradient of approximately 50°C. These specimens also showed no cracks.

In another experiment, 12 capsules 1 in. long and 0.8 in. in diameter (density 2.6 g/cm^3) were irradiated to doses ranging from 10^{21} to 5×10^{20} fast neutrons/cm^2 (> 1 Mev). The central temperature of these capsules was approximately 120°C, and again there was a temperature gradient of approximately 50°C. These samples had been prepared by isostatic pressing and firing at 1570°C. The samples with the highest neutron dose cracked during the irradiation and broke into several pieces. When the cracked samples were left open in the hot cell, their surfaces became powdery.

The low-density samples were also used in an experiment involving a variety of neutron doses and temperatures. Nine specimens had central

temperatures of 900°C and temperature drops of 100°C to the outer wall. They received doses of 10^{21} fast neutrons/cm^2 (> 1 Mev), and each cracked into several pieces and showed surface powdering in the hot cell. Three specimens that were irradiated at about 800°C (central temperature) with a temperature gradient of approximately 100°C and which received doses of $\sim 5 \times 10^{20}$ fast neutrons/cm^2 (> 1 Mev) started to crack but did not fall apart. Six specimens that were irradiated at central temperatures between 440 and 740°C and that received doses of less than 5×10^{20} fast neutrons/cm^2 (> 1 Mev) showed no cracking. Three cold specimens (irradiated at 120°C) received a dose of 10^{21} fast neutrons/cm^2 (> 1 Mev). The stainless steel container on these specimens ruptured during the irradiation and exposed the beryllium oxide to the cooling water. These specimens were also broken and exhibited surface powdering in the hot cell.

A beryllium hydroxide product of unusually high purity was obtained in gram quantities from the Versene—acetyl acetone purification process. Spectrographic analysis showed the following reportable impurities:

Impurity	Quantity (ppm)
Mg	5
Si	10
Sn	5
V	5

The following impurity elements, if present, were in quantities below the present spectrographic standards, that is, below the minimum levels indicated below:

Element	Spectrographic Standard Minimum Level (ppm)
Mo	< 50
Zn	< 25
Cr, Ni, Ti	< 20
Al, Ba, Ca, Fe, K, Li, Na	< 10
Co, Cu, Mn, Pb	< 5
Ag	< 0.5

Additional quantities of beryllium hydroxide are being prepared, both to serve as improved analytical standards and for conversion to BeO.

Three samples of BeO (totaling 500 g) prepared by steam precipitation from molten fluoride solvent were analyzed and submitted to GE-ANPD for evaluation of their ceramic properties. They were composed of single crystals with average diameters of 10 to 20 μ. The principal impurities were those characteristic of the solvent and container materials, and the three samples differed in impurity content as a consequence of different degrees of purification following the precipitation step.

NUCLEAR TECHNOLOGY AND GENERAL SUPPORT

Release of Fission Products on In-Pile Melting of Reactor Fuels. — The first meltdown experiment in the In-Pile Fuel-Element Catastrophe Program (ORNL-3062, p 11) has been approved by the Reactor Experimental Review Committee. The completed facility in the F-9 lattice position of the ORR was operated with a device for measuring gamma heating as a function of vertical position substituted for the meltdown capsule. Operation of the facility has demonstrated satisfactory performance of the positioning mechanism and of the assembly for control of the cooling water. Such operation will continue to establish gamma heating as well as thermal- and fast-neutron fluxes as a function of position. Meanwhile, equipment and materials for the first meltdown capsule are being assembled.

Basic Shielding Research. — An attempt is being made in the Lid Tank to verify a geometrical transformation concept that the axial dose rates in a homogeneous medium from a large source plate can be inferred from the dose rates from a small disk source. Expressed mathematically,

$$D(z,a) = D\left(z, \frac{a}{\sqrt{n}}\right) + D\left(\sqrt{z^2 + \frac{a^2}{n}}, \frac{a}{\sqrt{n}}\right) + \ldots + D\left(\sqrt{z^2 + \frac{(n-1)a^2}{n}}, \frac{a}{\sqrt{n}}\right),$$

where

a = the radius of the source plate,

z = the distance from the source plate along a line perpendicular to the center of the source plate,

n = an integer.

The source-plate power will be varied by using several different diameter irises in a 1/2-in.-thick boral shutter placed between the source plate and the beam from the ORNL Graphite Reactor. Measurements will be made of the fast-neutron and gamma-ray dose rates and the thermal flux along the z axis. Preliminary measurements are in progress.

Power Reactor Fuel Processing: Solvent Extraction Studies. — Thorium was removed from unmodified U^{233} pilot-plant product (2 N H^+) by adsorbing 97% of the thorium and 10% of the uranium on Dowex 50W ion exchange resin. When the acid in the product was decreased to 0.1 N, 97% of the thorium and 4% of the uranium was adsorbed.

Zirconium solutions containing Zr^{95} tracer were prepared by extraction with thenoyltrifluoroacetone or by complexing the zirconium with fluoride, followed by separation from the fluoride. In tracer experiments on the distribution of zirconium between nitric acid solutions and 30% tributyl phosphate in dodecane, the results showed excessive scatter.

Solvent satisfactory for solvent recovery studies was prepared by diluting highly degraded solvent with fresh, undegraded solvent.

Uranium was extracted directly from 21 N HNO_3 with TBP in Amsco.

Power Reactor Fuel Processing: Processing of Beryllium and UO_2-Al_2O_3 Fuels. — Unirradiated Idaho GCRE fuel pellets containing 70% UO_2–30% BeO were completely dissolved in boiling 2 M HNO_3–4 M HCl in 22 hr and in boiling HNO_3-H_2SO_4 solutions with molarity ratios of 3/5, 3/7, 5/3, and 7/3 in about 24 hr.

When unirradiated Maritime GCR prototype UO_2–8% Al_2O_3 and UO_2–13.5% Al_2O_3 fuel pellets were leached with boiling 10 M HNO_3 for 4 hr, uranium losses to the undissolved Al_2O_3 residue were 1.3 and 3.3% respectively.

Power Reactor Fuel Processing: Zirflex Process. – In laboratory tests in which 3 to 4.5% H_2O_2 was added to the 6 M NH_4F–1 M NH_4NO_3 dissolvent at rates of 3.6×10^{-6} to 1.6×10^{-5} mole/min per gram of U-Zr-Sn fuel alloy charged, the solution darkened for a time, indicating the presence of unoxidized elemental tin, and cleared as the tin was oxidized. There was no observable precipitation of UF_4. Stable solvent extraction feed containing 0.4 M Zr, 1 g of U per liter, 2.7 M F, 1 M Al, and 1 M HNO_3 was prepared from the dissolver product by adding 1.25 volumes of 1.8 M HNO_3–1.8 M $Al(NO_3)_3$.

Power Reactor Fuel Processing: Mechanical Processing. – Tests were made with the inclined-drum rotary leacher to determine the feasibility of using a conveyor of similar design for surge storage and feeding. The results indicated that delivery of a 3-liter batch of sheared stainless steel rods 1/2 in. in diameter by 1 in. long varied from 95 to 105% of theoretical when the leacher was rotated continuously at 2, 3, and 4 rpm at an elevation of 20°.

To simulate continuous leaching, 18.9 ml/min of 7 M HNO_3 was passed for 4 hr over a series of beds containing 600 g of PWR reject UO_2 pellets each. Steady-state conditions were approached in 5 to 6 stages, and the product composition profile varied from a maximum uranium loading of 550 g/liter and 0.8 M H^+ to 450 g of U per liter and 2.0 M H^+.

Power Reactor Fuel Processing: Corrosion Studies. – In Zirflex corrosion tests with flowing 5.4 M NH_4F–0.33 M NH_4NO_3–0.003 M H_2O_2, LCNA, Hastelloy F, and types 309SCb and 304L stainless steel were corroded at over-all rates of 1.16, 2.83, 2.27, and 13.36 mils/month, respectively, in exposures of 432 to 500 hr.

In tests to find a material of construction for a multipurpose power reactor fuel processing centrifuge, titanium-45, Carpenter 20, LCNA, Hastelloy F, and Hastelloy C were corroded at over-all maximum rates of 0.01, 0.03, 0.03, 0.03, and 0.33 mil/month, respectively, in 672 hr exposure to spent Thorex solution at 35°C. In spent UO_2 core dissolvent solution at 35°C, LCNA, Carpenter 20SCb, and Hastelloy F were corroded at maximum rates of 0.25, 0.48, and 0.69 mil/month respectively. Both Carpenter 20SCb and Hastelloy F suffered extensive weld corrosion.

Power Reactor Fuel Processing: Processing of Graphite-Containing Fuels. – Only 30% of uranium-free graphite specimens, density 1.48 to 1.65 g/cc, were reduced to 10 mesh powder in 4 hr by boiling 90% (21.2 M) HNO_3; 94% of a fuel sample containing 8% UO_2 was reduced to 10 mesh powder. From uranium-graphite specimens containing 2.8 to 16% uranium irradiated to 0.001% burnup, at least 99.8% of the uranium was recovered in two 24-hr leaches with boiling 90% HNO_3; after two 4-hr leaches, 60% of the Ce^{144}, 35% of the Ru^{106}, and 38% of the Zr^{95} remained in the graphite residue.

In engineering time-cycle tests, graphite–3% U fuel leached 24 hr at 60°C or 96 hr at 25°C with 88–91% HNO_3 contained ~600 ppm undissolved uranium, indicating that extended leaching at lower temperatures may be less effective than several cycles of leaching with acid and washing with water at a higher temperature and for the same total processing time.

In sedimentation tests, only 0.4% of unirradiated graphite–3% U fuel exposed to fuming nitric acid for 24 hr at 30 to 80°C was disintegrated to 10-μ particles. It is thought unlikely that mechanical attrition in subsequent water washes would measurably increase the fraction of < 10-μ particles.

Water flow rates through 2-in.-diam beds of disintegrated graphite-containing fuel, 6 to 18 in. thick, ranged from 24 to 32 gal hr^{-1} ft^{-2} per kilogram of graphite, and pressure drops ranged from 3 to 18 in. Hg.

In laboratory tests, uranium monocarbide reacted with 5.6 M HCl at 80°C to produce a green solution containing > 99% U(IV). The reaction rate was much slower with 5.6 M HCl than with water. The off-gas contained 91.6 ml (STP) of gas per gram of carbide and consisted of 10% hydrogen, 88% methane, 1% ethane, and small amounts of paraffins with 3 to 7 carbon atoms.

Uranium monocarbide reacted with 4 M HNO_3 to produce a dark-red solution. Near the end of the reaction the off-gas was 65% NO, 23% CO_2, 4% N_2O, 1% CO, and 5% N_2 (probably from atmospheric contaminants) and had a faint brown color indicative of NO_2.

Power Reactor Fuel Processing: Zircex Process. – A Zircex process was developed in the laboratory for recovering uranium from spent TRIGA reactor fuel. The aluminum cladding, the aluminum-samarium burnable poison, and the core (8% U–91% Zr–1% H) are hydrochlorinated at 300 to 600°C to produce volatile aluminum and zirconium chloride and relatively nonvolatile uranium chloride containing some oxide impurities. The residual uranium trichloride and the oxide impurities are converted to volatile chlorides by chlorination with carbon tetrachloride vapor at 550°C. The volatilized uranium chloride is selectively condensed at 300°C, and the zirconium and aluminum chlorides are condensed downstream at room temperature. The unreacted graphite plugs are then burned in oxygen at 750°C. The condensed uranium chlorides are dissolved in nitric acid prior to chloride removal and solvent extraction. Twenty-two hours is required for the entire process. The off-gas is hydrogen and carbon dioxide.

Treatment and Disposal of Radioactive Wastes: Low-Activity Wastes. – A bench-scale pilot plant designed to demonstrate the removal of fission product cesium and strontium from ORNL low-activity waste by ion exchange following a precipitation-flocculation step operated successfully under various conditions of feed flow and composition. Thirteen hundred bed volumes of effluent spiked with Sr^{85} and Cs^{137} were passed through a column of CS-100 resin (a carboxylic-phenolic cation exchange resin) before the 0.1% breakthrough point (df = 1000) was reached. Breakthrough of 1% occurred for cesium at about 1700 bed volumes. No strontium activity was observed in the effluent at any time. Upflow elution of this resin with ten bed volumes of 0.5 M HNO_3 removed 99.9% of the cesium and strontium. No gassing was observed when the resin was left in stagnant contact with 0.5 M HNO_3 for 72 hr, but some gassing was noted after five days.

Design of a pilot plant for demonstrating this process with ORNL waste was started. Equipment currently available for this plant will set its capacity at 10 gpm, or 1/50 the capacity of a plant needed to handle the entire Laboratory waste stream.

Treatment and Disposal of Radioactive Wastes: High-Activity Wastes. – The addition of phosphorous acid to Purex waste before batch calcination

decreased the ruthenium volatilized from 21% to 1.9, 0.04, and 0.03% when the solution was 0.61, 1.52, and 3.05 M, respectively, in H_3PO_3. In a mixture carried to 900°C, B_2O_3 and SiO_2 added to give a glassy melt did not interfere with the effect of phosphite on the ruthenium. Ruthenium material balances varied between 99.4 and 100.8%. Type 304L stainless steel heated at 900°C for 168 hr in contact with the powdered solid from calcination of Purex waste showed an over-all corrosion rate of 3.6 mils/month with no observable localized attack.

A design and cost estimate for a high-level pot calciner pilot plant indicated that installation in a new four-cell building at ORNL would cost $2.5 million. Installation of the same equipment in existing space at the Hot Pilot Plant of the Idaho Chemical Processing Plant was estimated to cost $1 million.

Waste Disposal Research and Engineering: Geochemical Studies. – The ability of anthracite coal, bituminous coal, lignite, and peat to remove tracer quantities of radionuclides in water was measured. Anthracite and bituminous coal showed very poor removal qualities for strontium, cesium, cobalt, and ruthenium. The decreasing order of removal by lignite and peat for the four elements was Co > Sr > Cs > Ru. In general, the best removal was obtained at pH 8 to 10; ruthenium removal was better in the acid pH range, in contrast to the behavior of the other elements. At extremely high pH (12) the test solutions gave evidence of degradation of the peat and lignite, which may account for the lower removal of the radionuclides.

Studies using natural inorganic materials have shown that high pH and phosphate ions are beneficial for increasing the removal of strontium from wastes when natural vermiculite is the sorbent. Further work has shown that the improved removal was due to the precipitation of magnesium and calcium ions by the phosphate at high pH. By replacing the naturally occurring exchangeable ions of magnesium and calcium with sodium, the addition of phosphate was found to be slightly detrimental for strontium removal. However, because sodium offers less competition to strontium than calcium or magnesium, removal of strontium is improved by saturating the vermiculite with sodium. Sodium vermiculite is twice as effective as natural vermiculite for strontium removal when no phosphate is in the waste.

Waste Disposal Research and Engineering: Disposal in Natural Salt Formations. – Operation of the neutralized waste cavity was terminated at the end of 392 days, and the system is now cooling to ambient temperatures. The acid-waste cavity has been emptied. The shape of the cavity was not significantly changed during operation of the experiment. Small experiments designed to observe alteration of "all-salt" cavities have been terminated, and the cavities are being opened and readied for examination.

The radiolysis of aqueous solutions by ionizing radiation is expected to be a major problem in direct liquid waste disposal. It has been found experimentally that by keeping a blanket of hydrogen over the solution the net production of radiolytic gases from simulated Purex-type waste solution can be reduced to a level undetectable by simple manometric measurements. The production of radiolytic gases can also be minimized by fixing the liquid as a surface film on solid salt or by sorbing the liquid on the lattice of sorbent materials like vermiculite or attapulgite.

Waste Disposal Research and Engineering: Disposal of ORNL Radioactive Wastes. – Three field-scale columns (2 in. ID and 10 ft in length) filled with vermiculite were tested for removal of cesium and strontium from simulated ORNL intermediate-level waste.

The first column was filled with potassium-treated vermiculite, and 600 liters of simulated waste (with 3×10^{-3} M KH_2PO_4 added) was passed through it intermittently over a three-week period; distribution coefficients (K_d) of 500 and 140 were obtained for strontium and cesium respectively.

Two hundred liters of simulated waste (with 3×10^{-3} M KH_2PO_4 added) was passed through the second column, which was filled with sodium-saturated vermiculite; K_d's of 150 and 85 were obtained for strontium and cesium respectively. The column was only about 20% saturated at the end of the run, so that the estimates of K_d may be low.

The third column was also charged with sodium-saturated vermiculite, but no KH_2PO_4 was added to the simulated waste. In this column both strontium and cesium appeared in the effluent before 200 liters of solution had passed through the column; K_d's of 55 and 40 were obtained for strontium and cesium respectively.

The addition of phosphate to the intermediate-level waste stream appears to be necessary for adequate removal of strontium, while potassium treatment of the vermiculite or of the waste improves removal of cesium. As found in bench-scale studies, the loading capacity increases with longer operation time.

Waste Disposal Research and Engineering: Process Waste Water and Treatment Plant Studies. – Since chemical precipitation techniques are limited in their ability to decontaminate process waste water, other methods of treatment will be necessary to increase removals. Mineral-filled columns offer promise as a second-stage treatment step. The head-loss characteristics and distribution coefficients (K_d) for Sr^{90} and Cs^{137} of four grades of vermiculite (effective sizes 2.1, 2.0, 0.41, and 0.17) and two grades of rock phosphate (effective sizes 1.1 and 0.25) were investigated. A synthetic solution typical of the effluent from the waste treatment plant was employed. Results show that of the materials tested, the smallest grade of vermiculite has the most attractive chemical properties. Distribution coefficients of 13,000 and 5300 for Sr^{90} and Cs^{137}, respectively, were obtained by slurry techniques. The loss of head across a column of the vermiculite was linear with flow rate, 10 cm of water per foot of bed at 2 ml min^{-1} cm^{-2} and 20 cm of water per foot of bed at 4 ml min^{-1} cm^{-2}. Small laboratory columns (1/2 in. in diameter) filled with 2 in. of vermiculite are now in operation.

Waste Disposal Research and Engineering: Clinch River Studies. – Collection and analysis of river-water samples, collected on a continuous basis in proportion to the flow, were begun November 1, 1960. Sampling stations are located at the intake to the Oak Ridge Water Plant, the intake to the Oak Ridge Gaseous Diffusion Water Plant, and at Clinch River mile 5.0 (0.6 mile upstream from the mouth of Emory River).

Chemical analyses of weekly composites of samples show that the major ionic constituents are Ca, Mg, K, Na, HCO_3, Cl, SO_4, NO_3, and PO_4. The pH averages 7.5. The suspended solid and total solid loads in suspension vary from 6 to 46 ppm and from 120 to 230 ppm respectively.

The weekly composites of samples are separated into suspended solids (larger than 0.5 μ) and solution. Each portion of the sample is radiochemically analyzed. Major radionuclide constituents sorbed by the sediments are Cs^{137}, Ru^{106}, and Co^{60}. Major radionuclide constituents in solution are Sr^{90}, Ru^{106}, and trivalent rare earths.

During August and September 1960, core samples of Clinch River bottom sediments were collected from 19 sections between Clinch River miles 4.7 and 22.5. Each cross section was sampled at 50-ft intervals across the stream by lowering a 3/4-in.-ID by 16-in.-long plastic tube, contained in a weighted stainless steel pipe, into the formation. Following collection, each core was frozen and sectioned into 1-in. segments for gross-gamma counting. A total of 143 core samples, approximately eight cores per cross section, were collected which yielded 870 separate 1-in. segments for analysis.

Analysis of field data collected during sampling shows that the mass of sediment between river miles 22.5 and 16.0 is a small fraction of the sediment deposits downstream from mile 16.0. Comparison of the relative gamma radioactivity for each cross section shows that the bottom sediments at river miles 20.8 and 11.0 have the highest radionuclide concentration.

In general, the relative gamma activity of the core samples shows the same order of distribution as the 1960 Applied Health Physics "flounder" survey. Additional analysis to determine specific radionuclide distribution within the core samples is in progress.

Waste Disposal Research and Engineering: Long-Range Evaluation of Over-All Waste Complex. – In the proposed long-range evaluation of the various methods of ultimate waste disposal being studied by ORNL, an economic feasibility study of each method will be carried out, and a hazards evaluation of each method will be appended later. In this way the cost figures, which can be calculated with some degree of confidence, may be displayed separately from the hazards evaluation, which must be much more qualitative in nature.

As a basis for the study, a 6-ton/day plant will be assumed, processing 1500 tons/yr of uranium-converter fuel at a burnup of 10,000 Mwd/ton and 270 tons/yr of thorium-converter fuel at a burnup of 20,000 Mwd/ton.

The processing steps and disposal methods to be considered are interim liquid storage, pot calcination, interim solid storage, shipment, and ultimate disposal in salt, vaults, shafts, deep wells, hydrofractures, and tanks. For each processing step a curve of mills of cost per kilowatt-hour of electricity will be calculated for each waste type as a function of time since reactor discharge. For each disposal method, costs will be calculated as a function of time interval between reactor discharge and time of ultimate storage. Curves will be constructed for acid and alkaline Purex and Thorex wastes. Conceptual designs of processing steps and disposal methods, which must be made for cost estimation purposes, should be a valuable product of this study.

The study of interim liquid storage has been completed, and costs for this step ranged from 0.001 mill/kwhr (electrical) for 0.5 yr storage to 0.003 to 0.005 mill/kwhr for 30 yr storage.

PHYSICAL RESEARCH PROGRAM

PHYSICS AND MATHEMATICS RESEARCH

Nuclear Measurements for Reactor Design. – The pulse-height response of silicon surface-barrier detectors to fission fragments has been found not to be strictly proportional to fragment energy. The relationship is linear, but extrapolation to zero pulse height yields a finite value for the energy [analogous to the ionization defect for fission fragments stopped in gases; see Phys. Rev. 102, 183 (1956)]. Thus $E = a \times PH + \delta$, where E = fragment energy, PH = pulse height, and δ for these detectors has been termed "pulse-height defect" [Semiconductor Nuclear Particle Detectors, Natl. Acad. Sci. – Natl. Research Council, Nuclear Sci. Ser., Rept. No. 32 (1961)]. Values of δ from 6 to 26 Mev have been found, depending on the resistivity of the silicon and the applied bias voltage.

Scintillation Spectrometry and Instrument Development. – Angular correlation measurements were made on the reaction $Be^9(\alpha,n)C^{12*}(\gamma_{4.43})C^{12}$ by associated-particle time-of-flight techniques. The bombarding energy was 3.35 Mev, in a region which shows no evidence of resonances. The target thickness was approximately 50 $\mu g/cm^2$. Plastic and NaI(Tl) scintillators were used for neutron and gamma-ray detectors. Fast signals from the detectors operated a time-to-pulse-height converter, the output of which was gated into a multichannel analyzer by coincident single-channel outputs from the slow energy-selection system. Measurements were made by movement of the gamma-ray detector with the neutron counter fixed at each of the following angles from the beam direction: 10, 20, 75, and 135°. The angular correlations were observed both in the reaction plane and in two planes perpendicular to it – one parallel, the other perpendicular to the axis of symmetry of the correlation curve in the reaction plane.

High-Voltage Experimental Program: Coulomb Excitation of Levels in Se^{77}. – Levels at 242 ± 6 (doublet), 440 ± 4, and 515 ± 6 kev have been Coulomb-excited with alpha particles of variable energies ranging from 2.1 to 8.0 Mev. The B(E2) for excitation of these levels are (1.9 ± 0.2), (2.6 ± 0.2), and $(0.10 \pm 0.02) \times 10^{-49}\ e^2\ cm^4$ respectively. These are 48, 43, and 2.5 times the independent-particle estimates (taking $R = 1.2 \times 10^{-13}\ A^{1/3}$ cm). Besides the three ground-state transitions, gamma rays with energies of 87, 161, 203, and 283 kev were observed. The excitation curves of the 87- and 161-kev transitions have the same shape as the curve for the 242-kev transitions. Coincidence studies revealed that the 203-kev gamma ray, which de-excites the 440-kev level, is in coincidence with a 242-kev gamma ray but not with the 87-kev gamma ray. This provides new evidence for the previously suggested level doublet at 242 kev (Nuclear Data Sheets, National Academy of Sciences – National Research Council). The angular distributions of the 203-, 242-, and 440-kev gamma rays with respect to the incident beam have been measured for several alpha-particle energies. From these distributions one of the levels at 242 kev must have spin 3/2. The measurements are compatible with a spin assignment of 3/2 or 5/2 for the 440-kev level.

High-Voltage Experimental Program: Afterpulser for the Oak Ridge Pulsed Van de Graaff. – With ion sources presently available [Rev. Sci. Instr. 30, 694 (1959)] it should be possible to obtain milliampere bursts of ions 10^{-9} sec and less in duration without the complications and limitations of bunching. However, fundamental limitations prevent achieving the very short times by terminal pulsing alone when the ion beam is large, or, for that matter, if the ions are heavy [Nuclear Instr. & Methods 7, 245 (1960)]. The limitations are reduced in magnitude if the pulsing is performed at high ion energy.

To accomplish ion burst production at 1 to 3 Mev ion energy and at ion currents in excess of 1 ma without overloading the Van de Graaff, an "afterpulser," synchronized with the bursts produced at the terminal, has been designed and built. The pulser is in routine use for the production of 2×10^{-9} sec, 500-μa bursts of He^3.

Heavy-Particle Physics. – A dE/dx-E counter was developed for the identification of heavy-ion reaction products. A thin proportional counter measures the energy loss, and a silicon surface-barrier counter measures the total energy of the ion. Boron, carbon, nitrogen, oxygen, and fluorine ions are well separated and readily distinguished with this system. The counter is being used to study transfer reactions. Alpha-particle spectra from the nitrogen bombardment of oxygen were measured at 12 angles from 0 to 160°. The angular distribution of the alpha particles is peaked fore and aft, with a minimum at 90°.

The 63-Inch Cyclotron was shut down for three weeks to allow changes to be made in the magnet structure for the stable-isotope separation program.

The 86-Inch Cyclotron: Nuclear Physics. – Internal-conversion-electron and gamma-ray spectra from the electron-capture decay of Eu^{149} (120 days) to Sm^{149} were analyzed. The data indicate excited states in Sm^{149} at 22.5, 277.2, and 350.2 kev which are depopulated by M1 radiation to the ground state. States at 528.6 and 558.3 kev are populated to a lesser extent; these are de-excited to the 22-kev and ground states. Since the parity of Sm^{149} is probably odd, the upper states (22, 277, 350, and 528 kev) are likewise expected to be odd.

The 86-Inch Cyclotron: Applied Physics. – The isotopes Na^{22}, Al^{26m}, Sc^{44m}, Ti^{44}, Co^{57}, Co^{61}, Zn^{65}, As^{74}, Sr^{85}, Te^{123m}, $I^{124,125}$, and Pm^{148} and recoil products from the proton bombardment of Pa^{231} were produced as service irradiations for customers representing four universities (in the United States and India), three commercial processors, and the Chemistry, Physics, and Metallurgy Divisions of ORNL. These irradiations represent 174 hr of bombardment.

The Isochronous Cyclotron. – Occupancy of the cyclotron building (Building 6000) was obtained on January 13, 1961. While the building contractor (Foster and Creighton Company) completes the remaining details, the installation contractor (H. K. Ferguson Company) is installing the cyclotron components.

The cyclotron magnet is now assembled in place in the cyclotron vault, and the heavy turret base (former gun mount) for the analyzing magnet has been installed in the large experiment room. The AVF power

supplies (17) were placed in the utilities room; wiring and plumbing are being installed. The piping and the major components of the vacuum system (accelerating tank, manifold and diffusion pumps, dee-stem house, and piping) were tested and are ready for installation. Designs for the sector coils, harmonic coils, and trimming coils were completed, and the assemblies are being fabricated.

Stable Isotope Development. — The research and experimentation initiated to increase the purity of the separated chlorine isotopes have resulted in the separation of gram quantities of Cl^{37} in the 99 to 99.3% range, as compared with previous values of 92 to 97%.

A device has been constructed to prepare targets for particle accelerators in which the material to be deposited is evaporated and recondensed on the target backing. The heat of evaporation is furnished by electrons emitted by a heated tantalum filament and accelerated through a potential of 5 kv to the feed material. The material is heated and evaporated in a vacuum by the electron bombardment and is immediately recondensed on the cooled target backing, which is located directly above the area of evaporation.

Methods of preparing targets in the calutron for use in particle accelerators are being investigated. Currently the approach is to apply a potential to the collector to decelerate the 35-kev ion beam to a few hundred volts. Experiments using calcium and magnesium deposited on copper and tantalum have been performed, and preliminary results are encouraging.

The new inhomogeneous-field separator now being designed employs an inhomogeneous field with $n = 0.5$. It will operate on a 20-in. radius and collect the particles at 255° from the source. A study of the problems involved in achieving the exact magnetic field shape has led to the decision to purchase an automatic data-logging system. Use will be made of the Hall effect to provide a voltage, determined rapidly and accurately by the data system, that is equivalent to the field strength. This instrumentation will permit the thousands of field measurements and corrective data associated with the accurate shaping of a magnetic field to be processed with an electronic computer. The magnetic-field measuring system will also be used in a program to evaluate and improve the linear magnetic focusing system of the calutrons.

Special Separations. — Cleaning and decontamination operations were completed on the eight calutron tanks and the associated track area which are to be used in the new facility for separating plutonium isotopes. This work was necessary to reduce the contamination to an acceptable level prior to the beginning of track alterations by an outside contractor.

Arrangements are being made for the conversion of approximately 10 kg of depleted uranium to metal foil. The uranium foil, containing about 300 ppm U^{235}, will be used to fill special requests for this type of material.

CHEMISTRY RESEARCH

Equipment Decontamination. — A mixture 0.3 M in ammonium oxalate and 1 M in H_2O_2, pH 5, at 95°C decontaminated cold-rolled carbon steel

(ASTM A 109) by factors of 25 to 10^4, depending on the isotope and the method by which it had been applied to the steel. The steel was corroded at a rate of 0.007 mil/hr. The reagent was not effective on rusted or scale-coated steel. A UO_2 slug jammed in a charger was 50% dissolved in 1 hr, 99% in 2 hr, and 100% in 2.5 hr. A carrier contaminated with UO_2 from a ruptured slug could therefore be decontaminated with this reagent.

Analytical Chemistry Research. – Spectral studies on molten fluoride salts have been extended to molten solutions of sodium fluoride at temperatures as high as 1100°C. The absorption spectra of praseodymium(III) fluoride and neodymium(III) fluoride in molten sodium fluoride were found to be in agreement, in general, with the spectra of these same elements in aqueous solutions. Spectrophotometric measurements at temperatures in excess of 1000°C have been made possible by the installation of a graphite resistance heater in the high-temperature cell assembly. Somewhat novel electric contacts were provided in which platinum wires are held in place by silver solder in cavities drilled into the top of the graphite heater. At temperatures exceeding the melting point of the solder, the confined molten pool of solder serves as a conductor of the current. No deleterious effects have been observed as a result of vaporization of components of the solder after numerous thermal cyclings to temperatures in excess of 1000°C.

The resonance, or atomic, absorption lines of sodium, potassium, and rubidium vapor have been observed when fluoride salts of these elements are melted in the graphite furnace; however, when a platinum wire heater is used to melt rubidium fluoride, no resonance absorption lines are observed. From these observations, it is postulated that alkali fluoride vapor reacts with heated graphite to produce alkali metal vapor. Further effects and ramifications of this reaction are being investigated.

Organic Chemistry. – As part of a series of measurements of the effect of distant deuterium atoms on the ionization constants of organic acids, the ionization constant of α,α,α-trideuterioacetic acid (CD_3COOH) was determined. The ratio of the ionization constants of the undeuterated and deuterated acids at 25° was found to be $K_a(H)/K_a(D) = 1.05 \pm 0.01$. The ionization constant is therefore lowered 1 to 2% per deuterium atom. This result, which has been confirmed by conductivity measurements made elsewhere, refutes earlier measurements of others.

Radiation Chemistry. – Yields were measured for decomposition of various nitrate crystals by 3.4-Mev alpha particles, cobalt gamma rays, and ultraviolet light at several temperatures. The increase in yield with increase in linear energy transfer (LET) along the path of the particle (alpha particles compared with gamma rays or ultraviolet light) parallels the increase in yield with increased temperature for decomposition by gamma rays or ultraviolet light. This suggests that thermal effects along alpha-particle tracks ("thermal spikes") are an important factor in the radiolysis. For example, the 100-ev yield of nitrite, $G(NO_2^-)$, for decomposition of $NaNO_3$ by gamma rays increased from 0.25 at 30°C to 1.0 at 150°C (the yield for photolysis increased by a factor of 3); the yield for decomposition by alpha particles was 1.3 at 25°C and 1.1 at 150°C. On the other hand, yields for decomposition of KNO_3, $CsNO_3$, and $Ba(NO_3)_2$, which are less sensitive to change in temperature for decomposition by

gamma rays or ultraviolet light, are also relatively insensitive to change in LET. For example, for decomposition of $Ba(NO_3)_2$ by gamma rays the yield changed from 1.8 at 30°C to 1.6 at 150°C; for alpha particles the yield was 1.6.

Chemical Physics: Microwave and Radio-Frequency Spectroscopy. — The electron spin resonance spectrum seen in gamma-irradiated single crystals of $NaNO_2$ was attributed earlier to oriented molecules of NO_2 produced in the $NaNO_2$ lattice by the radiation. The measured hyperfine interaction parameters were satisfactorily accounted for by unpaired electrons occupying N and O orbitals of NO_2. A surprising result of this interpretation was that the electron spin density associated with N is negative even though its numerical value is high. This interpretation depended upon the assumed orientation of NO_2, which was taken to be the same as that of NO_2^- in the perfect $NaNO_2$ lattice. An alternative orientation which is consistent with the data, the symmetry of the lattice, and the space available therein is obtained by rotating NO_2 90° about the line of intersection of the two mirror planes of NO_2 (or NO_2^-). A more satisfactory interpretation can be made on the basis of the rotated orientation. A cursory analysis indicates that electron spin density will then be positive on N as well as on O.

The complicated spectrum found in an electron spin resonance study of single crystals of $KClO_3$ irradiated with Co^{60} gamma rays at low temperatures was partially interpreted. Spectra characteristic of at least four different trapped free-radical species were seen. Two of these are especially interesting, in that hyperfine interactions from single chlorine nuclei are present.

In electron spin resonance studies of single crystals of KNO_3 irradiated with Co^{60} gamma rays, spectra from at least four different free-radical species were seen. At least two species, probably three, were observed with large hyperfine interactions from single nitrogen nuclei.

High-Temperature and Structural Chemistry. — Solutions of neodymium in molten neodymium trichloride were found to exhibit very little or no electronic conductance, quite distinct from the solutions of cerium in cerium trichloride, where the conductance of the saturated solution is more than 70% electronic. This difference reflects the much higher stability, in the liquid as well as in the solid state, of divalent neodymium, which is known to form a stable, insulating solid $NdCl_2$.

Early measurements of the electrical conductivity of solutions of cadmium in cadmium chloride (Aten, 1910) were confirmed. Together with other evidence, the nonelectronic conductance is best interpreted in terms of the formation of a relatively stable $(Cd_2)^{2+}$ ion.

A study of the effect of dissolved salt on the electrical resistivity (ρ) of liquid metals was initiated with measurements on solutions of potassium iodide in potassium metal. At 700°C, ρ increased linearly with KI mole fraction (N) up to N = 0.25, within ±2.5% according to $\rho = 920 \times 10^{-6}\ N + 58 \times 10^{-6}$.

Chemical Separation of Isotopes. — The solubility of BF_3 in dimethyl ether·BF_3 and diethyl ether·BF_3 as a function of temperature was determined as shown below:

Temperature (°C)	Moles of BF_3 per Mole of $(CH_3)_2O$		Moles of BF_3 per Mole of $(C_2H_5)_2O$ at 400 mm Hg
	400 mm Hg	760 mm Hg	
30	1.027	1.038	
22	1.034	1.050	1.047
6	1.051	1.093	1.073
− 9	1.082	1.138	1.114
−19	1.103 (−16.50)	1.201	1.163

Redetermined values for the single-stage separation factor between BF_3 and triethylamine·BF_3 are 1.023 (30°C), 1.021 (40°C), and 1.0195 (50°C).

Transuranium Elements: HFIR Target Development. – Fabrication of HFIR targets by compacting aluminum powder and oxides of plutonium, americium, or curium is planned. In studies on the stability of a 10 vol % PuO_2–Al compact to heating at 400, 500, and 600°C for 100 hr, the volume increased less than 0.8%. This is approximately the volume increase for pressed aluminum powder containing no PuO_2. These results indicate that PuO_2-Al cermets are stable and will be satisfactory for HFIR targets.

Transuranium Elements: Process Chemistry. – Development of solvent extraction methods for separating actinide and lanthanide elements was continued. For the system 30% triisooctylamine–10% isooctyl alcohol in Amsco vs 11 M LiCl, the order of actinide extractability was Cf > Am ≧ Cm, with a californium-americium separation factor of 20. Lanthanide extractability increased from lanthanum to europium and then decreased to lutetium. The americium-europium separation factor was about 100. In a countercurrent extraction run with seven scrub and seven extraction stages, americium tracer recovery was greater than 99.9%, with decontamination from macro rare earths by a factor greater than 10^4.

Separation of transplutonium elements by extraction into the half ester of 2-ethylhexylphenylphosphoric acid (2-EHΦPA) in xylene from dilute hydrochloric acid was investigated. The californium-curium separation factor with 1.0 M 2-EHΦPA in xylene vs 1.5 M HCl was approximately 100. A countercurrent extraction run with three scrub and three extraction stages gave (98 ± 1.3)% californium recovery and decontamination from curium by a factor of 10^3.

In tests on materials for tank storage of acid wastes from the transuranium facility, titanium-45A and Haynes 25 were corroded at rates of 0.1 mil/month in 500 hr exposure. Hastelloy C showed a uniform attack of 0.3 mil/month, Hastelloy F showed a localized attack of 10.9 mils/month, and types 304L and 309SCb stainless steel showed rates greater than 500 mils/month in 25 hr exposure. In 500 hr exposure to oxygenated, neutralized wastes all these materials showed rates greater than 0.1 mil/month, and SAE 1045 carbon steel scaled rather badly and showed somewhat indeterminate rates between 0.3 and 1.6 mils/month.

METALLURGY AND MATERIALS RESEARCH

Deformation of Crystalline Solids. – The new boiling calorimeter has been finished and tested. It is designed to measure the energy stored in metals and alloys during deformation.

Alloy Theory Research. – The conclusion may be drawn that size factor effects introduced during alloying of α-zirconium are indeed isotropic on the zirconium lattice dimensions. This is based on the equal and near-zero effects of both titanium and hafnium on the axial ratio during alloying, and on equal axial ratio effects of indium and gallium. The atom misfit is small in Hf-Zr and In-Zr but appreciable in Ti-Zr and Ga-Zr. For the solutes Ag, Cd, In, Ga, Sn, and Sb the axial ratio is mainly affected by a term proportional to the number of S-P valency electrons of the solute, as reported earlier, but in addition a smaller positive term occurs that is related to a linear restoration of the axial ratio distortion in pure α-zirconium.

Fields of 4400 gauss have been reached in a superconducting niobium solenoid designed for paramagnetic salt cooling. Dimensions of the core are 1-1/2 in. ID by 6 in. long.

X-Ray Diffraction and Metallographic Research. – To determine whether the change in electrical resistivity reflects the change in the state of local order in the alloy Cu-16 at. % Al, a single crystal was heated for 1 hr at various temperatures from 150 to 525°C and water-quenched. X-ray diffuse scattering measurements were made at selected positions in reciprocal space to ascertain the change in the intensity of the short-range order peaks. It was found that the change in electrical resistivity did indeed reflect the change in the state of order in the alloy. This information will be used to choose a heat treatment for the starting condition of this alloy in order to determine the effect of neutron irradiation on its local order.

A high-temperature diffraction study of Cu-32 at. % Au filings has been started in order to recheck the existence or absence of classical two-phase fields between ordered, anti-phased, and disordered single-phase regions. A specimen has been disordered at 440°C and then cooled to 340°C during the course of a three-month period. At temperatures of 357°C and below, the specimen has been maintained at the temperature of interest for at least 336 hr in order to attain equilibrium. An anomalous broadening of certain high-angle fundamental reflections appears at 351°C and persists to 340°C. The effect is accompanied by an increase in the thermal expansion coefficient. No satellited superstructure reflections characteristic of the anti-phased structure are observed in this temperature interval. It has been tentatively concluded that the effects are produced by a two-phase condition of the alloy between 340 and 350°C.

Single crystals of $Ca_2Be_3O_5$ have been grown and are currently being ground to approximately spherical geometry prior to observation of diffraction data. These crystals are the first obtained in a simple mixed oxide system containing BeO. Crystals of $Cr_3(CrF_6)_2$ have been obtained from a solid-state chemical preparation. They are probably twinned and are also too small to permit a complete structure determination to be made.

Refinement of the structure of $LuMnO_3$ based on the space group $P6_3cm$ has been virtually completed. Although a reasonably good agreement factor (13% for F^2) was obtained, the odd behavior of the β_{22} temperature factors of two sets of oxygen atoms located on mirror planes has not been explained.

Metal Surface Studies. – The investigation of the oxidation of tantalum was continued. It was definitely established that the platelets of oxide which develop in the metal at 500°C form parallel to (320) planes of the metal. Such "platelets" of oxide were observed to form parallel to the metal surface on single-crystal specimens with a (320) orientation. The oxide consisted predominantly of a highly oriented layer of a suboxide, probably Ta_4O, and a polycrystalline layer of TaO. Assuming the oxide at the oxide-metal interface to be Ta_4O, the application of the theory of the invariant plane for martensitic transformations to the Ta-Ta_4O system yielded the (320) as the predicted habit plane for the metal.

Fundamental Investigation of Radiation Damage in Solids: Investigation of F and M Centers in KCl. – Measurements of the temperature dependence of the magnetic susceptibility of ionic crystals permit determination of the total number of point imperfections containing unpaired electrons. This technique and optical absorption measurements are being used to investigate F and M centers in KCl. Optical bleaching of samples that originally contained only F centers causes a decrease of these and a production of M centers. The decrease of the number of magnetic centers has been found to be greater than the decrease in the optical absorption of the F band, indicating that the M center is not a one-electron center and, furthermore, that it absorbs not only in the M band, but also at the same wavelength as the F band.

Fundamental Investigation of Radiation Damage in Solids: Radiation Effects on Deep-Level Impurity States in Germanium. – The change in electronic properties of semiconductors during irradiation and during subsequent annealing depends critically on the type and energy of the initial bombarding particle and on the initial type, carrier concentration, defect concentration, and chemical impurity in the material. Substitutional tellurium acts as a double donor in n-type germanium. Extensive experiments have indicated that introduction of isolated Frenkel-type defects by Co^{60} gamma irradiation and any subsequent removal of such defects by vacuum-annealing at 450°C have no apparent effect on the concentration and donor action of the tellurium; however, the introduction and subsequent removal or rearrangement of fast-neutron-induced lattice defects under the same annealing conditions totally cancel the apparent concentration and donor action of the tellurium. This effect has not previously been observed with normal donor or acceptor chemical impurities, nor was it observed in germanium that was doped with copper acceptor impurities.

Preparation of Pure Single Crystals. – In order to prepare Li^7F single crystals more nearly free from strain than the crystal previously reported (ORNL-3062, p 23), two major changes in the crystal-growing apparatus were made. The linear rate of lowering the crucible was reduced to 1/30th that used in the previous test, and a thick-walled nickel crucible capable of withstanding a vacuum at high temperature was substituted for the thin-walled nickel container previously used. In addition, a vacuum system

was installed to ensure the absence of contaminating gases during the melting and crystal growing periods.

The very pure Li^7F previously prepared was found to contain, nevertheless, an undesirable trace of calcium and magnesium. Magnesium is an impurity which does not separate from the LiF on crystallization but forms solid solutions. A liquid-liquid extraction procedure was demonstrated on a 100-g scale to remove magnesium satisfactorily.

CONTROLLED THERMONUCLEAR RESEARCH

Arc Research. – In the gas arc facility, deuterium arc operation has been achieved with a gas input of 1.5 cc/sec, one-third the flow in previous operation. Ambient (corrected) pressure was 9×10^{-6}, a factor of 2 better than has been achieved before. The ionizing efficiency of anodes currently in use is sufficiently high that there is no need for isolation of the portion of the vacuum chamber which contains the anode during normal arc operation.

The ion temperature in the arc is indicated by Doppler studies to lie between 15 and 20 ev. This is true for five different ion species observed, and hence thermal equilibrium between ions is inferred. The apparent electron temperature, based on intensity ratios of certain spectral lines, is 70 ev in the region of maximum arc density.

DCX-1 Facility. – The stationary gas arc anodes using cooling water flow in swirl and vortex patterns were tested. Both failed at hydrogen gas bleed rates that were higher than those at which failures occurred when using the rotating anodes described in previous reports. Now being fabricated is a mild steel structure to surround the arc immediately in front of a rotating anode. The magnetic-field distortion produced by this structure may significantly increase the diameter of the arc as it impinges upon the anode.

Tests of the High Voltage Engineering Corporation ion source were continued. Fifteen- to 20-ma H_2^+ has been accelerated to 600 kev, but the requirement of a proper beam focus inside the DCX has limited the current actually injected to about one-quarter of this total. The focal properties of the equipment are being investigated to determine what changes can best be made to increase the injected current.

Scans across the undissociated portion of the H_2^+ beam made after the beam had emerged from the inner vacuum region have shown that it has a considerably larger divergence than was previously thought. This divergence has necessitated a change in the planned method for disposing of this exit beam. The beam was to have been passed through the bottom of the tank assembly and then directed onto a target inside a large Varian VacIon pump. The wide divergence requires that the beam target be much closer to the aperture through which the beam emerges from the inner vacuum region. It is now planned to stop the beam near this aperture on a rotating water-cooled copper drum. Pumping is to be provided by continuously evaporating titanium onto this drum.

Larger Magnetic Facility. – The first phase of the engineering design of the DCX-2 facility was completed by Y-12 Engineering on February 1.

This phase included site preparation, control room layout, structural supports, and mechanical design of the larger machine parts. On February 1, the construction contractor began work in the DCX-2 area. The second, and final, phase of the design is scheduled to be complete on March 20 and is proceeding on schedule.

Designing of the magnet coils, performed by the Thermonuclear Division, is complete. The "outboard booster" coils have been constructed and are being prepared for testing in conjunction with the test of the ion source and the magnetically shielded injector snout. Contracts have been let to outside firms for the construction of the other coils, and delivery is scheduled before June 1.

Vacuum. – Several vapor-deposited metal films (titanium, molybdenum, chromium, and lithium) have been tested for their ability to pump hydrogen. In this evaluation the net pumping speed is a practical measure of the usefulness of the material as a getter of hydrogen. Titanium gave the greatest net increase in pumping speed. The pumping speed of molybdenum was down by an order of magnitude. It appears that its surface is easily contaminated, which reduces its ability to pump hydrogen except in very clean systems. Chromium gave only one-third the pumping speed of molybdenum. The pumping speed of lithium was zero in this test.

In the large-scale pump test facility a comparison of pumping rates by a continuously evaporated titanium film for several gases other than hydrogen and deuterium has been made. Oxygen, nitrogen, carbon monoxide, and carbon dioxide are readily pumped by the titanium film during continuous evaporation. Argon, methane, and helium are not significantly pumped by a continuously evaporated titanium film.

BIOLOGY AND MEDICINE PROGRAM

BIOMEDICAL PROBLEMS IN ATOMIC ENERGY OPERATIONS

Internal Dose Estimation. – A need to evaluate and interpret the $(MPC)_w$ for Rn^{222} arose when recent measurements of Rn^{222} concentrations in spring waters supplying drinking water to some local areas indicated higher levels than in river waters surrounding the area. These concentrations were higher than the $(MPC)_w$ recommended by NCRP in Handbook 52. Although these levels can probably be considered as natural background, and the publications of ICRP and NCRP stipulate that their recommendations on permissible exposure do not apply to the natural background, there is nevertheless a need for an $(MPC)_w$ for Rn^{222} in natural waters because of the increased emphasis on estimating the total radiation dose received by human beings.

In order to calculate this $(MPC)_w$, experimentally measured values of f_2^i, the ratio of the total amount of Rn^{222} in the ith organ to the total amount in the whole body, and experimentally measured values of the fraction of the ingested radon undergoing disintegration in the body are required. The experimental data obtained by Nussbaum (1957, UR-503)

in an investigation of the distribution of radon in organs and tissues of rats after inhalation of Rn^{222}-contaminated air provided estimates of f_2^i. No direct experimental data on the fraction undergoing disintegration in the body could be found. It was assumed that 0.1 was a reasonable estimate. Employing this value, together with the f_2^i values, the calculated $(MPC)_w$ is 16×10^{-6} μc/cc for 168 hr/week exposure of occupational workers and 0.5×10^{-6} μc/cc for 168 hr/week exposure of the population at large.

Very recently some experimental data on a human subject were obtained by University of Utah investigators, and it is indicated that the above $(MPC)_w$ values are too conservative by a factor of ~20. Their measurements of the fraction undergoing disintegration in the body suggested 0.004 to 0.005 rather than 0.1, as was assumed. This one measurement can hardly be considered as adequate. More measurements on subjects of widely different body weights and different ages are required.

Waste Disposal Research and Engineering: Evaluation of Soil Disposal. – Work is in progress to identify and define all the contamination sources at ORNL and to determine the amount, type, and state of activity contributed to White Oak Creek from each source. A series of temporary sampling stations is being installed along White Oak Creek and its tributaries to assess the activity associated with the burial grounds and general and local fallout and to aid in determining the means by which radioactive materials move downstream. Samples will be taken at these stations with a portable, battery-operated, scoop-type device that will sample in proportion to the flow across a V-notch or rectangular weir.

Representative samples taken from the laundry effluent for November and December 1960 showed discharges to White Oak Creek during this period as follows (specific radionuclides with millicuries of radioactivity are given): strontium, 2.6; Cs^{137}, 4.0; Ru^{106}, 3.4; Zr-Nb^{95}, 8; Co^{60}, 1.9; Ce^{144}, 3.3; trivalent rare earths, 2.7; U^{233}, 0.1; $Pu^{239,240}$, 0.3; and Pu^{238}, 0.03. A portable weir box is being fabricated for use with one of the scoop-type sampling devices for taking continuous and proportional samples from the sewage treatment plant effluent and possibly from the LITR retention pond effluent.

Ecology and Marine Studies: Radioactive Waste Areas. – Continuation of previously reported studies of radioisotope bioaccumulation by small mammals from the White Oak Creek drainage system has revealed that muskrats are receiving the highest radiation doses from ingested radioelements. Calculated whole-body doses due to internally deposited radioelements (contents of GI tracts excluded) for a male and female muskrat are: female, 23.7 rads/week (90.4% due to Sr^{90}); male, 15.1 rads/week (94.0% due to Sr^{90}). Significant doses may also result from (1) material in the GI tract, for example, the GI tract of the male muskrat contained 67.6 μc of Co^{60} and 12.4 μc of Sr^{90}, (2) swimming in radioactive water, and (3) burrowing in radioactive mud and soil. The two muskrats were trapped from a population inhabiting the ORNL settling basin for low-level radioactive liquid wastes. The high dose rates of the muskrats suggest that they would be an excellent choice for studying radiation effects on wild mammals in areas contaminated by radioactive waste disposal. It is also apparent that the spread of radioactivity by muskrats migrating into uncontrolled areas should be investigated on the basis of the extensive use

made of muskrat pelts by the fur industry and the use of muskrat carcasses for food consumption.

Ecology and Marine Studies: Forest Ecology. – Concentration factors and release coefficients of Co^{60} and Cs^{134} for several microorganisms were determined in the course of an over-all study of biological cycling of radioisotopes in aquatic and terrestrial environments. After three days in shaking cultures, Co^{60} concentration factors relative to the nutrient medium were as follows: streptomycete, 34; fungi, 2 to 23; bacteria, 4 to 34.

For the common fungus *Trichoderma viride*, the Co^{60} concentration factors were lowest (12) near neutral pH, compared with 48 at pH 4 and 22 at pH 8 – probably due to changes in colloid size. Concentration of Cs^{134} was not affected markedly by pH and was about one-third the concentration of Co^{60} at neutrality. For both nuclides the greatest concentration factor was obtained after 7 to 16 days and decreased after prolonged growth. Concentration factors were not affected by a wide range of initial levels of activity (600 to 80,000 μμc/ml).

Release of Co^{60} and Cs^{134} from intact *Trichoderma* fungus mycelium was only 8 to 14% and 11 to 23%, respectively, depending on the leachant in the shaking culture. But drying or freezing of mycelium before leaching increased the release to 50 or 98% for Co^{60} and 87 or 90% for Cs^{134}. This difference suggests that previous uptake had been more physiological than physical. Drying or freezing at the soil surface, where most fungi are active, will tend to release quickly absorbed isotopes for subsequent movement in the environment.

Ecology and Marine Studies: Clinch River Program. – Tubificid worms are the most abundant macroscopic organisms living in the radioactive bottom sediments of the Clinch River. They feed with their anterior ends in the mud and with their posterior ends in the overlying water. There are weak bottom currents, created by differences in water density in Watts Bar Reservoir. Feeding activity of the worms and density currents might contribute to the downstream transport of radioactive bottom sediments. A mathematical model was used to evaluate the movement of bottom sediments by the worms. The parameters included in the model were current velocity, height of worm tail above the mud surface, rate of fecal pellet fall, rate of feeding, depth of burrowing, diameter of feeding worm, mud density, and worm population. The average population in the Clinch River (one worm/10 cm^2) would take 3.3 years to move the volume of sediment from 1 cm^2 to average depth of feeding, downstream a distance of 9.18 cm. This rate of movement is insignificant. Populations of 100 worms/cm^2 may occur in areas of high organic pollution. Such a population could effect a movement 800 m downstream during one half-life of Cs^{137}, an isotope which is readily adsorbed on bottom sediments. It is obvious that sediment transport by flowing water is of much greater importance than movements by the worms.

TRAINING, EDUCATION, AND INFORMATION PROGRAM

Biology and Medicine Program. – A two-week course was given for 16 students from the Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio, in September 1960.

A 60-hr (ten-week) course was given for the Oak Ridge School of Reactor Technology beginning on November 14, 1960.

The radiological physics course at Vanderbilt University began on February 6 with 26 students in attendance. There are 21 AEC Fellows, 2 foreigners, and 3 others.

A series of 22 lectures is being given to ORNL personnel as part of the over-all Radiation Safety Training Program.

One employee of the Martin Company, two Italians, one Indian, one Greek, and one Thai have been with the Health Physics Division for training in applied health physics.

ISOTOPE DEVELOPMENT PROGRAM

Radioisotope Research and Development. – The absorption in lead of bremsstrahlung x radiation from an $Sr^{90}TiO_3$ pellet in the proximity of Hastelloy C was measured to obtain data for the design of a biological shield. The radioactive pellet contained 1000 curies of Sr^{90} in 65 g of titanate powder which had been compacted and sintered to a specific gravity of ~4.5. The only radioactive contaminant in the pellet was Ce^{144}, which amounted to 0.305 curie at the time of the absorption measurements. The measured tenth value layer for the absorption of the more energetic components of the x-ray continuum averaged 1.60 in. of lead. This absorption coefficient is characteristic of a 1.25-Mev gamma ray. The radiation through 2.6 in. of lead at 1 m from the $Sr^{90}TiO_3$ pellet was 10 mr/hr. The radiation from the Ce^{144} contaminant was calculated to have contributed 0.25 mr/hr at 1 m through the 2.6 in. of lead. The data will be published in ORNL CF-61-1-25.

Several factors were investigated which are important in the conversion of strontium into sintered strontium titanate pellets. This study included the effect of variable amounts of calcium and barium impurities, the mole ratio of alkaline earths to TiO_2, the calcining temperature used to convert a mixture of alkaline-earth oxalates and TiO_2 to the titanates, the pellet pressing technique, and the sintering temperature. It was found that satisfactory pellets could be made from $SrTiO_3$ containing up to 15% calcium and/or barium in mole ratios of alkaline earth to TiO_2 in the range 0.95 to 1.15. Calcining temperatures between 1000 and 1200°C and sintering temperatures between 1400 and 1450°C were satisfactory. Pellet pressing technique was the most important factor in obtaining sintered pellets having good physical properties.

Radioisotope and Stable Isotope Production. – Suitable charge materials were prepared for the separation of the isotopes of calcium, chlorine, dysprosium, nickel, potassium, ytterbium, and zirconium in the isotope separation program. Small quantities of calcium, magnesium metal, and tin tetrachloride were supplied for the decelerating beam

study, in which an attempt is being made to make isotopically enriched targets directly in the separator.

The chemical recovery of osmium from calutron components used in the recent isotopic separation of the element is continuing. Associated with this has been the recovery and purification of 341 g of osmium metal, 99.9% chemically pure, for particle accelerator use. This metal as obtained from the vendor was 96 to 98% pure.

An estimated 2701 g of separated isotopes was collected. Elements processed included calcium, nickel, dysprosium, ytterbium, zirconium, chlorine, and potassium. Separations were performed in the four calutrons in Building 9731 and in the 24 calutrons associated with the expanded isotope separation facilities in Building 9204-3. Of these 24 calutrons, 8 represent the third segment of track 5 and were put into operation for the first time on January 5, 1961.

Separated isotopes of Cd^{112}, Ca^{46}, Os^{184}, and Pb^{208}, one returned lot of Cl^{35}, and one reclaimed lot of Zr^{90} were recovered, refined, and prepared for inventory.

The Pb^{208} samples were products of a second-pass separation in which Pb^{208} previously enriched in the calutron was used as charge material. The following table gives the initial isotopic distribution of the charge material and two analyses on the best sample of the final product:

Isotope	Charge	Product*	
		Assay No. 1	Assay No. 2
204	<0.05%	2 ± 1 ppm	2 ± 1 ppm
206	0.71%	48.6 ± 1.0 ppm	48.2 ± 0.8 ppm
207	0.44%	58.2 ± 0.6 ppm	58.0 ± 1.5 ppm
208	98.98%	99.98912%	99.98918%

*ORNL Referee Analyses Laboratory.

These analyses of the product represent two sample loadings, and results were calculated on the 95% confidence level. There was 195 mg of this sample; there was also 95 mg of better than 99.9% material.

Currently in varying stages of recovery and refinement are new enriched isotope lots of cadmium, calcium, chlorine, gallium, iridium, lead, iron, nickel, potassium, tantalum, and ytterbium.

Six special items were prepared for Los Alamos from enriched Pb^{206}, Pb^{207}, and Pb^{208}, each in the form of a cylinder. Three of these were 1-3/4 in. high with an outside diameter of 0.629 in. and an inside diameter of 0.060 in., and they weighed slightly more than 100 g each. The other three samples were formed into cylinders 1-1/8 in. in diameter and 3/4 in. high that weighed approximately 140 g each.

Two inventory lots of Ca^{46}, in the form of CaO with an isotopic content of 3.41 and 8.46% Ca^{46}, were diluted with high-purity Ca^{40} to 0.40 and 0.51% Ca^{46} respectively.

A zirconium metal wire weighing 926 mg was fabricated from an enriched isotopic sample of Zr^{90}. The procedure, essentially that described by DeBoer and Van Arkel, reduces the oxide to finely divided powder by use of calcium chloride and sodium metal. The powder reacts with iodine under vacuum in the presence of a hot tungsten filament and produces a deposit of zirconium on the filament. This can be rolled into thin films suitable for accelerator targets.

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