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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

APRIL 1966

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OAK RIDGE NATIONAL LABORATORY

STATUS AND PROGRESS REPORT

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DATE ISSUED

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

STATUS AND PROGRESS REPORT

APRIL 1966

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

REACTOR DEVELOPMENT

GAS-COOLED REACTOR PROGRAM

Fast Gas-Cooled Reactor Irradiation Tests. — Irradiation tests are being performed in the ORR poolside facility to support the development of fuel elements for a fast gas-cooled reactor. The test capsules are irradiated in a predominantly thermal-neutron spectrum, and data are obtained principally on fuel-cladding interactions. The General Atomic fast gas-cooled reactor design (GCFR) is used as a reference in the selection of test parameters, and the test specimens are supplied by General Atomic.

Irradiation of the third capsule, designated P4B-3, was completed as scheduled on April 10, 1966. This capsule contains two fuel element specimens: one of the fuel-supported clad type and one of the sodium-filled "flexcan" type. It was the first irradiation test of a flexcan fuel element. The specimens were irradiated for one ORR cycle at maximum cladding temperatures of 660°C and linear heating rates of 16 kw/ft for the "flexcan" element and 13.5 kw/ft for the fuel-supported element. During irradiation the specimens were subjected to 31 thermal cycles (660 to 300°C) and 11 external pressure cycles (1000 to 100 psig). The primary system blanket gas was monitored for fission product activity; since none was found, the fuel specimen claddings were apparently still intact. Postirradiation examinations are under way.

The fourth capsule in this series, designated P4B-4, contains two fuel specimens similar to those in capsule P4B-3. Irradiation began April 16, 1966, and test conditions will be approximately the same as for capsule P4B-3, except that cladding temperatures will be somewhat higher.

GCR-ORR Loop 1 Irradiations. — The irradiation of experimental fuel assembly 15 was completed. The assembly was retracted from the reactor core on April 1, 1966, and removed from the test loop on April 10 after an irradiation period of about nine months. The test fuel element consisted of a mixed bed of loose pyrolytic-carbon-coated particles of different sizes and compositions [$(Th,U)C_2$, $(Th,U)O_2$, and ThO_2] in two

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graphite containers; the upper container was vented to the loop coolant. The maximum fuel temperature was held at 1400°C throughout the test, except during periods of reactor shutdowns, and the burnup achieved was 12.1% fission of initial heavy-metal atoms. The ratio of fission-gas release rate to birth rate (R/B) was initially quite low – about 10^{-6} for krypton and xenon isotopes – but increased rapidly after 10% heavy-metal burnup and was about 10^{-4} at the end of the irradiation period.

Following irradiation of experimental assembly 15, the in-reactor portion of the loop, which has been in service about five years, was removed. It will be examined destructively for evidence of deterioration from unexpected mechanisms. The emergency-power motor-generator set and the loop compressors are being overhauled, and several other maintenance tasks will be performed while the existing spare in-reactor section is being modified and prepared for installation. An analytical study was initiated to determine the feasibility of increasing the operating pressure of ORR loop 1 to a value higher than the presently approved 300 psi.

Mathematical Simulation of Coated-Particle Behavior. – We studied the parameters in our mathematical model of coated-particle behavior in the light of postirradiation data now becoming available from the first ETR X-basket experiment. From the measured burnup of those lots of coated particles with only a small percentage of failures, we hope to arrive at reasonable estimates of the actual burnups at failure. Comparison of these values with those predicted by our model under various assumptions should permit us to fix more accurately the poorly known parameters. At present, it appears that experimental results can be explained by assumption of a 30,000-psi rupture stress for all outer coating materials, 100% release of fission gas from the fuel particles, and 25% densification of the inner low-density laminar or isotropic coatings. A somewhat higher percentage densification may apply for the porous inner coatings.

REACTOR FUELS AND MATERIALS

Behavior of High-Temperature Materials Under Irradiation. – Postirradiation examinations of specimens of monocarbides of Ti, Zr, Ta, W, and Nb, extruded translucent Al_2O_3, and Y_2O_3 irradiated in two uninstrumented assemblies at ~200°C are now in progress. Tungsten carbide specimens were more resistant to neutron irradiation than were the other carbides over the dose range (~0.5 to 7.2×10^{21} neutrons/cm^2, >1 Mev) of these experiments. Only minor gross damage occurred in hot-pressed and slip-cast tungsten carbide, even at the highest doses.

The volume expansions of carbide specimens irradiated to doses up to 7.2×10^{21} neutrons/cm^2 (>1 Mev) were as follows: W, ~0.5%; Ti, Ta, Nb, >2%; Zr, ~4%. The expansions were approximately the same over the entire neutron dose interval 0.5 to 7.2×10^{21} neutrons/cm^2 (>1 Mev) in each case.

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The volume expansion of translucent Al_2O_3 specimens increased nearly linearly from 1.3 to 2.5% over the dose range 0.5 to 3.7×10^{21} neutrons/cm^2 (>1 Mev). At a higher dose (~6.5×10^{21} neutrons/cm^2, >1 Mev), the volume expansion was 5%.

Yttrium oxide expanded by ~0.5% upon exposure to doses over the range 0.5 to 3×10^{21} neutrons/cm^2 (>1 Mev), and by ~1.5% after exposure to much higher doses (6 to 7×10^{21} neutrons/cm^2, >1 Mev).

Fuel Element Development. — We are investigating the intermetallic compound UAl_3 dispersed in aluminum for application in high-performance research reactors. Studies on the preparation of UAl_3 included both arc melting of the elemental metals and the solid-state reaction of uranium hydride with aluminum. The latter technique yielded essentially stoichiometric UAl_3 and was selected to prepare the material for irradiation performance testing.

Fuel plates containing Al—30% UAl_3 compacts were successfully fabricated and were assembled into prototype fuels for irradiation testing in the ORR. A visual examination of the elements after approximately 30% ^{235}U burnup showed no evidence of damage.

Although no difficulties were encountered in processing the UAl_3-bearing ORR elements, considerable problems arose from the subsequent processing of larger quantities of UAl_3. Unlike the 1.1% normal growth of the ORR compacts during degassing at 570°C, severe swelling of 14% was observed in compacts containing UAl_3 prepared in larger quantities. Lowering the outgassing temperature to 525°C, at which the volume changed only 4%, circumvented the problem, but the source of the difficulty remains unknown. We are actively evaluating this phenomenon.

FUNDAMENTAL HEAT TRANSFER AND FLUID DYNAMICS

Water Boiling in Multirod Geometries. — We improved the existing seven-rod boiler by reducing tolerances in the rod spacing and in the positions of six semicylinders affixed to the wall. These partial rods simulated the hydraulic boundary of a larger array, while a brass jacket containing six heater rods surrounded the flow shroud and provided thermal boundary simulation. A good thermal bond between the jacket and the shroud was obtained by filling the gap with molten tin. In preliminary experiments with a rod spacing of 1/16 in., a flow of 1 gpm of saturated water, no jacket heat, and equal heat generation in all rods, we estimated "burnout" on the central rod to have occurred at a heat flux of about 200,000 Btu hr^{-1} ft^{-2}. This result compares favorably with previous data in which the critical flux was achieved only on the peripheral rods. Repetition of the experiment following complete disassembly and reassembly of the test channel gave critical flux values in very close agreement with the initial values.

Parallel Flow in Rod Clusters. — We developed an accurate approximation method for determining the velocity, pressure drop, friction factor, and shear stress for laminar flow through a circular channel containing a rod cluster. This method employs a mapping function and the

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transformation of the complex potential from one polar coordinate system to another. The velocity may be expressed explicitly in any portion of the flow channel within the complicated physical boundaries before the boundary conditions are imposed. The boundaries are then point matched in the least-squares sense, and solutions are obtained and ensured to any desired degree of accuracy.

We applied this analysis to 13 variations of the basic configuration to test the effects of the number of rods, channel-rod radius ratio, and peripheral-rod eccentricity. The number of rods was varied from 5 to 11, the radius ratio from 4 to 5, and the eccentricity from 0.8 to 0.9. Velocity maps were developed for all conditions. We found also that the pressure drop was affected significantly by the channel-rod radius ratio but only slightly by the eccentricity.

Hydromagnetic Stabilization of Vortex Flow: Experimental. — We accumulated data using the 4-in.-diam vortex tube operated with one, two, and four injection slits feeding the aqueous ammonium chloride solution. The 75-kilogauss air-core solenoid in the Magnet Laboratory was the source of an axial magnetic field. In order to present the experimental results in a form most useful for examining the feasibility of magnetically stabilizing a high-temperature plasma-core reactor or magnetohydrodynamic (MHD) generator, we postulated that growth of the boundary layer between injection slits controls the stability of the flow; accordingly we examined the data in terms of the critical Goertler modulus (Reynolds modulus based on momentum thickness of boundary layer times square root of ratio of momentum thickness to radius at transition to instability) and the Hartmann modulus (based on momentum thickness). We found that the individual data points for one-, two-, and four-slit operation with the 4-in. vortex tube fall on three distinct curves of somewhat lower slope than that of a fourth-degree polynomial least-squares curve based on the data for all three slits collectively. However, by modifying the exponent of the ratio of momentum thickness to radius from 0.5 to 0.43, we obtained an excellent correlation of the results in which the individual data points for one-, two-, and four-slit operation fall on a fourth-degree polynomial least-squares curve based on all the data. Of course there is no justification from boundary-layer theory for an exponent other than 0.5, but the empirical correlation should nevertheless be more valid for interpolation than that based on the theoretically derived Goertler parameter.

Hydromagnetic Stabilization of Vortex Flow: Theoretical. — When we applied laminar boundary-layer theory to determine stationary velocity profiles in a jet-driven vortex tube, we found that the recovery ratio (ratio of tangential velocity extrapolated to the wall to jet velocity) should be a unique function of the dimensionless parameter $N_{Re,r}/(nN_{Re,j}/2\pi)^{1/2}$, where $N_{Re,r}$ is the radial Reynolds modulus, $N_{Re,j}$ is the tangential Reynolds modulus based on jet velocity, and n is the number of (equally spaced) injection slits. A comparison of calculations with experimental results obtained in a 4-in.-diam vortex tube using one, two, and four injection slits indicates that the recovery ratio can be predicted with an average deviation of about 5%.

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The essential difference between the jet velocity profile of direct interest and the velocity profile obtained between rotating cylinders is the variation of the jet-driven flow with peripheral distance. We calculated the stability of these jet profiles by ignoring the peripheral variation, that is, by choosing the most unstable profile and assuming it to apply around the entire periphery. The Taylor-Goertler theory is employed to make the parallel flow approximation, along with inclusion of the effect of radial flow. The results, presented in terms of the critical Goertler modulus and the Hartmann modulus, show that the Goertler modulus increases with the Hartmann modulus but falls significantly below the experimental values.

POWER REACTOR FUEL PROCESSING

Analysis for Chemical Processing of Nuclear Fuels. — Experiments were performed to determine whether isotopic sources could be used to excite characteristic x-ray spectra suitable for monitoring UF_6 and PuF_6 in process gas streams of the fluoride volatility process. In simulated tests, x radiation from aqueous uranyl solutions excited by a 1-mc ^{57}Co source was measured with a 3-in. NaI crystal scintillation detector. With uranium solutions in 1-cm-diam cylindrical containers surrounded by 15 mils of nickel foil and fluorescent radiation measured at an angle of 135° from the source (the most favorable practical geometry for resolution of the energies of the uranium K spectra from scattered radiation), only about 70 net counts/min were observed for concentrations corresponding to 10% UF_6 in the gas phase. This counting rate is only slightly above the statistical variation of the background. While a lower background would be expected in the measurement of gaseous systems, the improvement is estimated to be no greater than a factor of 4; therefore the technique would not offer adequate sensitivity, even with increased source strength. Similar experiments will be performed with a lithium-drifted germanium detector which offers partial resolution of the fluorescent x rays from the scattered background radiation.

Development of Close-Coupled Processes. — Since refabrication of thorium fuels will require shielded facilities, coupling the reprocessing plant to the refabrication plant (and possibly to the reactor) may decrease fuel-cycle costs to a minimum. We are developing methods for partial decontamination and recycling of thorium fuel by remote operation and are preparing an engineering cost evaluation of these new concepts.

Accurate density equations were derived for solutions of thorium and nitric acid in water and in tributyl phosphate—n-dodecane. When the thorium was less than 1 M and the acid less than 4 M, the equations gave results that were within 0.005 g/ml of the measured densities. This is about the limit of analytical accuracy.

The dense second organic phase that forms when the solubility of the thorium-TBP complex is exceeded contains about 80% TBP. Therefore an extractant containing 80% TBP would be free of the second-organic-phase problem. Eighty percent TBP was tested as an extractant and was found to

be free of this problem. However, the density difference between the organic phase and the equilibrium aqueous phase was not sufficient to give satisfactory phase separation.

The extraction of thorium nitrate with 30% triisopentyl phosphate in n-dodecane is similar to its extraction with 30% tri-n-pentyl phosphate in n-dodecane, which was previously reported. The thorium distribution coefficients with 30% triisopentyl phosphate are somewhat larger than with tri-n-pentyl phosphate but are less than with TBP. A second organic phase formed when the triisopentyl phosphate was 0.31 M in thorium and the equilibrium aqueous phase was more than 2 M in $Th(NO_3)_4$. The corresponding values with TBP are 0.26 M in the organic phase and 1.04 M in the aqueous phase. There is, therefore, little probability of encountering a second organic phase when triisopentyl phosphate is used as the solvent in a thorium extraction process.

The cost estimate for the close-coupled plant designed to process and remotely fabricate 165 kg of Zircaloy-clad, vibratory-compacted sol-gel $(U,Th)O_2$ fuel per day was completed. The capital costs were estimated to be about $16,150,000 for the facility, and the annual operating cost was about $4,400,000, not including the cost of the makeup uranium and thorium. The total cost was estimated to be about $157 per kg of U + Th. The cost of the batch extraction plant, which was technically impractical, was essentially the same as the cost of the continuous single-cycle solvent extraction plant. Compared with previous estimates (ORNL-TM-1308), this is a rather attractive cost for such a small plant. However, the close-coupled-plant study assumed well-developed technology and a frugal approach to design and plant staffing. The corresponding cost for a small plant study, made on a similar cost basis but with low-enrichment uranium fuel with complete decontamination processing and direct fabrication, was about $120 per kg of U + Th, indicating a cost premium of about 30% for the more complex thorium fuel cycle.

FUEL-CYCLE STUDIES

Gas Evolution from Sol-Gel Fuel Materials. — During the sphere-forming operations, organic chemicals are introduced into the microspheres prepared by the sol-gel process. These organics appear as carbon in the fired microsphere product unless special steps are taken to remove it. Oxidation at relatively high temperatures (above 650°C) is one approach, but it is advantageous in some instances to remove the carbon at lower temperatures. Studies are in progress to achieve this objective.

Our most recent work has been with UO_2 microspheres, with which simple oxidation of the carbon is not practicable. We have found that steam "scrubbing" is an effective treatment. The gel microspheres are enveloped in an excess of water vapor at room temperature, and the excess water is pumped off. The sample is then heated to the temperature being studied, and the gases released at this temperature are pumped off and collected and analyzed. The following observations can be made, based on a study of the steam scrubbing technique over temperatures ranging from 100 to 650°C.

1. Temperature intervals of maximum gas evolution appear to be 150 to 250°C and 400 to 650°C.
2. The principal gas evolved (primarily in the range 300 to 350°C) is methane. This is in direct contrast to the higher-molecular-weight organic products previously noted in the case of ThO_2 sol-gel microspheres.
3. Nitric oxide is evolved at 150°C.
4. There is a suggestion that if sintering can be avoided, steam scrubbing around 650°C might greatly minimize the amount of carbon present in the products. This conclusion is based in part on the following evidence. After a steam scrubbing at 650°C for 1 hr, the UO_2 beads showed little reactivity with an excess of CO_2 or H_2O (i.e., less than 0.02 cm^3/g). Previous studies had indicated that carbon impurities reduce CO_2 to CO. Metallographic examination of the beads showed little evidence of treatment-induced defects.

It must be emphasized, however, that steam heating at about 650°C cannot completely remove the carbon. To establish this point, the UO_2 beads after the 650°C treatment were ground down to smaller than 270 mesh. Subsequent reheating of the material resulted in additional gas evolution.

Thorium-Uranium Recycle Facility. — The Thorium-Uranium Recycle Facility is a shielded facility being built to provide for development of methods of remote processing and fabrication of Th-^{233}U fuels. The building is made up of four 20-ft-wide cells having a combined length of 106 ft.

Construction is now about 57% complete. Liners are now up in all cells to the top of the viewing-window forms. The building exterior has been completely prime coated, and about one-half has received the first finish coat. Equipment procurement is proceeding satisfactorily.

Design drawings of the uranyl nitrate storage system for the sol-gel process are complete except for very minor changes and drafting corrections. Design of a sol-preparation system to produce 1 kg of urania as sol in an 8-hr day is about 50% complete.

^{233}U Storage and Distribution. — The final shipment of $^{233}UO_2$ containing less than 5 ppm of ^{232}U was received and stored during the month. The present inventory of this material is 51.7 kg of ^{233}U, and the total inventory of ^{233}U in all forms is 189.4 kg. Four containers of scrap oxide and nitrate solution containing 599 g of ^{233}U (91%) were received and stored. Two shipments, amounting to 105 g of ^{233}U, were made during the month.

About 7 kg of ^{233}U as UO_2 was removed from the dry storage wells and dissolved in the Kilorod batch dissolver. The dissolver inventory is presently 10 kg, which will be processed during the first two weeks in May.

Calibration and boiling tests with the new safe-geometry dissolver were completed. The unit will be placed in operation upon final approval by the Criticality Review Committee.

TURF Fuel Fabrication Line. — We started the conceptual design of the equipment that will be installed in the Thorium-Uranium Recycle Facility to demonstrate the refabrication technology of fuel elements for a high-temperature gas-cooled reactor. A tentative flowsheet has been selected, and work is being concentrated on those parts of the process that are not dependent on the as-yet-undefined fuel element design. Designs for a particle classifier, a shape separator, and three-way particle diverter valves are complete. We are now designing the coater furnace, the pressing equipment for consolidating particles, and a pneumatic system to convey them.

Extrusion Fabrication of Sol-Gel-Derived Fuels. — We are investigating the extrusion fabrication of small-diameter fuel pieces to try to find economical ways to remotely fabricate sol-gel-derived materials.

Powders are being added to the 10 M ThO_2 clay prior to extrusion, in an effort to eliminate the internal cracking (which accompanies drying) and shrinking (which results from firing) of pure ThO_2 clay and to lower the cost of this fabrication technique by recycle of scrap materials as powder. We are investigating the effects of powder loading, powder particle size and size distribution, and heat treatment of the powders prior to addition.

An additional technique of interest in extrusion fabrication is being tested. A tube about 0.2 in. in inner diameter, with a 60-mil central hole, has been successfully extruded from pure ThO_2 clay. The uniformity of wall thickness and extent of internal cracking are being determined.

An initial effort to obtain an extrudable clay from 1.8 M UO_2 gel by vacuum drying has yielded a clay (about 7 M) with small pieces of drier UO_2 in it. This material appears to be of extrudable quality, and further evaluations will be made upon completion of the controlled-atmosphere glove-box system necessary for work with UO_2 sols. This system is nearly 50% complete and should be finished within the next month.

Firing Studies of UO_2 and UO_2-ThO_2 Microspheres. — A modification of the firing method previously used for firing $(U_{0.22}Th_{0.78})O_2$ gel microspheres to near-theoretical density and to very low carbon contents is being studied for possible use with pure UO_2 and for $(U_{0.8}Pu_{0.2})O_2$. In preliminary work, ThO_2 is being used as a stand-in for PuO_2.

The essential features of the procedure used previously included:

1. Firing with Ar–4% H_2 to some intermediate temperature so that the oxygen-to-uranium ratio is reduced and a possible phase transition to U_4O_9 or preliminary sintering of U_3O_8 is avoided in subsequent oxidative firing.

2. Making the atmosphere oxidizing (carbon dioxide is the gas of choice), at some intermediate temperature, and firing to the final temperature. At this stage, the carbon is removed ($CO_2 + C = 2CO$) and an assist in densification is achieved.

3. Densifying, at the final temperature, by firing in Ar–4% H_2. This also reduces the oxygen-to-uranium ratio.

Step 2 is not well understood. Uranium dioxide very slowly oxidizes during this step, and this oxidation enhances the sinterability of sol-gel-derived UO_2. Thus it is at this operation that we are studying various reaction times and temperatures.

The principal finding we have made is that by "soaking" the material in CO_2 for times up to 30 min, we can improve the product, both by increasing the density and by reducing the carbon content. Addition of 20% ThO_2 to the UO_2 makes a large change in the sinterability of the UO_2, even when the soaking period is used. Thus, while UO_2 sintered to greater than 98% of theoretical density at 850°C for 1.5 hr, UO_2–20% ThO_2 required a temperature of 1100°C to achieve the same density.

Coated-Particle Development Laboratory. – This laboratory was set up to develop methods for making carbon-coated microspheres of thorium and uranium oxides and carbides and their mixtures. Equipment and processes developed here will be used in the shielded Thorium-Uranium Recycle Facility now under construction.

About 23 kg of thoria microspheres having diameters larger than 420 μ was prepared for use in pyrolytic-carbon coating studies. The porous metal and the porous glass supports used to collect and retain the gel spheres clogged and required cleaning after prolonged use. Cleaning procedures amenable to remote operation must be developed to cope with this.

During this month, coating studies demonstrated that we could achieve low-density (1.5 g/cm^3) coatings on a reproducible basis for 1-kg batches. However, when we attempted to duplicate this density in a 30-μ-thick coating on 2-kg batches, we obtained densities of 1.67 and 1.69 g/cm^3 in two runs.

A slotted injector nozzle was tested, and 2 kg of particles coated by it at 1700°C in a 45-min period showed very good sphericity. The coating was 30 μ thick. Also, a water-cooled copper nozzle with gas at 2150°C was operated satisfactorily for 45 min, with no blockage due to carbon deposit.

Twelve experiments with the entrained-bed coater were made. The bed charges were each 2500 g of 250- to 300-μ-diam ThO_2 spheres, and the bed wall temperature was maintained at 1800°C. Coating efficiency decreased only slightly with large increases in methane flow rate; however, coating density is quite flow-rate dependent in the lower flow rate range.

Thoria-Urania Microsphere Preparation. – One of the objectives of our studies is to learn how sol-gel technology can be extended to include thoria and urania mixtures in all ratios rather than be limited to mixtures containing less than 10% urania, as in the original sol-gel process. We have now prepared more than a kilogram each of UO_2-ThO_2 microspheres (210 to 297 μ in diameter) having Th/U ratios of 3.25 and 4.6. Gel microspheres were prepared by mixing a UO_2 sol with a ThO_2 sol and forming the resultant sol mixture into microspheres in a forming column containing 2-ethyl-1-hexanol and the surfactants Ethomeen S/15 and Span 80. The gel microspheres were dried in argon at 120 to 140°C. The dried gel spheres fired to excellent ceramic spheres that had very nearly theoretical density and good resistance to crushing (about 1000 g), and which x-ray analysis showed to be solid solutions of UO_2 and ThO_2. A nonuniformity in color observed after firing to 1100°C in Ar–4% H_2 was corrected

by firing at 1400°C in hydrogen. The final product spheres were transparent and light amber.

MOLTEN-SALT REACTOR PROGRAM

Molten-Salt Reactor Experiment. — The revisions to the fuel off-gas system were completed when the particle trap and charcoal bed assembly was installed between the new, larger, pressure control valve and the reactor. While the fuel system was being filled, a variable restriction was observed at the inlet of the auxiliary charcoal bed. When we cut the line, we found a check valve poppet, apparently lost from one of the three check valves in the drain tank vent lines.

Fuel was circulated three days with the reactor subcritical with no indication of changes in the off-gas system. Operation at 1 Mw for three days followed. During this period the new particle trap and charcoal bed heated up, showing that some material was being absorbed, but there was no increase in pressure drop. There was, however, an increase in pressure drop through the main charcoal beds, from 1 psi to 1.5 psi. When the reactor was taken to 2.5 Mw, the pressure drop increased to over 3 psi within a day after the power increase. The pressure drop was restored to normal by pressurizing the beds, then allowing the helium to exhaust backwards into the inlet holdup volume. Operation at 2.5 Mw was then continued for two more days with no further difficulty. Similar behavior was observed when the power was raised to 5 Mw — the beds partially plugged shortly after the power increase, but were easily blown clear. We believe that the restrictions are caused by material coming from the holdup volume between the pressure control valve and the charcoal bed.

Dynamics tests at 2.5 and 5 Mw confirmed earlier predictions of stability.

Xenon poisoning at each power appeared to be lower by a factor of 4 than predicted, implying more effective removal.

Reactor Analysis. — A report was prepared which presents the experimental and theoretical work on period measurements on the MSRE with the fuel circulating. Work was begun on a summary report of all MSRE zero-power physics experiments.

An attempt is being made to extend the same spatial model used to analyze the period measurements during circulation to the calculation of the zero-power transfer function. The purpose is to provide a firmer foundation for the changes in core parameters (such as effective core volume and lag times) required to fit calculations from a simpler, space-independent theoretical model to the measured values of the frequency response. This calculation has been programmed and is being debugged.

During MSRE run 3, several sets of rod-drop experiments were performed as an independent check on the rod worths determined by period-sensitivity measurements. A convenient method of analyzing these experiments has been developed, which makes use of the MATEXP program to solve the neutron kinetics equations for the time integral of the flux following the rod drop. Several of the rod-drop experiments made with

the fuel stationary in the core have been analyzed by this technique. Results are found to be consistent with the rod worths determined by the separate techniques of period measurements and comparison with equivalent ^{235}U addition. Self-consistency in results from these three techniques is approximately 5% of the reactivity worths measured by the period-sensitivity method.

Development of Components and Systems. — Plugging of the off-gas lines while the reactor was being operated at 1 Mw was traced to oil-like and varnish-like hydrocarbons which collected at valves and capillary restrictors, where either radiation damage or thermal effects produced polymerization. Our solution to the problem was to install a particle and heavy-vapor trap in the line between the control valve and the reactor and to increase the opening size of the valves. In addition, a noncritical check valve and a restrictor were removed from the system, and provisions were made for the future installation of additional traps at critical positions in the off-gas lines if they become necessary. Neither the exact source nor the rate of inflow of the oil is known at this time; the trap is designed to supply some data from which this information can be deduced. Although the off-gas system is not free from the buildup of pressure drop, it is operating much better now.

Operation of the freeze valves and control rod drives has been satisfactory during the startup. A high-temperature alarm thermostat was shown to be faulty by inferring the temperature from the measured dc resistance of the windings in the motor which the thermostat was supposed to protect. Since the motor temperature was still within limits, the motor windings will be used as the element of a resistance thermometer, and the thermostat may be eliminated.

The sampler for the fuel salt system has operated to add 87 capsules of enriching salt and to remove a total of 103 samples. The last nine samples have been removed after varied periods of operation of the reactor at 1 to 2.5 Mw. The last sample removed near the end of the 2.5-Mw operation read 50 r/hr at 1 ft of air. Only two small radiation beams were found through the sampler shielding, and these were plugged.

The remote maintenance system was given its first real test under radioactive conditions and performed well. Several nonroutine tasks were performed easily, demonstrating the basic adaptability of the maintenance approach. Many of the routine tasks have been turned over to the craft forces.

MSRE Pump Program. — The prototype pump test facility was operated for 1200 hr, circulating salt at 1200°F, 1300 gpm, and 1165 rpm. The off-gas lines were modified to obtain gas samples and to test the effectiveness of various filter designs in the removal of particulate matter which may be in the gas stream. Some gas samples were taken and are being analyzed. The radiation densitometer was reinstalled in the facility to make additional measurements of the undissolved gas content in the circulating salt; this was necessitated by a change in impeller diameter, which in previous tests was 13 in. and presently is 11-1/2 in.

The spare rotary element for the MSRE fuel pump was assembled and was operated in the cold shakedown stand. It is being held in storage for the MSRE.

The PKP molten-salt pump continued to operate, circulating salt at 1200°F, 800 gpm, and 1650 rpm. It has operated continuously for 6350 hr. The endurance test of the MSRE lubrication pump was continued; it has operated for 23,600 hr, circulating oil at 150°F and 60 gpm.

The design of the Mark 2 pump tank for the MSRE fuel pump was reviewed. Additional analytical and design work will be required to provide a satisfactory design.

NUCLEAR SAFETY

Properties of Fission Product Aerosols Produced by Overheated Reactor Fuels. — A Zircaloy-clad highly irradiated UO_2 fuel capsule was melted in a pressurized steam-air atmosphere in the Containment Mockup Facility by use of rf induction heating. The heating period lasted about 10 min, but during a 20-min cooling period the pyrophoric nature of the melted Zr-UO_2 fuel residue was demonstrated by alternately slow and rapid oxidation in air.

We intended to provide sufficient steam for a complete metal-water reaction, but penetration of molten zirconium metal into the UO_2 prevented the oxidation in steam from going to completion. The unoxidized metallic zirconium apparently accelerated the oxidation of UO_2 to U_3O_8 in air as cracking of the cooling residue caused fresh surfaces to be exposed to the air stream.

In an effort to obtain a quick indication of changes in fission product content of the fuel that occurred during the experiment, aliquots of solutions of unmelted and melted fuel specimens were exposed to a lithium-drifted germanium detector, and the resulting gamma spectra were recorded with a 400-channel analyzer. Heath (Phillips Petroleum Company — NRTS) is developing gamma-stripping analysis techniques for spectra obtained with this high-resolution detector which promise to be more versatile than those making use of sodium iodide detectors. We identified 30 major gamma rays in the spectra obtained. The absence of gamma rays from iodine in the melted-fuel spectrum and a reduction in the cesium and tellurium peak heights were the major differences shown by the comparison of spectral curves. Analytical data needed to fully evaluate the experiment are not available at this time.

Behavior of Fission Products During In-Pile Destruction of Reactor Fuels in the ORR. — The fission product behavior in the recently performed experiment using Zircaloy-clad fuel melted in moist air apparently was quite similar to that observed in experiments using stainless-steel-clad fuel under similar conditions. However, the particles upon which the volatile iodine, tellurium, and cesium were transported from the furnace appear to be considerably larger than usual, since these materials were transported to the filter instead of being deposited on the diffusion tube. The particle size distribution on the filters is being determined. Approximately 30% of the iodine produced penetrated the F-700 high-efficiency filter as usual in an experiment using an oxidizing atmosphere, but less than 0.03% of the tellurium and cesium penetrated the same filter. Less than 0.06% of the iodine penetrated the first half inch of charcoal.

Physical analysis has been completed of an experiment performed to study the effects of overheating stainless-steel-clad UO_2 for 5 min at 1500°C in a slow flow of steam-hydrogen-helium. Visual examination had shown no evidence of fuel melting or central void formation characteristic of the residues of unmolten fuel specimens, although metallographic examination showed that the center of the fuel had partially melted and that central void formation had begun. This indicates that in experiments performed similarly to this one but with subsequent fuel melting, the central void is formed during the higher temperature phase of the experiment. In addition, visual and metallographic examination showed no eutectic formation between the stainless steel cladding and the UO_2, although oxides of stainless steel formed on both the inside and outside of the cladding. Other experiments (ORNL-3691, p. 34) have shown the formation of a low-melting eutectic between oxides of the stainless steel cladding and the UO_2 fuel.

In a previous experiment (ORNL-3843, p. 7) using a steam-air atmosphere, there was a possibility that an atmosphere capable of oxidizing fission products was not obtained because stainless steel cladding reacted with all of the oxygen. Therefore, an experiment has recently been performed in which stainless-steel-clad UO_2 was melted in a slow flow of steam-oxygen. Visual examination showed that the fuel completely melted and as expected did not appear any different from that of the steam-air experiment. The effects of the increased oxygen supply on the behavior of fission products will be determined as the radiochemical analyses are completed.

TREAT Fission Product Release Studies. – Radiochemical analysis of experiments 9 and 10 is now complete. In these experiments, stainless-steel- and Zircaloy-clad UO_2 fuel specimens were melted under water by transient fission-heat input of approximately 510 cal per gram of UO_2. Most of the water boiled out of the fuel-containing autoclave in 1 min to simulate a ruptured reactor pressure vessel system, followed by slow boiling to dryness in 10 min. Approximately 60% of the ^{131}I and ^{137}Cs was released from the melted fuel-cladding mixture. Twelve to 18% of the total ^{131}I and ^{137}Cs was carried out of the fuel autoclave with the escaping steam, and 35 to 45% of the iodine and cesium was collected later in distilled-water rinses of the walls of the disassembled fuel autoclave. We hypothesize the following simplified mechanism for transport of iodine and cesium in these tests. First, the portion (12 to 18%) which escaped from the fuel autoclave was carried only by steam which was formed at the heated fuel surface and moved out of the autoclave without being condensed. Second, the iodine and cesium which entered the liquid water either directly or by entrainment in condensing steam was essentially permanently trapped by chemical reactions to form mostly nonvolatile water-soluble compounds such as cesium hydroxide and metal iodides. Less than 0.01% of the total ^{131}I was found in penetrating forms such as CH_3I.

Characterization, Control, and Simulation of Fission Products Released Under LOFT Conditions: Development of Characterization Devices for Measuring Forms of Radioiodine. – Initial tests on May pack responses to tagged methyl iodide at an elevated temperature and in the presence and absence of water vapor have been completed. The May packs were loaded

with materials received from the LOFT program and involved the following loading sequence: three layers of high-efficiency filter material (HV-70), eight silver screens (80 mesh), five layers of charcoal-impregnated filter paper (ACG/B), two 0.75-in. charcoal beds (PCB, 12/30 mesh), and, finally, three layers of high-efficiency filter material (HV-70). In each experiment, two May packs were exposed in parallel, together with one composite diffusion tube at the same temperature and relative humidity and a second composite diffusion tube under dry conditions at ~25°C. This arrangement allows cross-calibration of the characterization devices, in addition to verification of the form of iodine in the source.

Under dry conditions at 90°C with an exposure of 5 min to a gas velocity of approximately 10 fpm, the methyl iodide was distributed about equally between the charcoal-impregnated filters and the first charcoal bed. About 0.05% of the sample entering the May pack deposited on the first high-efficiency filter pack and about 0.05% on the silver-screen section. The associated diffusion tube assembly showed depositions of 0.5, 16.6, and 81.7% on the silver, rubber, and charcoal sections respectively.

At 75% relative humidity at 90°C the methyl iodide was carried deeper into the packs, with about 96% depositing on the first charcoal bed. Deposition on the second charcoal beds was 1.8 to 3.4%. The silver screens took up only about 0.13%, while the first high-efficiency filters, the charcoal-impregnated, and the last high-efficiency filters retained 0.16, 0.65, and 0.04% respectively. The associated diffusion tube assembly showed a deposition distribution of 0.3, 12.5, and 86.6% on the silver, rubber, and charcoal sections.

The runs revealed that the methyl iodide is not carried completely into the charcoal beds of the May packs except under moist conditions. No significant amount of the organic iodide deposited on the silver screens under wet or dry conditions. As observed previously in the diffusion-tube studies, a large portion (about 15%) of the methyl iodide deposits uniformly on the rubber section of the composite diffusion-tube assembly before it reaches the charcoal tube.

Future tests will involve the May pack response to elemental iodine vapor under exposure conditions similar to those above. After this series of tests, modifications to the May packs will be accomplished, if needed, and additional response tests will be performed.

Characterization, Control, and Simulation of Accident-Released Fission Products: Removal of Methyl Iodide from Gases Under Reactor Accident Conditions. — Investigation of the removal of radioactive methyl iodide from flowing air-steam mixtures by impregnated activated charcoals has been continued. The tests at 212°F and atmospheric pressure have been extended to cover a wide variety of laboratory-impregnated and commercially impregnated charcoals. Also, an experimental system has been constructed and utilized for testing the more promising charcoals under more severe conditions, including steam-air pressures of 60 psia and temperatures in the vicinity of 300°F. In both types of experiments it has been noted that relative humidity (or the degree of steam saturation) ranks very high among the variables controlling methyl iodide removal efficiency.

To illustrate the type of results obtained at the higher pressure and temperature (60 psia and around 300°F), a few examples will be given. The data that follow are for a commercially available charcoal (MSA 85851) impregnated with a form of iodine, and the mechanism for trapping radioactive methyl iodide is isotopic exchange. For a 2-in. depth of this charcoal, a methyl iodide loading of 2.5 mg per gram of charcoal, and a superficial linear velocity of air-steam through the charcoal of around 40 fpm for 5 hr, radioactive methyl iodide removal efficiencies as a function of percent relative humidity were observed as follows: 98.5% removal at 70% RH; 93% removal at 87% RH, and only ~40% removal at near 100% RH. Charcoal adsorbers must therefore be operated at relative humidities somewhat below 100% if useful methyl iodide removal efficiencies are to be obtained. Preliminary results on samples of iodine-impregnated charcoals from other manufacturers indicate that their behavior under high relative humidities will be very similar to that noted above for MSA 85851 charcoal.

Particulate Materials from Reactor Accidents: Characterization, Simulation, and Filtration. — Studies involving the calibration and application of the fibrous filter analyzer (FFA) for aerosol behavior studies and particle sizing under moist conditions at ambient and elevated temperatures are continuing.

While we are awaiting the completion of the large recirculating aerosol system for the study of the behavior of stainless steel—uranium oxide aerosols under various combinations of reactor accident conditions, preliminary studies are being carried out with the small test facility. The FFA is being utilized in this study of the effects of temperature and humidity on a newly formed aerosol.

Some 40 experimental runs have been made in the effort to calibrate the response of the FFA to a zinc oxide aerosol. Variables have included gas velocity, humidity, and age and concentration of the aerosol. As a part of the effort to interpret the results obtained, single-fiber efficiencies are being calculated from the combined particle collection mechanisms of diffusion, interception, and inertial impaction. A computer routine has been developed to aid in this data reduction process.

Ignition of Charcoal Adsorbers Under Reactor Accident Conditions. — An experimental assembly for studying the ignition of charcoal under in-pile conditions is 75% completed. Ignition temperatures have been measured in the laboratory with this apparatus and are being analyzed statistically to establish the characteristics of the system. A series of ten identical experiments, of ten cycles each, have been run in the ignition tube which will be installed in the in-pile unit. A cycle consists of igniting the charcoal by slowly increasing the temperature, while air flows through the charcoal, and then extinguishing the burning by stopping air flow and turning off the heater. After cooling, the process is repeated for the next cycle. An in-pile experiment will be run in the same way and in the same apparatus, and its results will be compared with the results of the laboratory experiments. Such a procedure will ensure that the in-pile experiment will be sensitive to any effect which fission products will have on the ignition of charcoal.

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Laboratory work is continuing to establish the effect on ignition temperature of other parameters, such as ionized air or iodine adsorbed on charcoal before ignition. Under the four conditions most studied, dry and moist air at 10 and 20 fpm, the initial ignitions show little response to the changes, but subsequent ignitions of the same charcoal occur at 10 to 20° lower when the air flow is changed from 10 fpm to 20 fpm. No definite effects can be detected when changing the moisture content from dry, <1% relative humidity, to <95% relative humidity.

Steam-Carbon Reaction and Fission Product Release and Transport Studies. – The major effort during this period has been directed toward procurement and assembly of the equipment required for the projected high-temperature experiments. Two high-temperature furnaces have been received, and a third is being fabricated locally. Power supplies for these furnaces have been designed and requested. Various alumina items, including specialized tubing, have been received, and orders were placed for additional alumina shapes. The latest communication on the gas chromatograph indicates delivery around the middle of May.

Four runs were made at 1000°C in which sections of ATJ graphite pipe (15/16 in. OD, 11/16 in. ID, 2 in. long) were exposed to helium-steam mixtures at a total pressure of 1 atm. A partial pressure of steam of ~270 torrs and total flow rates of 555 or 1250 cm^3/min (STP) were used. Reaction rates of 45 mg g^{-1} hr^{-1} or 11 mg cm^{-2} hr^{-1} were obtained for both flow rates, indicating that the reaction is not sensitive to flow rate changes in this range. For rates expressed on a weight basis (as mg g^{-1} hr^{-1}), the reported rate, 45, is higher than the 25 found for ATJ graphite spheres at a partial pressure of steam of ~730 torrs and compares more favorably with the 42 reported by Walker et al. for a partial pressure of 142 torrs. If rates are expressed on a geometric area basis (as mg cm^{-2} hr^{-1}), the currently found rate, 11, is to be compared with 29 for ATJ spheres and 21 for graphite used by Walker et al.

Additional studies of the reaction of unirradiated pyrolytic-carbon-coated fuel particles with low partial pressures of water vapor have been made in the temperature range 1000 to 1200°C, using a recording balance to measure reaction rates. Helium–water vapor mixtures containing 250 or 500 ppm, by volume, of water vapor were used at flow rates of ~200 cm^3/min (STP) at a total pressure of 1 atm. Inhibition of the water vapor–carbon reaction has been observed in many of these tests, but the cause of the marked decrease in reaction rate after various periods of heating is still unknown.

LOFT Support in Out-of-Pile Nuclear Safety Studies. – Construction of the Containment Research Installation (CRI) is progressing at a rate which should ensure hydrostatic testing and instrumentation checkout of the complete system by May 20, 1966.

The vessel liner and containment vessel have been assembled, and a leakage test was performed with the assembled units. Minor problems concerning insert and vessel misalignment are being overcome by machining the affected components. The first liner to be inserted in the CRI is being painted with Phenoline 302, which is the coating to be used in the Containment Systems Experiment (CSE) at Hanford and is, consequently, the coating to be tested as part of ORNL's commitment to Battelle Northwest

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Laboratory. Four more liners, which will be coated with various other materials, are being fabricated. Final polishing of the containment vessel and welding on of studs for the placement of heater cables and insulation media are being performed.

Scaled-down models of the LOFT recycle filter and Demister unit have been fabricated. The components consist of a 2-in. Teflon Demister, a 4-in. roughing filter, a 9-in. absolute filter, and a 5-in.-diam, 3-in.-deep charcoal adsorber. These units will be tested in the CRI to determine the effects of a saturated atmosphere on the overall efficiency and on the efficiency as a function of the number of times the containment volume is cycled through the filter train.

The sampler assemblies and fission product sampling devices which will be used in the LOFT facility have been received from Phillips Petroleum Company and are being checked out prior to being tested in the CRI. Part of the procedure requires testing the units in a high-humidity, high-temperature atmosphere. A steam chamber approximately 7 ft long and 8 in. in diameter is being fabricated in which to perform these tests.

Nuclear Safety Pilot Plant. — The first high-level irradiated fuel run in the Nuclear Safety Pilot Plant was carried out on April 1, 1966. The fuel consisted of eight 5/16-in.-diam by 4-1/2-in.-long Zircaloy-clad UO_2 pins which had been irradiated to 20,000 Mwd/ton in the NRX reactor in 1960–61 and which were reirradiated in the ORR to produce 25 mc of ^{131}I as of March 28, 1966. The furnace atmosphere was helium-argon-hydrogen-steam, and the model containment vessel atmosphere was air-steam. One heating pass was made at a boat travel speed of 1 in./min, followed by a second pass at 1/2 in./min, after which heating was terminated. The plasma jet appears to have been deflected to one side of the fuel array, and examination of the residue indicates approximately 20% complete melting, with more melting at the ends than near the center of the fuel array. The cladding was oxidized and breached on all eight pins. Only scattered analytical data are available at this time, and we are not in a position to state more than that significant release of fission products from the fuel and transport to the model containment vessel occurred.

Analytical data from runs 8, 9, and 12 were processed through the data handling program, and the data from runs 8 to 12 inclusive are now stored on magnetic tape. A program to retrieve May pack sample data in tabular form has been developed.

HTGR Safety Program Office. — The HTGR Safety Program Office was established April 1, 1966, in response to a request from the AEC's Division of Reactor Development and Technology (DRDT). The purpose of the Office is to act as an extension of the DRDT nuclear safety organization in evaluating and coordinating HTGR safety research being conducted, and to provide the DRDT with competent and independent recommendations on the direction and implementation of such research for the future. While the Office forms a part of the ORNL effort in the nuclear safety field, it will necessarily report independently to the Laboratory Director's staff, since its mission involves evaluation of the ORNL research program on HTGR safety.

The Office staff is now being assembled and is to consist of a director and 1 or 2 full-time professional people, with an additional

part-time complement of 4 to 6 specialists who will evaluate areas relating to specific disciplines. Its initial efforts will involve the preparation of a long-range plan for HTGR safety research and the evaluation of test reactor facilities for obtaining physics parameters.

Engineering-Scale Steam-Graphite Reaction Rate Experiment. — Preparation of an analytical model is continuing; the model will be used to interpret steam-graphite reaction rate data obtained with small laboratory-scale graphite samples in a low-velocity flow system in terms of reactor system conditions. The model will consider such inhibiting effects on reaction rate as turbulent mass transport in the bulk stream, reactant blanketing, and in-pore diffusion.

A proposed design of the graphite test piece for the higher temperatures is essentially complete. The higher graphite temperature will be produced by the use of four heating elements sealed inside the graphite test piece, spaced so that the variation in temperature in both the circumferential and the longitudinal direction of the reaction surface will be minimized. Other remaining items of design involve insulation for the test section and the desuperheating of the steam as it leaves the test piece.

Desorption of Fission Products in a Steam Environment. — The Fission Product Deposition Test Facility is being upgraded in order to meet the additional requirements of an experiment to compare desorption of fission products in helium and steam atmospheres. The first test is in process. This test uses helium only, so that it can be used as a standard against which the results of the steam runs can be compared.

The facility is a closed loop consisting of a helium circulator, a bellows-sealed throttling valve, a heater section (17 kw capacity), an orifice flowmeter, a water injector, a test section, and the associated instrumentation. The water injector is a low-flow diaphragm pump with a minimum flow rate of approximately 10 cm^3 of water per hour. The test section is a straight piece of 1-in. sched 40 IPS stainless steel pipe 12 ft long. The normal test procedure is to place a removable section of test surface inside the test piece, so that it can be removed and sectioned for further radiochemical analysis following testing. The special instrumentation includes a differential pressure transmitter, a gas chromatograph, and a 256-channel pulse-height analyzer.

HTGR Nuclear Safety Tests in ORR Poolside Capsule Facility. — The conceptual design of the ORR poolside nuclear safety capsule, which is intended to further the investigation of the release of nongaseous fission products under simulated HTGR accident conditions, is nearing completion. The capsule will consist of an instrumented fuel element simulating HTGR core conditions, using the Public Service of Colorado reactor as reference. The element is an annular bed of loose coated fuel particles in a graphite body, with thermocouples in a central graphite spine and in the surrounding graphite body. A purged gas gap surrounds the fuel body. The outer surface of the gap is graphite.

The capsule will have two modes of operation. Initially it will operate at normal HTGR conditions (1400°C maximum fuel temperature and

1070°C graphite fuel body surface temperature) until 5% heavy metal burn-up is attained. The second mode or accident condition will then be initiated, to simulate a loss-of-coolant accident, by raising the maximum fuel temperature to above 1800°C. The annulus surrounding the fuel body will be continuously purged to one of two sets of trap systems for fission product characterization. Each trap system consists of three traps in series operated at 1200, 600, and 100°F respectively and containing different filter media.

Pressure Vessel Technology Report. — In order to better assess the risk of catastrophic failure of reactor pressure vessels, the AEC has authorized ORNL to prepare a summary report on the current state of pressure vessel technology. The information developed on the reactor vessels is to include design, materials, fabrication, and inspection. Information is also to be supplied which could be used to evaluate the worth of upgrading codification and standards, along with the identification of any research which would materially augment the safety of reactor vessels.

A preliminary outline for the report has been prepared, and assignments have been made to members of the Laboratory staff for the preparation of the various sections. Visits are planned in order to consult with experts in the field, and industrial organizations are being contacted in order to inspect fabrication facilities. It is intended that consultants will be employed to review sections of the report and prepare certain portions of the document if the need arises. Currently, the first draft of the report is scheduled for completion by September 30, 1966. A period of six months following this date is scheduled for review and final report preparation.

RADIOACTIVE WASTE DISPOSAL

Intermediate-Level Radioactive Wastes. — Incorporation of waste in asphalt and storage or burial of the product is being studied throughout the world as a general method for treating all types of low- and intermediate-level wastes. The ORNL process consists of adding wastes directly to emulsified asphalt in a stirred evaporator, evaporating the water, heating the product to about 160°C, and draining the product into a steel drum.

Asphalt products containing 10 to 80 wt % salts from simulated aluminum-fuel dejacketing waste (FDW) and Purex second-plutonium-cycle waste (2CW) were made. Leaching tests were started on the products containing 20 to 60 wt % salts from waste. The leach rates, expressed as fraction of sodium in the sample leached per day through unit specific surface area, fluctuated between 10^{-4} and 10^{-3} g cm^{-2} day^{-1} during the first four to five weeks. But after six to seven weeks, they appear to have settled down to between 2×10^{-4} and 4×10^{-4}. These results are practically the same as obtained earlier with ORNL intermediate-level waste (evaporator concentrates).

The radiation stability of products containing 60 wt % salts from FDW and 2CW samples was determined by irradiation with a ^{60}Co source. A dose of 10^6 rads showed negligible effect; a dose of 10^7 caused slight swelling; a dose of 10^8 caused the FDW sample to swell about 36% in volume and the 2CW sample about 33%. Again, these results are about the same as those obtained earlier with ORNL evaporator concentrates — about 36% volume increase at 10^8 rads.

Low-Level Radioactive Wastes: Low-Level Waste Treatment. — The water-recycle process for treating low-level radioactive waste (LLW) is being developed as a replacement for the traditional treat-and-discharge approach. The process is based on demineralization of all new water entering a nuclear installation, and recycle and reuse of used water which has been completely decontaminated and demineralized. Our micro pilot plant system includes means for (1) continuous clarification, using zeta-potential-controlled additions of the coagulant (alum) and the coagulant aids (activated silica and a nonionic organic polyelectrolyte), (2) demineralization by cation-anion exchange, and (3) sorption of remaining radioactive and nonradioactive contaminants on granular activated carbon. The product water is returned for plant use, thus providing a closed circuit.

The first test of low-salt-content water gave results that agreed well with those projected from tests with high-salt-content water (LLW). Decontamination factors of 1000 to 10,000 were obtained for all major radioactive species for up to 2400 volumes of waste treated per bed volume of cation resin: in the thousands for ^{131}I, ^{106}Ru, ^{90}Sr, and total rare earths; in the ten-thousand range for ^{60}Co and ^{137}Cs. Other radiation emitters — ^{144}Ce, ^{125}Sb, and ^{95}Zr-Nb — were reduced to the analytical limits of detection. The high decontamination factors for cobalt and ruthenium were comparable with those obtained in LLW tests. Similarly, specific conductance was a simple indicator of resin-bed exhaustion, because ionic breakthrough precedes the breakthrough of the radioelements.

Clarification of the recycle water required the addition of a nonionic organic polyelectrolyte (75 ppb) in addition to activated silica (0.5 ppm as SiO_2) and one-tenth of the alum used in the treatment of LLW [2 to 3 ppm $Al_2(SO_4)_3$]. The water, after coal-sand filtration, was comparable in clarity with distilled water, indicating more than a hundredfold decrease in turbidity.

Disposal in Natural Salt Formations. — Operation of Project Salt Vault is proceeding routinely. A test installation, consisting of 37 roof bolts, spaced 4 ft on centers, has been made in one room of the experimental area. The tentative conclusion drawn from the test is that roof bolts will effectively bolt the 2-ft-thick salt layer in the roof to the 3-ft layer above it, thus producing a 5-ft roof system which has not yet shown any signs of separating from the material above it.

Safety Evaluation. — Recent information on metabolic processes of children and adults enabled a preliminary assessment of their importance

in estimating internal dose. In particular, Kulp and Rivera have examined the effects of bone growth, rate of bone turnover, and the observed ratio on the retention of ^{90}Sr in the skeleton of man. The "observed ratio" is the fraction

$$\frac{(Sr/Ca) \text{ in bone}}{(Sr/Ca) \text{ in diet}} .$$

With the age-dependent metabolic model developed by Kulp, estimates were made of the dose received by the skeleton from ingestion of Clinch River water. Values of calcium intake and calcium content of the body were taken from Albritton and Mitchell, respectively: values of bone turnover rate and the observed ratio were from Rivera. For the age groups considered, the metabolic model gave an average dose 15% larger than the adjusted ICRP model. Unquestionably, changes can be expected in the values of metabolic factors as new information becomes available.

Engineering and Economic Evaluations. — Costs were estimated for management of the major wastes from the fluidized-bed volatility processing of Zircaloy-clad and stainless-steel-clad UO_2 reactor fuels. Management alternatives are encapsulation of the wastes without additional treatment, encapsulation following preparation of glassy solids, and encapsulation of the fission products as a glass following their separation from the inert materials by a phosphoric acid leach. In all cases, the final products are shipped to a salt mine for final disposal.

It was concluded that there is not a clear-cut economic incentive to make glasses or to leach the fission products, but that if these conversions are desirable for greater safety in handling and shipment, credits of 6 to 36% from cheaper management of the products can be applied against the cost of the waste-treatment step. Also, more economic waste management cannot be cited as an advantage of fluidized-bed volatility processing over aqueous processing for low-enrichment fuel.

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PHYSICAL RESEARCH

PHYSICS AND MATHEMATICS

Medium-Energy Nuclear Physics: Experimental. — The analysis of the differential cross sections and of the asymmetry in the inelastic scattering of polarized protons from the collective states of ^{28}Si, ^{58}Ni, ^{60}Ni, and ^{54}Fe was completed. It was found that the asymmetry is very sensitive to the coupling and clearly indicates the need for complex coupling as well as a distortion of the spin orbit potential. Good fits to the ^{28}Si and ^{60}Ni data were obtained in this way. A deuterated polyethylene target was bombarded with 52-Mev ^{3}He ions. The broad-range spectrograph at 0° showed a distribution of tritons as expected, but the cross section was found to be too low to make any inferences about the p-p residual interaction. A second run for this experiment is planned at the end of April. The excitation function for the elastic scattering of protons from ^{12}C was extended from 30 to 40 Mev and from 45 to 50 Mev. We now have cross sections at every Mev from 30 to 50 Mev for the elastic and several inelastic groups. The analysis of these data will probably take several months.

Differential cross sections for tritons from the reaction of 25-Mev ^{3}He ions on ^{48}Ti and ^{50}Ti were measured at eight angles, from 24 to 49°. Yields to the more strongly populated states in the vanadium isotopes range from 10 to 100 μb/steradian. Triton groups leading to $^{48,50}V$ states that correspond to the analogs of the respective target ground states were identified. If the (^{3}He,t) reaction populates these states by means of the (t·T) isospin term in the optical potential, the yields should be proportional to the target neutron excesses. Within our uncertainties, the $^{50}Ti/^{48}Ti$ ground state analog yield is in the anticipated ratio of 6/4.

The excitation function for the reaction $^{12}C(p,x)^{7}Be$ was measured. The data are important for estimating residual radiation to be encountered in high-intensity accelerators. Previous measurements were done at bombarding energies greater than 75 Mev. The present measurements, covering the energy range below 60 Mev, were made by bombarding stacks of carbon wafers with a monitored proton beam at the ORIC and subsequently counting the 480-kev gamma rays emitted in the decay of ^{7}Be. A high-resolution Ge(Li) gamma spectrometer was used. The lowest measured cross section is 0.05 mb at 25 Mev, which is the $^{12}C(p,^{6}Li)^{7}Be$ threshold. The excitation function rises to 20 mb at 38 Mev and slowly decreases with energy to a value of 16 mb at 60 Mev. A stack of carbon wafers was sent to Harvard University for bombardment with a 160-Mev proton beam at the Harvard Synchrocyclotron. This will close the gap between the ORIC data and previous measurements.

The conclusion drawn from the $^{16}O(d,^{3}He)^{15}N$ reaction that the ground state of ^{16}O contains two-particle configurations is dependent upon the fitting of the angular distribution of the unresolved second and third excited states of ^{15}N with a combination of $\ell = 0$ and $\ell = 2$ distorted-wave calculations. To determine if the predicted individual angular distribution was correct, the $^{19}F(d,^{3}He)^{18}O$ reaction was studied. Preliminary

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results indicate that the calculated and observed angular distributions for the ground ($\Delta\ell = 0$) and first excited ($\Delta\ell = 2$) states are in agreement.

The installation of the broad-range spectrograph system was essentially completed. The focal plane was located, and the system was initially calibrated with elastically scattered protons at 20, 31, and 42 Mev. The spectrograph was found to operate with characteristics very close to those calculated from ray tracing studies. The technique of matching the energy gradients of the beam preparation and the spectrograph magnets was shown to be very important for this system. With a prepared beam having an energy spread of 1 part in 1000, total line widths observed with the spectrograph approached an equivalent resolution of 1 part in 2000. Typical resolutions of 1 part in 1500 were obtained routinely.

Because of the good results obtained in the preliminary operation of the broad-range spectrograph, several experiments were undertaken. A study of (p,d) reactions with 31-Mev protons yielded results for the reactions $^{89}Y(p,d)^{88}Y$, $^{90}Zr(p,d)^{89}Zr$, and $^{91}Zr(p,d)^{90}Zr$. For all of these reactions the deuterons of interest have an energy in the region of 22 Mev. The observed resolutions were about 18 kev. In the first reaction a previously unresolved doublet in the low-lying levels of ^{88}Y was found. The second two reactions were part of a study of the neutron-neutron residual interaction between the shell model $1g_{9/2}$ and $2d_{5/2}$ levels. A closely spaced sextuplet of states at an excitation of about 5 Mev in ^{90}Zr was observed at one angle. Positive identification of these states must await the processing of data taken at other angles.

Medium-Energy Nuclear Physics: Theoretical. — The study of 1p-shell nuclei using $(1s)^4(1p)^{A-4}$ configurations and an effective interaction between the 1p nucleons was continued. Previous calculations used a first-order effective interaction, due to Kuo. We repeated these calculations using an interaction due to MacKellar and Becker; the resulting p-shell spectra are much the same. The p-shell spectra were also calculated with a second-order effective interaction, due to Kuo. The resulting spectra are again much the same, but with one additional encouraging feature: the $p_{3/2}$-$p_{1/2}$ splitting required to fit the p-shell data is much more nearly constant over the shell.

Electronuclear Systems Development. — A new system for measuring the energy of the external beam was constructed, and preliminary tests were made in the ORIC. The time for the beam to travel between two probes capacitively coupled to the beam, but separated by a known distance, is measured by using variable-length delay lines. Therefore, the system measures the velocity of the particles directly. Bench tests indicate that with a sharp beam pulse the accuracy should be a few parts in 1000. The first test results on the external beam were less accurate because the cyclotron was adjusted for multiturn extraction at that time; the external beam pulse had several peaks. With this system it should be possible to continuously monitor the beam energy.

For the polarized-ion injection system a hole will be required in the shield wall between the cyclotron vault and the large experiment room. Radiation leakage was measured from test holes 2 and 4 in. in diameter aligned along the axis of the cyclotron. Exposure rates of 5 to 10 mr/hr of fast neutrons in a well-collimated beam were observed when the cyclotron

beam was 30 μa of 62-Mev protons. This leakage should be significantly reduced by thermalizing and trapping the neutrons on the cyclotron side of the opening.

Advanced Accelerator Studies. — To supplement the development and design work of such separated-orbit cyclotrons as the 50-Mev machine described in preceding reports, an experimental SOC is being designed to accelerate protons to 4 Mev. The machine characteristics may be summarized as follows:

Injection at	1 Mev
Sector magnets	6
Turns	4
Cells, total	24
Cavities, coaxial	6
Voltage, peak	80 kv
Rf power	~25 kw
Momentum compaction factor	~2
Focusing	n = ~0.4
ν_r	~1.5
ν_z	~1.4
Field	8.9–7.0 kilogauss
Radius of path	54 to 104 in.
Aperture	1.5 × 3 in.

The large circumference factor of an SOC leads to large values of ν and to operation below the transition energy. The constant-n lens is suitable for energies up to ~200 Mev. Since this machine will have all the characteristics of any SOC, the solutions obtained to problems of construction and operation apply directly. Experiments are planned to obtain information concerning alignment techniques for pole tips and the effect of random errors in the magnetic field and tips; to evaluate the effects of rf voltage, phase, and noise on the quality of beam transmission; and to determine space-charge limitations for operation under heavy beam loading.

Isochronous Cyclotron Operations. — ORIC operation was scheduled for 1288 hr during the past two-month period, with distribution of the total time as follows: 77% to nuclear research bombardments, 15.2% to beam and machine development, 4.5% to scheduled installation, and 3.3% to scheduled downtime for routine maintenance. Of the approximate 992 hr (77% of total) assigned to nuclear research, beam "on target" time averaged ~67%; target setup and obtaining acceptable beam to the target station, ~15%; startup and restart after shutdown, ~7%; and miscellaneous downtime, such as rf, vacuum, power supplies, and electrical troubles, ~11%.

Extracted beams of 42- to 80-Mev alphas, 36- to 63-Mev protons, 25- and 51-Mev ^3He, 34-Mev deuterons, and 18- to 40-Mev H_2^+ were provided for nuclear physics experiments.

Physics of Fission. – The average number of neutrons ν emitted as a function of m*, the prompt fragment mass, has been experimentally determined for 13-Mev proton-induced fission of ^{226}Ra. The method of determination of ν consisted in correlated three-parameter measurements of both fragment energies and one fragment velocity, as described in ORNL-3928, p. 21. The trend of $\nu(m^*)$ is found to be similar to the familiar "saw-tooth" shape observed in low-excitation fission. The value of ν increases in the fragment mass range 82 to 122 amu, decreases rapidly in the range 123 to 134 amu, and then increases slightly in the range 134 to 144 amu. The sharp drop does not occur at or near symmetry, but occurs in the mass region just below closure of the Z = 50 and N = 82 shells for neutron-rich nuclei. These results are qualitatively consistent with a previously suggested description of ^{226}Ra fission involving liquid-drop model considerations for mass divisions in the region of symmetry (the central peak in the fragment mass distribution), and "fragment-shell" or "fragment-structure" considerations for asymmetric mass divisions. The results are also qualitatively consistent with a prediction by Terrell based on the systematics of neutron emission in low-excitation fission.

High-Voltage Experimental Program: Coulomb Excitation of ^{79}Br and ^{81}Br. – Coulomb excitation studies of ^{79}Br and ^{81}Br have been continued as part of our program to investigate low-lying levels in medium-weight nuclei with $I_0^\pi = 3/2^-$. Information on level properties of ^{79}Br was supplemented through a study of the radioactive decay of ^{79}Br. The energies and proposed spins of the Coulomb excited states in ^{79}Br are 217.2 (5/2), 261.3 (3/2), 306.4 (1/2, 3/2, 5/2), 397.5 (1/2, 3/2), 522.9 (3/2, 5/2), 605.6 (1/2, 3/2), and 760.7 kev (5/2, 7/2). In ^{81}Br, Coulomb excited states are observed at 275.9, 565.9, 767.3, 836.6, and 1308 kev. The B(E2)'s for excitation of these states have been determined. Efforts to explain the level properties by coupling an odd proton in a $p_{3/2}$, $p_{1/2}$, or $f_{5/2}$ orbital to the core in the ground state or first excited state have been initiated.

High-Voltage Experimental Program: Neutron Spectra from the $^{59}Co(p,n)^{59}Ni$ and $^{65}Cu(p,n)^{65}Zn$ Reactions. – The terminal pulsed proton beam (width <1 nsec) from the ORNL 5.5-Mv Van de Graaff has been used to measure (p,n) reactions for ^{59}Co and ^{65}Cu targets. The neutron energy resolution was 10 to 20 kev when thin targets and a 6-m flight path were used. Eleven excited states were observed in ^{59}Ni below an excitation energy of 2 Mev. All the states observed in high-resolution $^{58}Ni(d,p)$ studies [Nucl. Phys. 51, 641 (1964)] are confirmed, and in addition it is found that the state at 1.68 Mev is a doublet with 8 kev separation. The largest difference in partial cross section occurred for the states at 1.31 and 1.35 Mev, where a cross section smaller by a factor of 30 is observed for the excitation of the 1.31-Mev state at E_p = 4.3 and 4.5 Mev. Nineteen states were observed in the first 2 Mev of excitation in the ^{65}Zn nucleus. High-resolution charged-particle work is not available for comparison, and hence many new states are observed. Doublets are found at

1.05 Mev (20-kev separation), 1.26 Mev (11-kev separation), and 1.59 Mev (15-kev separation).

High-Resolution Electron Microscopy. — A detailed study is being performed of the effect on the resolution of the proposed high-resolution electron microscope produced by errors in lens tilts, displacements, and excitations, as well as by beam energy variations. Control of beam momentum to an accuracy of between 10^{-6} and 10^{-7} will be required. It is planned to sense the beam energy by magnetic deflection in a suitable analyzer magnet, and control the energy accordingly. The normal energy spread of a beam extracted from a cathode will be sharply reduced by magnetic analysis at low energy, before the bulk of the acceleration is performed. We are attempting to find a reasonable solution to the problem of regulating the magnetic field of the objective lens system. The use of adjustable permanent-magnet excitation of the lenses and analyzer magnets is a possibility, as is field sensing with Hall probes. An alternative capable of very fine performance is the use of a cryogenic system, with superconducting windings for the essential magnets, although this is unfortunately much too complicated unless we are forced to it. Some study of detector and data systems has been made, with the conclusion that an available image intensifier (RCA) will probably be adequate.

CHEMISTRY

Preparation and Properties of Actinide-Element Oxides: Thoria. — Studies have been conducted to determine the effect of CO_2 on the electrophoretic behavior of colloidal thoria sols. The electrophoretic pattern observed for oxalate-precipitated "CO_2-free" thoria sols appears to be very nearly the same as the pattern for thoria which had not been treated to remove CO_2; that is, there are two electrophoretic bands. However, the same thoria sol when saturated with CO_2 showed essentially only one electrophoretic band, whose constituents moved considerably slower than those of the CO_2-free sol. Preliminary studies with CO_2-free ammonia-precipitated thoria have shown only one electrophoretic band, indicating that the anomaly of multiple electrophoretic bands is not a general property of all thoria.

Preparation and Properties of Actinide-Element Oxides: Urania. — Urania sol having, on a per-liter basis, 314 g of uranium [with 11.9% as U(VI)], 28.4 g of formate, and 14.0 g of nitrate was measured for pH and conductivity while being diluted. The suspension effect on apparent pH was weak, being only about 0.3 pH unit, and the buffering action of the sol with respect to dilution was also weak compared with that for analogous thoria sol. These behaviors indicate weaker adsorption of the nitrate in urania sols than in thoria sols.

Viscometry of this sol and its twofold dilution with water gave reduced viscosities of 13.0 and 11.8 respectively. These viscosities correspond to hydrodynamic flocs of crystallite having axial ratios of 9.6 and 9.0, which are somewhat larger than those found for analogous thoria sols.

Irradiation Effects on Heterogeneous Systems. — An examination of previously accumulated data indicates that there might be a relationship between the catalytic-cracking ability of various solids and the efficiency of radiolytic hydrogen production from water sorbed on various materials. It is known that substituting calcium ion for sodium ion in some molecular sieves enhances the catalytic activity, so experiments were carried out to see if a similar effect was observed for the hydrogen yield. Measurements were carried out on materials such as Linde SK-40 molecular sieve in the sodium and calcium form as a function of water content, and it was indeed found that the calcium form had a greater effect. At 11 wt % water, $G(H_2)$ for the calcium form was 1.3, while for the sodium form it was only 0.4.

Protactinium Chemistry. — Measurements of the solubility of protactinium sulfate in strong sulfuric acid solutions were carried out starting with protactinium sulfate (instead of an oxide or hydroxide) as the solid phase. Consistent results were obtained for an initial set of measurements: solubilities were 0.024, 0.019, 0.036, and 0.15 mg/ml in 19.5, 17.4, 14.4, and 11.6 M H_2SO_4 respectively. In every set of measurements, the lowest solubility was observed in the 17.4 M solution, approximately the composition of concentrated sulfuric acid. Solubilities were higher in all cases after the mixtures were heated, but they returned to the above values after a few days at room temperature. However, when additional amounts of protactinium compound were added to the solutions, there was a substantial increase in protactinium solubility in all cases, and even after two weeks the values had not decreased significantly except in the case of 17.4 M acid.

Additional absorption spectra of protactinium sulfate in 2.5 M H_2SO_4 are being measured, and these will include a broad range of protactinium concentrations to determine if any features of the spectra can be correlated with concentration. Attempts are being made to resolve these spectra, as well as some obtained previously, by using a computer code developed at ORNL and applied to studies of uranyl spectra [J. Mol. Spectry. 18, 247 (1965)]. Preliminary results indicate that the observed spectra can be reduced to three to five Gaussian bands, but we do not yet have enough data to attempt any correlation. However, it was observed that the protactinium spectra are qualitatively quite similar to the spectra of uranyl solutions in the same wavelength range. This is surprising, since Pa(V) is presumed to be very different from the UO_2^{2+} structure and, in particular, not to form a PaO_2^+ ion.

Analysis of Molten Salts by Electrochemical Methods: Voltammetric Studies of Uranium in Molten MSRE Fuel. — A study of the voltammetry of uranium in a sample of $LiF-BeF_2-ZrF_4-UF_4$ (65.0-29.1-5.0-0.9 mole %) in the molten state at 500°C is in progress. This investigation was undertaken to ascertain the feasibility of utilizing rapid-scan voltammetry in monitoring the concentration of tetravalent uranium directly in the molten fluoride salt fuel and also to observe if it is possible by means of this technique to distinguish between tetra- and trivalent uranium.

Well-defined waves that correspond to the reduction of uranium(IV) to uranium(III) were observed at approximately −1.1 v vs a platinum quasi-reference electrode. The waves generally exhibit peaks at voltage scan rates of 1 to 10 v/min, which indicate that the transfer process to the

electrode is mainly diffusion controlled. From a plot of peak current (i_p) vs voltage scan [$(v/sec)^{1/2}$] a value of the diffusion coefficient for uranium was calculated as being approximately 1.5 to 2.0 $\times 10^{-6}$ cm^2/sec. This is the same order of magnitude as that previously observed for iron, nickel, and chromium in molten fluorides.

Well-defined reduction waves were observed at platinum, platinum–10% rhodium, and pyrolytic graphite indicator electrodes. From the standpoint of reproducibility, however, platinum and platinum–10% rhodium appear to be superior to pyrolytic graphite for this particular study. The uranium(III) produced at the electrode interface during the reduction appears relatively stable, that is, slow disproportionation to U^0 and U^{4+}, because on reverse scans a well-defined anodic wave is observed and the ratio of cathodic to anodic currents is close to unity, a fact which agrees with theory for a reversible charge transfer process involving soluble species. G. Mamantov has demonstrated that the addition of uranium(III) to the melt increases the anodic:cathodic current ratio and that this may be a fruitful way of determining the uranium(III) content of fluoride melts.

It is planned to continue the voltammetric studies and to carry out chronopotentiometric studies on the uranium in the MSRE fuel, and to compare the n value (electron change) and the value of the diffusion coefficient calculated by this method to that obtained by rapid-scan voltammetry.

Instrumental Methods for Chemical Analysis: Controlled-Potential Differential DC Polarograph. Investigations of ΔE, Subtractive, and Comparative Modes of Differential DC Polarography. – The design has been completed of a polarograph for differential (two-cell) dc (constant-voltage) polarography with, for example, natural drop-time D.M.E.'s. Investigations of three modes of differential polarography (ΔE, subtractive, and comparative) have demonstrated the utility of these modes, that the performance of this instrument is very satisfactory, and that advantages and simplifications have been realized over prior instrumentation, which increase the practicability of differential polarography. Moreover, this polarograph has been used to establish the feasibility and advantages of our new electrochemical method, one-cell comparative amperometric dc polarography; concurrently, the high degree of reproducibility of measurements made with these polarographic cells and circuitry was demonstrated. Operational amplifier systems provide electrolysis at controlled potential, highly stable initial current compensation, a selection of scan rates, and current averaging and electrical channel gain adjustment, which entirely eliminate (except for the subtractive mode at extreme sensitivity) the need for two synchronized drop-time D.M.E.'s having identical characteristics. An X-Y recorder presents (vs D.M.E. voltage) the current at either D.M.E. or the difference of the average currents at the two D.M.E.'s with identical D.M.E. voltages (subtractive or comparative modes) or with voltages differing by a constant ΔE (ΔE mode). It is also possible to record first-derivative dc polarograms using either the signal from one cell or the subtractive mode signal from two cells as the input to the derivative computer.

Equations for the ΔE-differential mode were derived and verified, and results obtained were compared with those obtained by analogically computed first-derivative dc polarography. True derivative dc polarograms

are recorded by the latter method. In subtractive-mode differential polarography, for example, the signal from a cell containing the supporting electrolyte is subtracted from the signal from the cell that contains the electroactive species of interest. This mode can provide increased sensitivity or selectivity or both. It was shown that polarograms that have about equal accuracy and precision are obtained with Cd^{2+} solutions ($\geq 10^{-4}$ M) in oxygen-saturated and in highly sparged solutions. Two-cell comparative polarography is a scale expansion subtractive technique (using in one cell a reference solution of accurately known concentration) whereby the concentration of the electroactive species in the second (sample) cell is determined with high accuracy and with high precision. Relative errors and relative standard deviations both equal to 0.1% were obtained. This has only been achieved with other methods of comparative polarography, at some inconvenience, by the use of two D.M.E.'s whose drop times are synchronized and which have identical characteristics that must remain very closely matched throughout the voltage sweeps and the series of measurements. Also, the requirement for matching cell voltages is less stringent with our method. The same excellent high accuracy and precision were also achieved with this instrument with our one-cell comparative amperometric dc polarographic method.

Properties of Aqueous Solutions: Species and Thermodynamic Properties. – In hyperfiltration, or reverse osmosis, solute and solvent are separated by forcing a solution by pressure through a membrane which partially rejects solute; the feed solution is at the same time circulated past the membrane to carry away salt that is being concentrated at the interface by the process. We have found that the salt filtration properties of tight cellophanes are substantially increased, and permeation rates decreased, by pretreatment with certain metal ions or the presence of these ions at low concentration in feed solutions. (As used here, the term "tight cellophane" signifies a film which water under 2500 psi permeates at a rate of about 1 cm/hr.) In a typical case, a cellophane which initially rejected about 20% of salt from a 0.05 M NaCl solution rejected over 70% in the presence of 10^{-3} M $ThCl_4$. Permeation rates were decreased, usually by a factor of 2 or 3. In cases for which thorium rejection was measured, it was high, 97% or greater. Other additives found to have a marked effect were Fe(III), Cu(II), Zr(IV), and hydrolyzed U(VI) and Pb(II); Mg(II), Ba(II), La(III), and unhydrolyzed U(VI) and Pb(II) had little effect. The mechanism by which additives affect the cellophane is not clear, but, except for Cu(II), effective ions were either hydrolyzed, or acidities of the solutions were not greatly higher than the pH range for which hydrolysis would be substantial.

Spectrophotometry of Solutions over Wide Ranges of Temperature and Pressure. – Our studies of the effect of temperature on the near-infrared spectra of CO_2 and simple hydrogen-containing solvents (H_2O and D_2O) were extended to include NH_3, n-hexane, and cyclohexane.

We are determining the temperature dependence of the NH_3 spectrum in the condensed state between 0.6 and 1.4 μ. This spectrum, arising from harmonics and combinations of vibrational modes of the NH_3 molecule, is especially complex, because the four fundamental and the combination bands resulting are all infrared-active. We have accounted for the general features of the spectrum in terms of all possible summation bands. The

principal band maxima occurring near 1.04, 0.93, 0.89, 0.80, and 0.65 μ have molar absorptivities of 16.2, 0.85, 1.22, 1.93, and 0.19 ($\times 10^{-3}$ liter $mole^{-1}$ cm^{-1}) in the liquid at the freezing point (−77.7°C). Freezing causes an abrupt shift (decrease) by 62 to 116 cm^{-1}, as compared with about 200 cm^{-1} for comparable bands of H_2O. The wave numbers of the maxima increase slowly and linearly (0.06 to 0.14 $cm^{-1}/°C$) from the freezing point to 60°. For liquid H_2O and D_2O the shifts near the freezing point are initially very large (3 to 5 $cm^{-1}/°C$) and nonlinear, decreasing to (linear) shifts of 0.2 to 0.3 $cm^{-1}/°C$ above about 100°C. The large shifts are consistent with the breaking of strong H bonds. The difference in behavior between water and ammonia is also consistent with the presence of fewer and weaker H bonds in the latter liquid.

Both n-hexane and cyclohexane have a spectrum which consists of complex bands centered at 1.4, 1.2, 1.02, 0.92, and 0.75 μ. The bands at 1.2, 0.92, and 0.75 μ arise from the 3d, 4th, and 5th harmonic absorptions of the C-H stretch fundamentals. The band frequencies and intensities are relatively insensitive to temperature changes, compared with those of NH_3, H_2O, D_2O, and CO_2. No change was detected in cyclohexane due to solidification. These observations agree with the behavior to be expected for C-H bonds, as compared with more polar N-H and O-H bonds.

Radiation Chemistry: Flash Photolysis and Pulse Radiolysis. — We have assembled the fast-recording spectrographic-spectrophotometric detection apparatus and have initiated studies of transient species produced by the flash photolysis of aqueous solutions.

In flash photolysis, to examine the buildup and decay of intermediates on the shortest time scale, it is necessary to minimize the flash duration, the electrical noise, and the scattered light. We studied a number of lamp designs, using several packing materials, and using various rare-gas and rare-gas-quencher fillings. The best light pulse was obtained from a relatively simple lamp constructed of a quartz tube with a coaxial quartz rod between the electrodes. Using a 1-cm-ID tube and a 7-mm rod, the discharge is constrained to a $1\frac{1}{2}$-mm annulus. The resistance of two 25-cm lamps in series is adequate to provide critical damping, and essentially all of the power is dissipated in the lamps. For a discharge of 300 j, 90% of the light is emitted in 3.2 μsec. Also, the scattered light and electrical noise have been reduced to the extent that we can compare the buildup of transients with integrated energy input to within 0.1 μsec. Thus it was shown that in the photolysis of basic hydrogen peroxide solution with oxygen present, O_3^- is produced rapidly during the flash by reaction of O_2 with the O^- produced initially; with no oxygen present, O_2^- is produced by the somewhat slower reaction of HO_2^- with the O^-.

The absorption spectrum of e^-_{aq} is known from pulse radiolysis studies, and has proven very useful in kinetic studies of aqueous systems. The absorption spectrum of its counterpart, the hydroxyl radical, had not been established. We have now measured the absorption spectrum of the OH radical produced by the flash photolysis of pure water and H_2O_2 solution. There is a broad peak in the absorption centered at ~235 nm. The spectrum is similar to that of O_2^- (or HO_2) except that it extends further into the visible region (to ~400 nm), and the extinction coefficient at the peak is about half that of O_2^- ($\epsilon_{OH} \sim 500$).

METALLURGY AND MATERIALS

Deformation of Crystalline Solids. — When metals are rolled, the crystalline lattices become oriented relative to the sheet thickness and the direction of rolling. This results because deformation is accomplished by the slipping of the atoms relative to each other, which takes place on certain crystallographic systems and leads to rotations. A description of the resulting orientations is important in understanding the deformation as well as in the use of the metal, since the physical properties are now anisotropic.

Conventionally the intensity of several crystallographic planes relative to the physical directions has been used to represent the orientations. Such a representation makes possible the recognition of only grosser features.

One can define a function in three angular variables that represents a complete description of the texture; the conventional representation is related to averages of the function along lines threading through this volume. While the need for such a function has been recognized for some time, no one has worked out the transformation of conventional data into these new coordinates.

So far we have defined minimum volume, determined the important symmetries, and programmed an iterative technique to carry out an inversion. We are now correcting known errors in the calculations and expect to obtain entirely satisfactory inversions shortly.

Reactions at Metal Surfaces. — We have continued our studies of the thin film stage of alloy oxidation. Significant differences in oxidation kinetics, oxide surface topography, and degree of strain in the oxide were noted between the 110 planes of a Cu—1.0 at. % Ni single crystal and a 99.999% Cu specimen. At 250°C an approximately parabolic oxidation rate curve was observed for the (110) of pure copper, whereas an incubation period occurred during the oxidation of the alloy crystal. During the initial stages the alloy oxidized more slowly than pure copper, but after 30 min at 250°C it oxidized twice as rapidly as pure copper. Electron microscopy studies of surface replicas of oxidized specimens revealed the alloy oxide to be smoother, and initial x-ray results indicated that the alloy oxide films were more strained during the induction period than were their counterparts on pure copper. Further efforts to correlate the state of strain in the films with changes in the oxidation rate are continuing.

Theoretical Research. — We have begun preliminary programming on the Green's function method for calculating the electron energy bands of crystalline solids.

Spectroscopy of Ionic Media. — The coordination geometry of ions in molten salts is expected to play an important role in determining the physical and chemical properties of these media. We have recently completed a study (by electronic spectroscopy) of the coordination geometry of nickel ions in melts and glasses of 0.1 to 33.3 mole % $NiCl_2$ mixed with tri-n-butylbenzylphosphonium chloride at temperatures of 22 to 174°C. We concluded that the nickel in these melts was present almost exclusively as approximately tetrahedral $NiCl_4^{2-}$ anions under all conditions studied.

So that we could interpret absorption intensities in terms of volume concentrations, the densities of molten mixtures containing 5.03 to 33.44 mole % $NiCl_2$ were measured as functions of temperature. The molar volume at a fixed temperature was a linear function of the mole percentage.

Transition Metals and Alloys. — Previously we established that the Mössbauer effect could be observed in ^{73}Ge following Coulomb excitation (ORNL-3864, p. 28). In the first experiment a 0.01-in.-thick absorber (136 mg/cm^2 Ge) was used. The observed maximum absorption was an order of magnitude smaller than the expected value. In a new experiment on the ORNL Tandem Van de Graaff accelerator we observed the Mössbauer effect in ^{73}Ge with two absorbers, 68 and 272 mg/cm^2 thick. The thickness dependences of the observed line width and of the maximum absorption allow the separation of the recoilless fraction of target and absorber. We found that the recoilless fraction of the absorber must lie in the range 0.12 to 0.17, in agreement with the value 0.125 obtained from the Debye formula. The recoilless fraction in the target is then about 0.008 to 0.011. This strong reduction of the recoilless fraction in the target is probably caused by the ion bombardment, although at the moment it is not clear which of the possible mechanisms actually influences the recoilless fraction.

Preparation of Pure Materials. — The Stockbarger-Bridgman method has proved to be so successful for preparing large pure single crystals of LiF that its application has now been extended to the growth of other fluorides and complex fluorides. A second Stockbarger crystal-growth apparatus, which is capable of operation at temperatures as high as 1800°C, was constructed for this work. In initial tests of the apparatus, crystals of $7NaF \cdot 6ZrF_4$ were grown by lowering a melt of the stoichiometric composition through the freezing gradient at 0.5 mm/hr. Single crystals of dimensions 10 × 10 × 6 mm were obtained. A recent determination of the crystal structure at ORNL showed that subunits within the $7NaF \cdot 6ZrF_4$ structure enclose a cavity containing an asymmetrically positioned fluoride ion. That its configuration and stoichiometry are both unusual has begun to generate interest among research workers in measuring the physical properties of $7NaF \cdot 6ZrF_4$. In accord with this interest, single crystals of the compound were furnished on a loan basis to the Sandia Corporation for investigation of its ferroelectric properties.

CONTROLLED THERMONUCLEAR RESEARCH

The DCX-2 Injection-Accumulation Experiment. — The highly energetic proton plasma trapped in DCX-2 usually consists of two components, the "side lobe plasma" that retains the axial energy it had at injection and the more energetic "central peak plasma" that is formed with very small axial energy. During this report period the central peak plasma and the rf spectrum and its decay after beam turnoff have been studied with hydrogen gas breakup in the presence and in the absence of a dense side-lobe plasma.

It was found that suppressing the side-lobe plasma with an obstructing paddle that left the central peak plasma undisturbed increased the

central peak and total plasma densities. The intense rf present with the molecular ion beam on persists for a considerable time after beam turnoff with the side lobes unobstructed. With the side-lobe plasma suppressed, these rf fields at the cyclotron harmonics decay rapidly to noise in about a millisecond, although the central peak plasma density (1.6×10^8 ions/cm^3) has not changed appreciably in this time (half the energetic protons remain after 250 msec).

The stability of the central peak plasma with respect to rf emission after beam turnoff has been used to test some theoretical models of ion cyclotron instabilities. The electron density in these experiments significantly exceeds the highest instability threshold predictions of the Soper-Harris theory or the Guest-Dory theory modified to include the finite plasma size. The DCX-2 results indicate that finite-size effects must be incorporated more carefully into the theory.

It is observed that under some conditions the molecular ion harmonics are double, with peaks above and below $n\omega_{ci}$. When a probe is inserted to stop the reflected molecular ion beam, the lower peak disappears, suggesting that under these conditions the rf seen at the molecular ion harmonics results from a streaming interaction. These double peaks are also seen at the proton harmonics with a strong peak at $n\omega_{ci}$ between the side peaks. It is found that as the pitch angle of the side-lobe plasma decreases, the frequency separation of the side peaks also decreases, indicating that some of the rf at the proton harmonics also results from streaming interactions, this time involving the protons.

End-injection studies are continuing in an effort to increase the amount of proton current that is being reflected off the mirror opposite the injector by optimizing the injection angle.

Active and Passive Stabilization of Plasmas and Ion Beams. — The development of a computer program to simulate beam plasma interactions in the calutron has been temporarily stopped by lack of sufficient memory in the computer being used.

Various boundary impedance networks are being tried in DCX-1. All give large changes in the amount of rf radiated by the plasma, but only the most recent configuration shows any correlation between rf decrease and plasma increase.

Properties of Vacuum Arcs. — In the vacuum carbon arc, earlier work here had established the existence of an anomalous equilibrium temperature situation: the carbon ions were much hotter than the electrons. An explanation was proposed according to which electron collisions excite C^{2+} ions into a metastable state that is known to exist, and, as a result of subsequent collisions between the metastable ions, this energy of excitation is released as kinetic energy. Such a mechanism implies a substantial continuing population of the metastably excited ions. We have now performed experiments upon the self-absorption of the ultraviolet spectral lines from the carbon ions, and as a result we can now state that the metastable C^{2+} ions are indeed sufficiently abundant to support the proposed heating process.

Experimental Plasma Physics. — We have developed a plasma source in which cesium chloride is dissociated, and the cesium ionized on a hot surface. Such thermal plasma sources have been very useful in that they

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produce a dense plasma, $n_e > 10^{10}$ cm^{-3}, that is quiescent. Our source is unique in that the hot surface is electrically nonconducting. Previously used surfaces were metallic, and acted as a short circuit across the end of a magnetically supported plasma column. This short circuit made plasma instability studies difficult. The short circuit does not exist in our apparatus.

Magnetics and Superconductivity. – The single-sweep experiments on flux flow through the surface of hard superconducting cylinders in isothermally changed longitudinal fields have been continued. Various samples with different surface treatments are being used, and the results are reproducible. These experiments are closely related to the theoretical calculations by Fink, Park, and Rothwarf.

Preliminary additional experiments showed the great influence of the surface currents on ac measurements. Taking the results of our single-sweep experiments into account, discrepancies between the current density values determined by C. P. Bean's method and values measured in different ways can be understood. Especially at low external fields, appreciable corrections of the data obtained with the first-mentioned method are necessary.

Vacuum Studies for Fusion. – Study has continued of the mass spectral changes due to addition of hydrogen gas to an oil-diffusion-pumped vacuum system. Previously, organic compound mass peaks were found to increase significantly upon addition of hydrogen or deuterium. These observations were interpreted as being due to the presence of adsorbed organic compounds in or near the spectrometer ion source which reacted with thermally dissociated hydrogen or deuterium atoms. This study was pursued with the design and construction of a trap which did not greatly alter the gas conductance but which could be operated to prevent surface transport to the spectrometer source and which could be operated at temperatures higher than the ambient temperature of the vacuum system. It was found that the organic spectra were decreased markedly by operation of the spectrometer at about 100°C after baking at 300° for several hours and that the addition of hydrogen then yielded insignificant changes in the organic spectra, still with some increase in the water vapor spectrum. The earlier interpretation appears to be substantiated. Application to a practical system was found to require control to ±1 to 2°C of the system temperatures near the spectrometer in order to avoid profound changes in the mass spectrum.

The response time of the spectrometer and of a fast ion gage was decreased to 0.009 sec to allow a study to be made of the pressure pips due to water being periodically evolved from an elastomer O-ring in the spectrometer test facility. (Air pips have been found which originate in an aluminum-wire-gasketed flange.) The rise-time constant was found to be equal to or less than this number, 0.009 sec. As predicted, the decay-time constant was found to be fixed by the system geometry at 0.10 ± 0.01 sec. (The calculated value of the volume-pumping speed ratio was 0.11 sec.) Thus for this type of pressure pip the gas (water, 95%; carbon monoxide, 5%) is introduced during a time period no longer than 0.01 sec. This determination eliminates some processes from consideration as the origin of this type of gas source.

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BIOLOGY AND MEDICINE

TERRESTRIAL AND FRESHWATER ECOLOGY

Movement of Nuclides in Terrestrial Environment. – Detailed analyses of soil cores pulled from each of these plots (bare, clipped, and tall) showed marked evidence of ^{137}Cs movement along the surface due to runoff and erosion and relatively little evidence of downward movement into the profile.

The upper 2.5-cm layer contained on the average 54, 37, and 77% of the applied ^{137}Cs on the bare soil, clipped meadow, and tall meadow plots respectively. Variations in concentration, amounting to as much as 30%, were observed between locations and could be ascribed to the microtopography and density of vegetation.

The ^{137}Cs present in the soil is concentrated within an upper 2.5 cm of the profile. However, if the specific activity of the eroded material is taken into account, an upper 1 cm would have to be taken as the zone of heaviest concentration.

During the first nine weeks of operation, 6, 4, and 2% of the ^{137}Cs moved into the 2- to 10-cm layer of bare soil, clipped meadow, and tall meadow plots respectively. During the following ten-month period, an additional 4% of the applied ^{137}Cs moved down into this layer on the bare soil and tall meadow plots, but no net increase was observed on the clipped meadow plot. On the average, after one year, only 1% of the applied ^{137}Cs was found in the 10- to 20-cm layer on all plots.

Very low recovery of ^{137}Cs from the soil cores on the clipped meadow plot indicated that more radionuclide should be present on the litter material than was reported earlier. Studies are under way to verify this observation.

Three additional small-plot studies, incorporating bare soil, mulch, and tall fertilized meadow, have been started. These plots are to serve as replicates for the previous studies and are to provide information on the relative movement and deposition of radionuclides within the plot. The lower half of each plot has been dosed with ^{134}Cs and the upper with ^{137}Cs. In addition, these plots will isolate the litter effect (on mulch plot) and help to evaluate seasonal variations with regard to radionuclide movement by runoff, erosion, infiltration, and possible plant translocation.

RADIOLOGICAL PHYSICS, HEALTH PHYSICS, AND RADIATION INSTRUMENTATION

Radiation Physics: Optical Properties of Sodium in the Vacuum Ultraviolet. – The index of refraction of sodium films evaporated onto a quartz substrate has been determined in the spectral region from 420 to 2000 A from interference patterns observed in the reflected beam and from the critical angle marking the onset of "total reflection." The extinction coefficient is shown to be less than 0.015 in this region. It is found that the net phase change resulting from reflections at the vacuum-sodium interface and the sodium-quartz interface is 0.3π. The thickness of a

film as calculated from interference patterns agrees with that calculated from the measured mass of the film. The growth of an oxide layer on the surface of a film is shown not to appreciably alter the positions of interference maxima and minima. The interband contribution to the real part of the dielectric constant was found to decrease from 2000 A to approximately 850 A, and then remained constant down to 500 A. This behavior agrees with calculated band structures which predict that the highest-energy transition between the conduction band and the first excited band is induced by photons with a wavelength of 880 A. The volume plasmon energy was found to be 5.69 ± 0.03 ev, in agreement with the value of 5.70 ev determined by Kunz from the characteristic energy losses observed in an electron beam experiment.

Radiation Physics: Formation and Autoionization of SF_6^-. — Using microscopic reversibility, the autoionization lifetime of SF_6^- was calculated. Agreement with experiment is good. Implications of this agreement have been studied with respect to the mechanics of "superexcited" molecules.

Radiation Physics: Finite-Medium Effects in Radiation Chemistry of Gases. — Expressions have been formulated from which the extent of all neutralization of charged species in irradiated gases may be calculated. Under most circumstances ambipolar diffusion will predominate. Comparison with experiment suggests that convection currents are usually present which enhance surface neutralization to a largely uncontrollable extent.

Radiation Physics: Effects of Phantom Geometry on Dose Distribution. — To assist in the evaluation of the hazard associated with exposure to high-energy neutrons and protons, the effect of the geometry of the tissue phantom on the energy deposited as a result of irradiation by a broad beam of protons and neutrons with energies up to 400 Mev has been investigated. In previous investigations [Second Symposium on Protection Against Radiation in Space, Gatlinburg, Tennessee, 1964, NASA SP-71, p. 161; Health Phys. 10, 783 (1964); Health Phys. 12, 227 (1966)] it was assumed that a body can be approximated by a 30-cm-thick slab of tissue. The present calculations were based on a more realistic model in the form of a right elliptical cylinder, which approximates the dimensions of a human torso. The calculations were performed under the assumption that the cylindrical phantom was exposed to neutrons and protons isotropically distributed in space, as well as neutrons and protons normally incident with respect to the planes passing through the axes of the phantom. Monte Carlo methods were used in calculating the dose at various points within the cylindrical phantom, with particular reference to the location of critical organs. The data are presented in a flexible manner, so that any relationship between quality factor (QF) and linear energy transfer (LET) can be used in order to obtain estimates of dose equivalent at any point within the phantoms under consideration. The calculations take into account nonelastic and elastic interactions, nuclear evaporation, and nuclear recoils.

Radiation Physics: Calculation of Radiation Dose Due to High-Energy Protons. — The Monte Carlo technique has been used to obtain estimates of dose and dose equivalent in a homogeneous tissue slab irradiated by broad beams of monoenergetic protons having energies extending from 400 Mev into

the Bev range. The protons were assumed to be normally incident on one face of the tissue slab. The calculations were simplified greatly by using the straight-ahead approximation. Specifically, in the cascade processes the emitted nucleons were assumed to have the same direction as the incident nucleon which initiates the nuclear reaction. Statistical information obtained by Metropolis et al. [Phys. Rev. 110, 204 (1958)], such as the average number and energy distribution of protons, neutrons, and pions emitted in a cascade process and the excitation energies of the residual nuclei, was used in preparing these calculations. However, the code was written with all the statistical information being read in as input, and, therefore, as new statistical information becomes available it can be easily incorporated. Both absorbed dose and dose equivalent were calculated at various depths in the phantom. The estimates of dose equivalent were made on the basis of the QF-LET relationship endorsed by the International Commission on Radiological Protection for occupational exposure.

Radiation Physics: Swarm Determination of the Cross Section for Momentum Transfer in Ethylene and in Ethylene Mixtures. — Data have been obtained on the temperature dependence of electron swarm drift velocities, w, in ethylene. The analysis of the data gave a velocity-independent cross section for momentum transfer, σ_E, for ethylene at thermal electron energies. Using a constant σ_E, investigation was made of the dependence of w_E/w_M on gas concentration, where w_E is the drift velocity in ethylene, and w_M that in mixtures of ethylene with a polar impurity, P. Three regions of concentration were distinguished: initial, intermediate, and final. The initial region can provide information about the velocity dependence of σ_P, the intermediate region (better suited for experimentation) can easily provide σ_P if the velocity dependence of σ_P is known, and the final region provides a new way to measure the drift velocity for pure P by simply measuring w_E/w_M in mixtures of P with ethylene. The latter is extremely important when P cannot be studied alone.

Radiation Physics: Use of Fourier Transforms in the Unfolding of Experimental Data. — It has been shown that when experimental data are subject to smearing effects which may be approximated in convolution integral form, Fourier transform methods used in conjunction with a nonlinear least-squares analysis may be of use in recovering the unsmeared data. Application of the method to the unfolding of experimental data in a time-of-flight experiment in gaseous electronics has been made.

Radiation Physics: Modified Formulation of Dielectric Properties of Absorbing Media. — It is suggested that the dielectric constant for a dispersive and absorbing medium be reformulated by replacing the conventional expression, represented by a complex number, by a real expression which is independent of the dissipating properties of the medium. In the conventional description of a dispersive and absorbing medium, the dielectric constant is represented by a complex quantity; that is, it has the form $\epsilon = \epsilon_1 + i\epsilon_2$. Such a representation has caused considerable difficulties in the analysis of the energetic behavior of electrical fields interacting with such media. Some of the fundamental concepts, such as the mean energy density U_e stored by an electric field E oscillating at a frequency ω, have not as yet been formulated in terms of macroscopic parameters of

an absorbing and dispersive medium. Thus, the well-known expression $U_e = (1/16\pi)(d/d\omega)(\omega\epsilon)E^2$, derived by Brillouin for a nonabsorbing medium (ϵ is real), does not give physically acceptable results in the case of absorption. The present analysis deals with a new formulation of the dielectric constant which eliminates the above-mentioned difficulties and discrepancies.

Radiation Physics: Ladder Corrections to the Static RPA Dielectric Constant and to Positron Annihilation in Metals. — A calculation of the static dielectric constant of a free electron gas was made in which account was taken of repeated (ladder) electron-hole interactions. An analytic approximation was given for the results. The effective interaction thus obtained is used in a high-order (ladder) perturbation calculation of the two-photon annihilation rate of positrons in an electron gas. The results agree quite well with experimental data for $r_s \leq 5$, where r_s is the radius of a sphere containing, on the average, one electron in the gas measured in units of the Bohr radius. Comparison was made with similar calculations made using the static random phase approximation and Hubbard interactions.

Radiation Physics: Study of Elementary-Particle Interactions for High-Energy Dosimetry. — Types of interactions have been classified from the standpoint of their relative importance in the dosimetry of particles with energies up to 10^{10} ev or more. The known elementary particles were treated individually; threshold energies, cross sections, and important reactions leading to their production and absorption were determined. The principal reactions by means of which these particles can contribute to dose and dose equivalent were studied. The data provide a starting point for detailed calculations or analyses of dose from high-energy particles.

Radiation Physics: Investigation of the "Surface Plasma" Radiation from Electron-Bombarded Silver. — The optical emission spectrum from a thick silver target bombarded by high-energy electrons at grazing incidence shows a peak at 3500 A. This peak was identified by Boersch et al. [Z. Physik 187, 97 (1965)] as being due to the decay of surface plasmons. In an effort to verify this hypothesis, the radiation emitted by self-supporting silver foils 700 to 1700 A in thickness was investigated as a function of angle of electron incidence, foil thickness, and degree of polarization. For a foil 700 A in thickness, the peak at 3300 A characteristic of transition radiation was found in the emission spectrum when the foil was bombarded by normally incident electrons ($\theta_i = 0°$). The wavelength at which the peak appeared increased monotonically with increasing angles of electron incidence until it appeared at $\lambda = 3400$ A for $\theta_i = 87°$. The dependence of the peak intensity on θ_i, electron energy, and polarization agrees with the bremsstrahlung theory of Howe and Ritchie [ORNL-TM-1105 (August 1965)] for electrons of nonnormal incidence.

Radiation Physics: Radiation from Thick Silver Foils Bombarded by Electrons at Grazing Incidence. — Silver foils several thousand angstroms in thickness were bombarded by electrons at grazing incidence. A peak was observed at approximately 3550 A in the optical emission spectrum. For an angle of incidence of 89° from the foil normal, the peak intensity was approximately ten times the previously observed intensities of transition radiation at 3300 A from silver foils bombarded with normally

incident electrons. The intensity was found to be directly proportional to the electron energy from 40 to 80 kev and showed a marked dependence on the quality of the foil surface. The results are in agreement with the findings of Boersch et al. [Z. Physik 187, 97 (1965)].

Radiation Physics: Cyclotron Excitation of Electromagnetic Waves by a Gyrating Electron Beam. — When a tenuous electron stream gyrates around the field lines of a static magnetic field in the presence of a stationary plasma, electromagnetic waves are excited which propagate perpendicular to the magnetic field. Previous theoretical studies involved the assumption that these excited waves are almost longitudinal (quasi-static approximation). Such an assumption was not made in this analysis. It was shown that the excited waves have the electron-cyclotron frequency or some multiple thereof. The electric intensity vector rotates in a plane perpendicular to the magnetic field, the phase velocities are of the order of or exceed the velocity of light, and for certain plasma-beam systems lower harmonics of the electron-cyclotron frequency cannot be excited. Expressions have been formulated for the rate of growth of the waves as a function of the plasma-beam parameters and the harmonic number.

Radiation Physics: Coupled Electron-Hole Cascade in a Free Electron Gas. — The coupled electron-hole cascade in an electron gas has been described by a space- and angle-independent transport equation. Analytic cross sections employed for the various processes are appropriate to a high-density electron gas and for low to moderate excitation energies. The solution of the integral transport equation may be expressed in analytic form. It was found that the cascade process results in the accumulation of extremely large numbers of electrons and holes close to the Fermi surface.

Internal Dose Estimation: Age-Dependent Model for ^{90}Sr Metabolism. — When exposure of a population to radionuclides via the dietary pathways occurs, estimates of the deposition in the body and the various organs and the resulting doses to body organs and tissues are needed. All such estimates depend upon a model for the metabolism of the material by the human body. For radionuclides of long radioactive half-life, this model must include data on the retention in the body over many years. Strontium-90 has been measured in organs and tissues of individuals of all ages during the past ten years, although data are relatively scanty for the younger age groups. These data, along with estimates of the dietary intake, can be used to test metabolic models for ^{90}Sr metabolism.

An age-dependent model for ^{90}Sr metabolism by humans has been developed by a modification of the single-exponential metabolic model. A study of data on dogs given ^{90}Sr intravenously suggests that the fractional amounts taken up by the skeleton vary with age. The values of the uptake parameter for individuals of various age groups are estimated using Comar's observed ratio model for metabolism of ^{90}Sr and data on the Ca content of the body. However, the rate of elimination from bone is assumed to be the same for individuals regardless of age. The model has been used to estimate the levels of ^{90}Sr in human beings for the years 1957, 1958, 1960, 1961, 1962, and 1963 using Kulp's estimated dietary intake. The actual diets consumed by the individuals whose bones were sampled remain unknown, and so estimated diets for individuals of these

ages were used in obtaining the estimates of bone levels. Taking into account this lack of precision of the data on intake, the estimates produced by the model are found to be in reasonable agreement with the observed values in bone samples.

TRAINING, EDUCATION, AND INFORMATION

Professional Health Physics Training. — The course in General Health Physics (Physics 473) at the University of Tennessee was completed. The course in Applied Radiation Physics (Physics 234) at Vanderbilt University will continue until June. There are eight students enrolled in this course.

Three 1-hr lectures in Applied Health Physics were given to personnel of the Biology Division at the request of the division director. Two 2-hr lectures were presented to the Institute in the Fundamentals of Radiation Protection at the Nashville Center of the University of Tennessee. A two-day lecture and tour program was arranged for the University of Arkansas Radiological Health course. Two lectures were given for the Medical Radioisotopes course at ORAU. Several lectures were given at the educational series of the East Tennessee Chapter of the Health Physics Society.

Thirty-three colleges and universities were visited by senior staff members of the Division. Health physics research and educational activities were discussed, as well as the U.S. Atomic Energy Commission Fellowship Program.

ISOTOPE DEVELOPMENT

Stable Isotopes Development. — Alterations in collector design have enabled simultaneous collection of ^{196}Hg (needed for production of radioactive ^{197}Hg) and ^{204}Hg (required for cross-section measurements). In the initial separation of high-purity ^{196}Hg, all other mercury isotopes were rejected by the collector; through a series of modifications in equipment design, ^{204}Hg can now be collected at a useful purity level (~60%) without decreasing the purity level or collection rate of ^{196}Hg.

Recent experiments have shown that palladium in a heated region can be transported by CCl_4 vapor to produce a useful calutron feed. Previously, palladium compounds had been found too unstable for use as source materials, and isotopic separation involved transport of palladium vapor formed by electron-bombardment heating of metallic palladium charge material. In the present work, ion source ovens operate at ~1500°C, and corrosion of source components exposed to CCl_4 is a limiting factor in the usefulness of this new approach for palladium separations. One run (which failed because of corrosion problems) operated successfully for 30 hr at outputs equal to those achieved before. The isotopic purity of the collected samples analyzed showed definite improvement (^{102}Pd increased from a previous high of 50.9 to 75.6%, ^{104}Pd from 78.3 to 92.2%, and ^{105}Pd from 78.1 to 94.7%).

The relationship between the calutron arc and source optics is being studied. The equipment in use facilitates visual correlation between the

ion beam profile and the arc parameter being varied. The ultimate goal of these investigations is to provide an ion source that will emit an ion beam having a very limited angular divergence. The development of such a source would promote the sector separator concept and enhance the usefulness of recently developed calutron receivers.

Target Preparation. — Evaporation-condensation techniques have been used to prepare supported targets of arsenic on beryllium substrates. Because of the low sticking coefficient of arsenic vapors on metal surfaces, attainment of film areal densities >1 mg/cm^2 required excessive quantities of evaporant. Furthermore, the tenacity of the arsenic coating on beryllium was found to be poor. Experimental evidence indicated that this latter problem was associated with impurities in the elemental arsenic being evaporated; only material of >99.9% purity yielded satisfactory films. Efforts are being made to investigate substrate parameters in order to derive the most suitable combination of substrate temperature, evaporation rate, and substrate cleaning which can be used to obtain heavy deposits on beryllium substrates.

A gross estimate of the vapor pressure of Cm_2O_3 (or a suboxide species of ^{244}Cm) was attempted by evaporation of material from molybdenum and tungsten tubular crucibles using electron bombardment heating. Comparison of data obtained, in terms of mass loss from the crucible as a function of time at a given temperature, with the calculated weight loss of PuO_2 at the same temperature indicated that $P_{CmO_x} = 5 \times 10^{-7}$ atm at 2100°C.

Development studies were initiated to determine condensation parameters as a function of the bonding capability of selenium and tellurium on aluminum surfaces. It is hoped that this investigation will delineate requisite conditions for permanent bonding of ^{210}Po to aluminum and titanium surfaces at temperatures up to 1000°C. Samples of ^{210}Po on these substrates have been requested by Wright-Patterson Air Force Base. Efforts to bond tellurium to aluminum were totally unsuccessful, and recourse to an intermediate coating of erbium metal was attempted with marginal success. Bonding was maintained up to 200°C, but quantitative loss of tellurium was observed at >200°C; tellurium vapors did not condense at substrate temperatures of ~300°C. Preliminary results indicated that an intermediate coating of yttrium metal may yield bonding of sufficient stability to withstand surface temperatures >300°C.

Radioisotope Research and Development. — A detailed study of the neutron flux of the HFIR is in progress in order to evaluate its full potential for radioisotope production. The flux mapping will cover the various irradiation facilities, and thermal, epithermal, and fast neutron fluxes will be investigated. The monitor assemblies have been fabricated and are ready for a scheduled irradiation at 100 kw. Future study will include spot checks on the validity of extrapolating the low-power readings to high-power flux and to determine the effects of the transuranium program target rods and the exhausted fuel core on the neutron fluxes.

A computer code has been written to treat the problem of neutron capture chains in cycled irradiations with intervening periods of cooling and time-dependent neutron fluxes not only for long-term irradiations in the ORR, but especially in future HFIR irradiations. It is also of interest in production chains when one of the steps involves decay of a radionuclide; ^{242}Cm production is an example of this type. The program

numerically integrates the differential rate equations of the system and is adapted to cycled irradiations and a time-dependent flux, which should be particularly important in the HFIR, where flux variations during each cycle are considerable. The program was written for ^{242}Cm production and must be rewritten for other isotopes, but such changes are easily incorporated. The program provides both printed and graphic output.

A computed effective cross section for the neutron-capture conversion of ^{125}I to ^{126}I was compared with experimental data. The experiment was performed with ^{124}Xe as the target. The reactions are as follows (the diagonal arrows indicate radioactive decay):

$$^{124}Xe \xrightarrow[\text{200 barns}]{\hat{\sigma}} {}^{125}Xe \xrightarrow{\text{18 hr}} {}^{125}I \xrightarrow[\text{1360 barns (computed)}]{\hat{\sigma}} {}^{126}I \,.$$

$$^{125}I \xrightarrow{\text{60 days}} \qquad {}^{126}I \xrightarrow{\text{13.2 days}}$$

Two vials, each containing 2.86% ^{124}Xe, were irradiated in hydraulic tube No. 13 of the ORR for one and four days respectively. Although the agreement between the experimental and calculated yields was not good, the assumptions used to calculate the production of ^{126}I from ^{125}I appear to be reasonable.

Tellurium-123m has potential medical use in being substituted for sulfur in methionine in a manner similar to ^{75}Se substitution. Tellurium-123m has a 0.159-Mev gamma ray and a 0.035-Mev x ray, which may make it useful in diagnostic scanning procedures and as a long-lived gamma standard. The gamma-ray spectrum of 104-day ^{123m}Te is very close to that of ^{123}I, a newly developed 13-hr iodine radioisotope. In large quantities, ^{123m}Te may be useful as a portable x-ray source for radiography. A 9.4-mg sample of 94.8% ^{122}Te was irradiated for 25 days in the Savannah River Reactor at a flux of 1.4×10^{15} neutrons cm^{-2} sec^{-1} to yield 12 mc of ^{123m}Te with no other tellurium radioactivities. The thermal-neutron cross section of the $^{122}Te(n,\gamma)^{123m}Te$ reaction was ~0.35 barn. Previous experiments have shown the reaction to have a high cross section for resonance neutrons; therefore, larger yields can be expected from a high-fast-flux reactor, such as the HFIR, than from the well-thermalized flux in the Savannah River Reactor.

Testing of the tritiated-phosphor-illuminated U.S. Army compasses has been completed, and the detailed results have been transmitted to the U.S. Army Mobile Defense Command. In general, the results indicated that this particular type of compass is contaminated during and after manufacture to a level above that normally considered acceptable. Certain areas of the compasses had tritium smears in the range 10^5 dis/min.

Classification testing of commercial prototype ^{144}Ce-plated sources being considered for use in controlling certain manufacturing operations in the automobile industry has been completed. These sources, which were mechanically sealed, showed an unusual degree of containment of the radioactive material under simulated accident conditions and are considered to be satisfactory, from a safety standpoint, for the intended application.

The first pass of the second 2000-curie batch of ^{85}Kr processed in the thermal-diffusion equipment was completed in March; the product showed an isotopic concentration of 6% ^{85}Kr. This is substantially less than the expected 15 to 18%, and a subsequent investigation indicates that several valves permitted depleted gas from the ends of the column to leak into the product section. The equipment is undergoing maintenance, and faulty valves are being replaced; a radiation monitoring system is being installed to provide a method of locating the peak enrichment of ^{85}Kr.

CIVILIAN APPLICATIONS OF NUCLEAR EXPLOSIVES

Chemical Applications of Nuclear Explosions. — Measurement of rates of exchange of tritium and hydrogen in water and natural gas is being made to aid in the feasibility studies of the stimulation of gas production from wells by nuclear explosives (Project Gasbuggy). After 145 days of mixing methane and tritiated water vapor (100°C, 1 atm, and 1 mc of tritium per liter of vapor), the rate of formation of tritiated methane is about 2.5×10^{-6} mc liter^{-1} day^{-1}.

[redacted]

OPEN LITERATURE PUBLICATIONS

Alexeff, I., and W. D. Jones, Erratum: "Collisionless Ion-Wave Propagation and the Determination of the Compression Coefficient of Plasma Electrons," Phys. Rev. Letters 16, 675 (1966).

Ard, W. B., R. A. Dandl, and R. F. Stetson, "Use of Silicon Solar Cells as Detectors of Energetic Electrons from Plasmas," Rev. Sci. Instr. 37, 380 (1966).

Auxier, J. A., J. S. Cheka, F. F. Haywood, T. D. Jones, and J. H. Thorngate, "Free-Field Radiation-Dose Distributions from the Hiroshima and Nagasaki Bombings," Health Phys. 12, 425 (1966).

Baldock, Russell, "Some Problems Relating to the Thermal Emission of Ions in the Mass Spectrometric Determination of Isotopic Abundances of Neutron Cross-Section Measurements," pp. 749–54 in Advances in Mass Spectrometry, vol. III, Institute of Petroleum, London, 1966 (Proceedings of a conference held in Paris, France, September 1964).

Boegly, W. J., Jr., R. L. Bradshaw, F. M. Empson, W. F. Schaffer, Jr., F. L. Parker, and J. O. Blomeke, "Project Salt Vault: A Demonstration Disposal of High-Level Radioactive Solids in Lyons, Kansas, Salt Mine," Health Phys. 12, 417 (1966).

Boyd, G. E., A. Schwarz, and S. Lindenbaum, "Structural Effects on the Osmotic and Activity Coefficients of the Quaternary Ammonium Halides in Aqueous Solutions at 25°," J. Phys. Chem. 70, 821 (1966).

Brown, G. N., and F. G. Taylor, Jr., "Interaction of Radiation and Photoperiodism in Xanthium Chinense: Effects of Variation in Time of Radiation with Respect to Photoinduction," Radiation Botany 6, 145 (1966).

Chernock, W. P., R. M. Mayfield, and J. R. Weir, Jr., "Cladding Materials for Nuclear Fuels," Proc. Intern. Conf. Peaceful Uses At. Energy, 3rd, Geneva, 1964 9, 35 (1965).

Christophorou, L. G., and J. G. Carter, "Excimer X-Ray Investigation," Nature 209, 678 (1966).

Crossley, D. A., Jr., "Radioisotope Measurement of Food Consumption by a Leaf Beetle Species, Chrysomela knabi Brown," Ecology 47, 1 (1966).

Eldridge, J. S., and A. A. Brooks, "Gamma-Ray Spectrometry Using Computer Library of Spectra," Nucleonics 24(4), 54 (1966).

Gibbons, J. H., and R. L. Macklin, "$^{51}V(p,n)^{51}Cr$ Cross Section from Threshold to 2.25 MeV," pp. 342–46 in Proceedings Second International Conference on Accelerator Targets Designed for Production of Neutrons (Grenoble, France, June 1965), EUR-2641.d,f,e, 1966.

Gilat, G., and R. M. Nicklow, "Normal Vibrations in Aluminum and Derived Thermodynamic Properties," Phys. Rev. 143, 487 (1966).

Gilliland, R. G., and J. T. Venard, "Elevated Temperature Mechanical Properties of Welds in a Ni-Mo-Cr-Fe Alloy," Welding J. 45, 103s (1966).

[redacted]

Gruber, U., B. Koch, B. P. Maier, O. W. B. Schult, J. B. Ball, K. H. Bhatt, and R. K. Sheline, "Energies and Character of the Low Lying Levels in ^{94}Nb," Nucl. Phys. 67, 443 (1965).

Gurinsky, D. H., J. R. Weeks, C. J. Klamut, L. Rosenblum, and J. H. DeVan, "Corrosion in Liquid Metal Systems," Proc. Intern. Conf. Peaceful Uses At. Energy, 3rd, Geneva, 1964 9, 550 (1965).

Hanson, W. F., and E. T. Arakawa, "Reflectance of Concave Diffraction Gratings for Polarized Vacuum Ultraviolet," J. Opt. Soc. Am. 56, 124 (1966).

Harris, L. A., and H. L. Yakel, "The Crystal Structure of Calcium Beryllate, $Ca_{12}Be_{17}O_{29}$," Acta Cryst. 20, 295 (1966).

Holden, A. N., B. Weidenbaum, and C. F. Leitten, Jr., "Control Rod Materials," Proc. Intern. Conf. Peaceful Uses At. Energy, 3rd, Geneva, 1964 9, 419 (1965).

Jelinek, T. M., R. N. Hamm, E. T. Arakawa, and R. H. Huebner, "Optical Constances of Vacuum Evaporated Films of Cd and Tl in the Vacuum Ultraviolet," J. Opt. Soc. Am. 56, 185 (1966).

Kim, H. J., "The ^{24}Mg$(\alpha,\alpha)^{24}$Mg Reaction and a $J^{\pi} = 10^{+}$ State of ^{28}Si," Phys. Letters 19, 296 (1965).

Kittel, J. H., T. K. Bierlein, B. R. Hayward, and W. C. Thurber, "Irradiation Behavior of Metallic Fuels," Proc. Intern. Conf. Peaceful Uses At. Energy, 3rd, Geneva, 1964 11, 227 (1965).

Lindenbaum, S., "Thermodynamics of Aqueous Solutions of Tetra-n-alkyl Ammonium Halides. Enthalpy and Entropy of Dilution," J. Phys. Chem. 70, 814 (1966).

McNally, J. R., Jr., M. R. Skidmore, P. M. Jenkins, and J. E. Francis, "On the Possibility of Excitation-Heating of Ions to High Temperature," Appl. Opt. 5, 187 (1966).

Melton, C. E., and P. S. Rudolph, "Mass Spectrometric Evidence for H_3O, HD_2O, and D_3O," J. Catalysis 5, 387 (1966).

Minturn, R. E., Sheldon Datz, and R. L. Becker, "Alkali-Atom–Halogen-Molecule Reactions in Molecular Beams; The Spectator Stripping Model," J. Chem. Phys. 44, 1149 (1966).

Nelson, F., "Ion Exchange Procedures. VIII. Separation of Silver from a Number of Elements by Partition Chromatography," J. Chromatog. 20, 378 (1965).

Neufeld, J., W. S. Snyder, J. E. Turner, and Harvel Wright, "Calculation of Radiation Dose from Protons and Neutrons to 400 MeV," Health Phys. 12, 227 (1966).

Payne, J. A., and G. E. Cosgrove, "Tissue Changes Following Cuterebra Infestation in Rodents," Am. Midland Naturalist 75, 205 (1966).

Posey, F. A., and T. Morozumi, "Theory of Potentiostatic and Galvanostatic Charging of the Double Layer in Porous Electrodes," J. Electrochem. Soc. 113, 176 (1966).

[redacted]

Raaen, Helen P., "Instrumentation for Polarography of Glass-Corroding Media," p. 219 in Analysis Instrumentation – 1965, Plenum Press, New York, 1966.

Shults, W. D., "Controlled-Potential Differential dc Polarography," Ph.D. Dissertation, Indiana University, Bloomington, April 4, 1966.

Stanford, J. L., R. N. Hamm, and E. T. Arakawa, "Effect of Photon Refraction on Angular Dependence of Photoelectric Yields in the Wavelength Region 300–1200 Å," J. Opt. Soc. Am. 56, 124 (1966).

Steinhardt, R. G., Jr., W. Neilsen, H. W. Morgan, and P. A. Staats, "Infrared Spectroscopic Study of Condensed Phase Transitions in CF_4," J. Chim. Phys. 63, 176 (1966).

Stoughton, R. W., and R. L. Ferguson, Book Review: Fission Product Yields and Their Mass Distribution, by Yu. A. Zysin, A. A. Lbov, and L. I. Sel'chenkov, authorized translation from the Russian, Consultants Bureau, New York, 1964, 121 pp., Nucl. Sci. Eng. 24, 310 (1966).

Thorngate, J. H., and J. E. Parks, "A Portable Scaler," Health Phys. 12, 385 (1966).

Tobin, J. C., M. S. Wechsler, and A. D. Rossin, "Radiation-Induced Changes in the Properties of Non-Fuel Reactor Materials," Proc. Intern. Conf. Peaceful Uses At. Energy, 3rd, Geneva, 1964 9, 23 (1965).

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