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PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING MAY, 1959

by

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A. ASSISTANCE TO HAPO

F. R. Shober

The thermal conductivities of irradiated uranium and UO_2 are to be measured. Measurements of the electrical resistivity of unirradiated UO_2 have been made from room temperature to 800 C. In the evaluation of the creep properties of annealed and of 15 per cent cold worked Zircaloy-2 at elevated temperature, it appears that cold-worked material results from prolonged exposure under stress at 290 and 345 C. Cyclic temperatures do not greatly alter the creep properties of Zircaloy-2. Work has been completed on the preparation of molybdenum single crystals with the growth of four additional single crystals. Two of these were approximately 3 in. in length.

The density distribution of graphite irradiated at 450 C is being compared with that of unirradiated graphite. Sink-float density measurements are being used to study the density distribution. The simulation of conditions after a loss-of-coolant incident in the PRTR by means of a digital computer has continued. The effect of loss of coolant on the 19-rod plutonium-aluminum element and the UO_2 concentric-ring element is being studied. The end-capping studies on coextruded fuel specimens are being completed. Several different joint designs and joining techniques have been examined and evaluated.

A study of the corrosive action of high-temperature water on defected Zircaloy-2-clad uranium fuel specimens is being made. Time-lapse colored pictures will be taken of the corroding specimens through a windowed autoclave. A successful preliminary run has been made. A related study concerned with development of a detector which will detect a defected fuel element in the NPR prior to catastrophic corrosion of the fuel has been initiated. The detector is to be based on isotropic or elemental halide exchange. A loop for the evaluation of silver halide exchange columns will be designed and constructed. A need has been expressed for a thermal-flux monitoring system in connection with the reactors at HAPO. A study has been initiated at Battelle to develop such a system based on the principle of balancing the temperature of a fission-heated uranium-fueled sensing element and a nearly identical electrically heated balancing element.

Thermal Conductivity of Uranium and UO_2

C. F. Lucks and H. W. Deem

Uranium

The thermal-conductivity apparatus for irradiated specimens has been readied for use. Special thermocouples have been made up. Jigs for specimen cleaning and drilling have been provided.

An unirradiated natural-uranium specimen has been successfully plated. The plating apparatus has been moved to the hot cell.

An attempt will be made to complete thermal-conductivity measurements on three uranium specimens with different burnups.

Uranium Oxide

During May, the thermal-conductivity apparatus was moved to the hot cell. All construction work and fitting have been completed. Due to changes at the hot cell it was found necessary to modify some auxiliary equipment. This has been completed.

Also during May the electrical conductivity was determined on the same unirradiated UO_2 specimens which had been used for thermal-conductivity measurements. This will give some information on the mechanism of heat transfer in UO_2. Tentative values are reported in Table A-1.

During the next month, it is planned to make thermal-conductivity measurements on two high-density UO_2 specimens with different burnups.

Mechanical Properties of Zirconium Alloys

F. R. Shober and J. A. VanEcho

A study of the creep properties of annealed and of 15 per cent cold-worked Zircaloy-2 at 290, 345, and 400 C is in progress. Tests include studies of long-time creep behavior (10,000 to 15,000 hr) and studies of behavior under relatively short-time loading and cyclic-temperature conditions.

Results from tests discontinued after 13,000 hr at 290 and 345 C indicate that the additional strength obtained by cold working is retained for long periods of time at these temperatures. There is no evidence of recrystallization or softening as the result of prolonged exposure under stress at 290 and 345 C. Total creep deformations and minimum creep rates were small, 0.5 per cent and less than 0.00001 per cent per hr, respectively. The effect of cyclic test temperatures, 6 days at temperature (290 or 345 C) and 1 day at room temperature, does not alter the creep properties of Zircaloy-2 greatly. Those specimens loaded to high stress levels showed slightly more total elongation under cyclic temperature conditions than those tested at constant temperature.

Long-time tests (10,000 to 15,000 hr) have been started on annealed material at 290 C.

Preparation of Molybdenum Single Crystals

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

High-purity molybdenum single crystals are being grown in a modified Andradè furnace. A steep temperature gradient along the length of the specimen is obtained by

TABLE A-1. TENTATIVE ELECTRICAL CONDUCTIVITY OF UNIRRADIATED UO_2

Temperature, C	Electrical Conductivity, 10^{-3} ohm^{-1} cm^{-1}				
	Specimen 65, First Run	Specimen 65, Second Run	Specimen 68	Specimen 70	Specimen 1000
		Heating[(a)]			
25	4.3	--	0.12	0.0052	0.016
100	7.7	20.0	0.98	0.068	0.098
150	12.0	26.0	2.8	2.3	2.8
200	16.0	33.0	6.2	6.1	6.3
250	27.0	40.0	12.0	17.0	13.0
300	30.0	49.0	19.0	24.0	21.0
350	34.0	60.0	32.0	43.0	34.0
400	44.0	70.0	44.0	65.0	47.0
450	66.0	84.0	63.0	100.0	65.0
500	120.0	140.0	100.0	140.0	85.0
550	180.0	230.0	160.0	190.0	110.0
600	280.0	280.0	280.0	250.0	130.0
650	380.0	380.0	450.0	300.0	140.0
700	560.0	550.0	680.0	380.0	170.0
750	730.0	730.0	1000.0	460.0	180.0
800	1000.0	1000.0	1500.0	560.0	180.0
		Cooling[(a)]			
800	1100.0	1000.0	1400.0	--	180.0
700	600.0	550.0	780.0	350.0	140.0
600	280.0	280.0	410.0	240.0	110.0
500	120.0	120.0	190.0	160.0	75.0
400	70.0	--	87.0	87.0	52.0
300	50.0	30.0	37.0	40.0	33.0
200	37.0	22.0	13.0	14.0	17.0
100	23.0	13.0	2.5	2.4	5.7
25	13.0	6.8	--	0.31	1.1

(a) Heating and cooling rates were approximately 2 to 3 C per min.

resistance heating the molybdenum rod and water cooling the ends of the rod. Ten single crystals of high-purity molybdenum have been grown in 1/8-in.-diameter rods at a current of 240 amp at approximately 3.0 v for 4 to 6 hr. Recent runs have produced two crystals approximately 3 in. in length.

No further work is planned.

Physical Distortion of Graphite

J. Koretzky, W. C. Riley, and W. H. Duckworth

Research to develop a method of sink-float density measurement to identify factors affecting irradiation-induced volume changes in graphite was continued.

Attempts to improve the reproducibility of the sink-float density distribution of minus 325-mesh TSGBF samples were continued. Repeated separations were made at each liquid density to eliminate entrapment of float material by sink material. Also, a tighter control on the density of solutions was maintained. Through the use of these techniques, satisfactory reproducibility was obtained.

The density distribution of a minus 270-mesh sample of a TSGBF graphite sample irradiated for about 1100 MWD/T at 450 C was measured. The density distribution of an unirradiated sample of minus 270-mesh TSGBF graphite will be measured and the results compared.

Evaluation of Possible Loss-of-Coolant Incidents in the Plutonium Recycle Test Reactor

C. A. Alexander, L. E. Hulbert, A. W. Lemmon, Jr., and R. B. Filbert, Jr.

The simulation of conditions after a loss-of-coolant incident in the PRTR by means of a digital computer was continued this month. In past months attention has been mainly directed to effects of loss of coolant on a fuel rod of a 19-rod cluster element. Attention was directed this month to two other types of elements, the 19-rod plutonium-aluminum element and the UO_2 concentric-ring element which is composed of a center rod and two concentric rings with a gap between them for coolant flow.

Machine computation of Case I (that of a double 14-in. rupture in the outlet pipe) for the concentric-ring element indicated that its thermal behavior after an incident would be closely the same as a 19-rod UO_2 cluster. The plutonium-aluminum fuel element was found to present a somewhat more severe hazard due to its low melting point. Comparison of a plutonium-aluminum cluster element with a UO_2 cluster element under conditions in which no coolant would be added indicated that plutonium-aluminum element would reach its melting point in 262 sec as compared to nearly

1000 sec before the Zircaloy cladding of the UO_2 element reached the melting point. However, for this case, cooling water covers the element in about 106 sec so that no critical condition would arise.

An analysis of Cases III and IV, that of a 1-3/4-in. jumper break at the top (III) and bottom (IV) indicated that neither the victim tube (the tube whose jumper had ruptured) nor the intact tubes would be a hazard until after the fuel elements would be uncovered. Long delays in recovering or providing cooling water to the fuel elements, however, which might occur in Case IV, might prove hazardous, particularly for the plutonium-aluminum elements. Preliminary analysis indicated that delays could be minimized if the motorized valve at the reactor primary-loop outlet could be closed after blowdown. It is now considered that this will be made standard operating procedure.

In addition to the machine computations, an analysis of heat transfer by radiation was made to determine if the aluminum shroud tube could receive enough heat by radiation to cause melting. The analysis was made for the adiabatic phase in which it was considered that no safety injection water would be added. Results of this computation indicated that the aluminum shroud tube would melt at 710 sec, at which time the Zircaloy process tube would be at 2000 F, the 6-rod elements at 2460 F, and the 12-rod elements at 2290 F. From the above analysis the 12-rod elements are at a lower temperature than the 6-rod elements due to radiation to the process-tube wall, even though the decay heat-generation rate is greater in the 12-rod elements.

During the coming month, it is planned to complete the injection phase of Cases III and IV and to prepare a report covering this study.

End-Capping Methods for Coextruded Fuel Pins

R. M. Evans, J. J. Vagi, and R. L. Koppenhofer

This is a survey project on methods for end capping Zircaloy-2-clad-uranium core fuel pins. The literature survey was completed and materials for qualitative tests of joining methods were prepared early in May.

End-cap welding experiments included a combination spot-seam weld and arc-seam weld. Arc sintering of powdered zirconium was tried qualitatively on stainless steel simulated fuel rods. Several induction-brazing experiments also were conducted. All of these experiments appear worthwhile for further study.

All laboratory work was concluded at the end of May. A report is under preparation.

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A Photographic Study of the Corrosion of Defected Fuel Elements in High-Temperature Water

E. F. Stephan and F. W. Fink

The corrosive action of high-temperature water on defected Zircaloy-2-clad uranium-core fuel elements is to be photographed. Time-lapse colored motion pictures are to be taken of the corroding specimens through a windowed autoclave.

The equipment has been calibrated and a successful preliminary run was made. The specimen was natural uranium with a 30-mil Zircaloy-2 cladding. A defect, 0.030 in. in diameter, was drilled through the cladding to the core. In 625 F water, the hole began to enlarge in 12 min. The cladding surrounding the defect ruptured in 28 min with a rapid evolution of hydrogen. The cladding opposite the defect ruptured in 48 min. In 68 min, enough UO_2 had escaped from the cladding to cover the windows of the autoclave making further visual observation impossible. Pressure records showed that the reaction was virtually complete in 100 min.

The defected specimens which are to be examined in this program have been received and work will be started immediately.

Development of a Fuel-Element Leak Detector

J. E. Howes, T. S. Elleman, and M. Pobereskin

Battelle has undertaken the development of a fuel-element leak detector based on isotopic or elemental halide exchange for the Hanford high-pressure water cooled dual-purpose reactor.

Evaluation of the performance requirements necessary for satisfactory operation of a silver halide leak detector has been initiated. A preliminary experimental program has been designed to measure the fission-product halide retention and decontamination efficiencies of silver halide columns under various conditions of operation.

A loop for the evaluation of silver halide exchange columns will be designed and constructed. The effect of temperature, flow rate, halide surface, and other system parameters on the physical characteristics of the exchange material and on the retention and decontamination of fission-product halide will be investigated.

Thermal-Neutron-Flux Monitoring System

C. V. Weaver, J. W. Lennon, and J. F. Quirk

The ultimate objective of this program is to develop a thermal-neutron-flux monitoring system for the Hanford reactors. A flux-measuring instrument has been developed at Battelle which appears suited as the basic unit for the proposed system. It is based on the principle of balancing the temperatures of a fission-heated uranium-fueled sensing element and a nearly identical electrically heated balancing element. A description of the development of the prototype instrument is contained in BMI-1083.

Work has begun on developing ceramic elements for use in the flux sensor. The two elements in each assembly are to be alike in chemical composition, but the fission-heated element will contain enriched uranium while the balancing element will contain natural or depleted uranium. Compositions to be investigated first will be in the $MoSi_2$-Al_2O_3-UO_2 system because of encouraging results obtained with these ceramics in earlier work.

One of the problems encountered in the previous work was obtaining the required degree of reproducibility and stability in the resistivity of the balancing element. The first phase of the current research concerns efforts directed to resolving this difficulty. Variables in materials and processes are being studied.

Thus far, raw materials have been obtained and are being analyzed, a purification treatment for $MoSi_2$ is being evaluated, dies for fabricating specimens are being made, and other processing equipment (mixing, furnaces for atmosphere firing, etc.) is being readied. A number of resistor specimens have been formed preparatory to sintering.

Work next month will include further characterization of raw materials and an effort to assess the effects of composition and processing conditions on the electrical resistance of sintered compacts.

It is proposed to prolong the service life of the flux sensor by surrounding the sensing (and balancing) elements with a flux absorber to reduce the rate of fuel burnup. In a preliminary analysis of the problem, it appears that silver will be a suitable shim material.

Work has also begun to develop the mechanical design of the flux sensor. Bench testing of components and the over-all design will begin as soon as suitable ceramic elements become available.

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

Preparation of Aluminum-Uranium Alloys

N. E. Daniel, E. L. Foster,
and R. F. Dickerson

The first dispersion-type fuels proved feasible for use in nuclear reactors were those of the aluminum-uranium system. The alloys of this sytem are used extensively in production and research-type reactors operating at low temperatures utilizing water cooling. Continued interest in these alloys is reflected in the present efforts to develop more satisfactory fuels and improved fuel-element production techniques. Previous studies have shown that a small quantity of a third element added to the binary aluminum-uranium alloys improves their fabricability and makes them more amenable to coextrusion with aluminum. The effects of selected additions on the castability and fabricability of the binary aluminum-35 w/o uranium alloy have been investigated. Since the alloys are desired in the form of hollow cylindrical extrusion billets suitable for coextrusion with aluminum into tubular fuel elements, centrifugal casting has been investigated to determine the feasibility of producing sound extrusion billets with acceptable end-to-end homogeneity with a minimum of scrap production.

The evaluation of six centrifugally cast 26-in.-long aluminum-35 w/o uranium extrusion billets has been completed. Chemical analyses samples taken by drilling through the walls of the hollow billet, along the length of the castings, revealed a variation of uranium content from 32.1 to 37.3 w/o in three castings. The best of these castings exhibited a low of 32.1 w/o and a high of 35.3 w/o uranium. Chemical analyses of material removed from the inner surface of the castings to a depth of approximately 0.062 in. showed that this surface was depleted in uranium, containing from 14.5 to 16.8 w/o uranium.

Tensile data have been obtained on binary and ternary aluminum-35 w/o uranium alloys in both the as-cast and as-extruded condition. The extruded specimens were obtained from 3-in.-diameter ingots which were extruded at 800 F through a shear-face die to form 3/4-in.-diameter rod. The alloys of interest were the binary alloy and the ternary alloys containing 3 w/o silicon, 3 w/o tin, or 3 w/o zirconium. The data are given in Table B-1.

TABLE B-1. TENSILE DATA FOR ALUMINUM-35 w/o URANIUM ALLOYS WITH 3 w/o TERNARY ADDITIONS

Ternary Addition to Aluminum-35 w/o Uranium Base Alloy, w/o	Ultimate Strength, psi		Reduction in Area, per cent		Maximum Extrusion Pressure, tons per in.^{2}
	As Cast	Extruded	As Cast	Extruded	
None	17,000	21,500	2.35	1.85	12.2
3 tin	19,000	17,900	4.34	3.5	9.37
3 zirconium	19,600	17,000	3.75	3.62	9.37
3 silicon	12,500	22,000	3.5	3.75	9.37

An examination of Table B-1 reveals that in the as-cast condition the alloys containing tin and zirconium exhibited greater ultimate tensile strengths than did either the binary alloy or the alloy containing silicon; whereas, in the extruded condition the opposite was true. The latter two exhibit increases in their tensile strengths after fabricating. An explanation of the large increase in tensile strength shown by the alloy containing silicon after extrusion must lie in casting quality since the as-cast strength was abnormally low when compared with the other alloys. The reductions in area of the alloys containing the ternary additions were greater than the reductions in area of the binary alloy in both the as-cast and the as-fabricated conditions and are attributed to the reduction in the amount of compound in the ternary alloys. In reference to the extrusion pressures, these are the values obtained from one series of alloys in effecting a 16 to 1 reduction of a 3-in.-diameter ingot at a ram speed of 20-in. per min.

C. RADIOISOTOPE AND RADIATION APPLICATIONS

P. Schall

In the program on the development of radiotracer techniques for cement quality control a study was made of the effect of foreign elements on the radiometric titration of magnesium. Only calcium appears to be present in sufficient concentration in typical cement samples to cause interference.

Experimental work has been started to determine the feasibility of intrinsic tracer control in one unit process in the ammonia-leach nickel-refining process.

Chemical analyses of irradiated inclusion compounds are continuing. It is expected that the analyses will be completed during the next month and summarized in the next report.

The investigation of the susceptibility to radiation-induced grafting of a series of polymethacrylic esters is continuing. It has been found that peroxy-site production is about the same for polyoctyl methacrylate as for polymethyl methacrylate. This is in contrast to the large difference in radical-site production for these two polymers.

Equipment construction for the study of radiation-induced nitration is in progress. Several preliminary irradiations under ambient conditions have been completed and analytical techniques are being established.

Development of Radioactive-Tracer Quality-Control Systems

T. S. Elleman, C. T. Brown, D. N. Sunderman,
and M. Pobereskin

The effect of interfering elements on the radiometric method for magnesium analysis has been investigated. Samples containing 10 mg of MgO and 10 mg of a specific contaminant in 100 ml of solution were run under the normal experimental conditions. The following contaminants were used: $Al_2(SO_4)_3$, $CaCl_2$, $Fe(NO_3)_3$, KCl, $MnSO_4$, and NaCl. Calcium and manganese were the only elements that interfered. Calcium yielded results 17 per cent high, and manganese yielded results 14 per cent high. One-milligram quantities of $MnSO_4$ and $CaCl_2$ per 10 mg MgO in 100 ml of solution did not interfere.

In a typical cement sample only 0.025 mg of manganese will be present for each 1 mg of magnesium. Therefore, interference due to manganese is no problem. However, calcium is present in a ratio of 26.4 mg to 1 mg of magnesium. In this later case a separation must be effected before the magnesium is determined.

A re-evaluation of the neutron-activation analysis technique was made using various standard samples. Corrections for the geometry of the detector on the hundred-channel analyzer were good, but dead-time corrections were not. In order to obtain good quantitative results it is necessary to run the samples at a low and fairly consistent dead time.

During the next report period work will continue on the activation analysis of cement and raw materials, and work will be concluded on the calcium and magnesium analyses.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, D. N. Sunderman,
and M. Pobereskin

During May, preliminary experimental study began on methods of radiotracer control of the copper-stripping operation in the ammonia-leach nickel-refining process. The purpose of these experiments is to determine the limits of sensitivity and accuracy obtainable by the various radioactivity-measurement techniques which might be used. This experimental study is expected to pinpoint the technique most suitable for this application and to define the limits of accuracy of this technique under actual operating conditions.

A sample of Matheson electrolytic copper was irradiated in the Battelle Research Reactor to determine its radiochemical purity. Following the neutron irradiation, the induced radioactivity was followed for several days. After 130 hr, corresponding to ten copper-64 half-lives, the activity was still essentially pure copper-64. It was concluded that this electrolytic copper will provide a suitable radiotracer for this work.

A commercial producer has been asked to cooperate in ascertaining the usefulness of the proposed application under actual plant conditions. Plans for proceeding with the study of radiotracer control of the copper-strip operation, beyond the preliminary phase discussed above, are contingent upon the producer's favorable reaction.

Next month, experimental tracer work on the copper-strip process will be continued, and it is expected that some definite arrangements with the producer will be discussed. If time permits, experimental evaluation of a different radiotracer process-control application will begin.

Radiation Chemistry of Inclusion Compounds

M. J. Oestmann, M. J. Landon, E. J. Kahler,
and W. S. Diethorn

Chemical analyses of irradiated urea-dodecane and the nickel cyanide-benzene complex are continuing. There is nothing new to report this month. Results will be reported as soon as they become available.

C-3

Graft-Polymerization Studies

I. S. Ungar, J. F. Kircher, R. A. Markle,
and R. I. Leininger

Work has continued on the free-radical and peroxy grafting of polymethylmethacrylate and polyoctylmethacrylate. As discussed in the last report, polyoctylmethacrylate is much more susceptible to radical-site production than is polymethylmethacrylate. Under comparable irradiation conditions about six times as many sites are formed in polyoctylmethacrylate as in polymethylmethacrylate. This large difference was not found in the irradiations in oxygen to form peroxy sites. Here, site production was about the same in both cases. No reason can be advanced at this time for this difference in behavior.

The length of the vinylpyrrolidone chains grafted onto the polymers has been measured. These measurements are based on the number of sites produced and the nitrogen content of the fractionated polymer. In the case of postirradiation grafting, polyoctylmethacrylate and polymethylmethacrylate were found, respectively, to have side chains of 14 and 66 vinylpyrrolidone units. This difference in the length of the side chains on the two polymers may reflect the effect of the number of sites produced in each polymer. This would be analogous to the effect of catalyst concentration on molecular weight in vinyl polymerizations.

The side chains grafted onto peroxy sites on polymethylmethacrylate were much longer than those obtained by free-radical grafting. Again this may be a consequence of the relatively few sites formed. The analyses for the comparable case with polyoctylmethacrylate are not complete.

Further studies of the influence of the concentration of vinylpyrrolidone on the side-chain length have been made. Qualitative observations indicate that, as expected, lower concentrations lead to the formation of shorter chain lengths.

Polybutyl-, polylauroyl-, and polystearoylmethacrylate have been prepared and it is planned to prepare polysecondary butyl- and polytertiary butylmethacrylates. These will complete a comprehensive series of polymethacrylic esters that will show the effects of varying ester length and structure.

Nitration of Hydrocarbons

M. J. Oestmann, J. F. Kircher, R. E. Fulmer,
and G. A. Lutz

This is the second monthly report on the project "The Effect of Phase on the Radiation-Induced Nitration of Hydrocarbons".

During the month of May the design of equipment meeting the temperature and pressure requirements of the irradiation program was completed. Construction of the equipment is about 50 per cent complete.

C-4

Several preliminary irradiation runs were made with nitric acid-cyclohexane mixtures in glass containers. The irradiated mixtures are being used to develop analytical techniques for the separation and identification of radiolysis products.

Completion of all equipment is expected in June. Thermal runs in the absence of radiation will be made in order to define the conditions which do not produce rapid thermally induced nitration reactions.

D. PROCESSING OF FEED MATERIALS

E. L. Foster

National Lead of Ohio (NLO) as the operator of the Feed Materials Production Center is concerned with the quality of uranium fuel slugs. One process step that contributes to the qualities of the fuel slugs is the melting and casting operation, and NLO is seeking a convenient method of evaluating the factors present in the solidification of uranium ingots. Because direct experiments would be costly and cumbersome, a program of study to evaluate these factors analytically is in progress. It is anticipated that if such a method is proved feasible it would be applicable to present and future casting questions.

Solidification of Uranium

E. L. Foster, C. K. Franklin, B. L. Fletcher,
A. B. Pritsker, and R. F. Dickerson

The manner in which uranium solidifies in cylindrical graphite molds is being studied. A mathematical model, incorporating the characteristics of the freezing metal, has been developed to predict the transfer of heat from the ingot to the mold during cooling. In order to calculate the theoretical heat flow in the system, the various thermal properties of the mold and ingot were determined experimentally. Casting experiments designed (1) to measure the temperature of the ingot, mold, and furnace surroundings from the time of pouring and (2) to investigate the physical events occurring at the interface of the mold and ingot were conducted. The data obtained from these experiments are to be used to refine and check the theoretical model.

Several melts, producing castings 3 in. in diameter by 20 in. long, were poured. These ingots were found to solidify within 1 min after pouring. In addition to obtaining temperature measurements, the nature of the interface between the mold and ingot was investigated. Results of interface determinations indicated a gap formation occurring shortly after pour.

Currently, efforts have been concentrated on recording the separation phenomena (gap formation) and temperatures in large masses of uranium. To do this, melts, which produced ingots 7 in. in diameter by 48 in. long, were cast at the Feed Materials Production Center at Fernald, Ohio. Employing plant melting and casting procedures, two series of three ingots were poured. Ingots in the first series were cast to measure time and position of separation between the mold and the ingot as freezing progresses. Using electrical probes as the detecting devices, gap formations were recorded. Early gap formation was seen at the top portion of the ingot while the bottom section generally remained in contact with the probes during the entire time of the tests. The gap formations were seen to vary greatly from pour to pour with an average time of 2-1/2 min for total separation to occur in the top two-thirds of the mold. A mold fill time of about 35 sec was also observed in the tests. However, at the start of pour a number of probes at the top and bottom of the mold reacted in an erratic fashion. These probes indicated contact with the metal before the liquid level had reached their level. This action is believed to be the result of splatter and fan-out of the pour stream as the liquid filled the mold.

The ingots in the second series were cast with groups of thermocouples placed in or on the mold to measure temperatures of the mold and ingot. To obtain metal temperatures platinum-platinum 10 w/o rhodium thermocouples were cantilevered to various distances into the mold cavity. The thermocouples extended 0.00, 0.50, 1.50, 2.50, and 3.50 in. from the inner mold-wall surface at locations 10, 20, 30, and 40 in. from the bottom of the mold cavity. Except for the hot junctions, which were directly exposed to the liquid metal, the thermocouples were protected from chemical attack by graphite cylinders. Along the same planes as the platinum-platinum 10 w/o rhodium couples, Chromel-Alumel thermocouples were placed in the mold to read mold temperatures. In addition, a thermocouple was attached to the furnace wall and one was located above the mold to measure environment temperatures. Temperature readings were recorded for a period of 15 min, starting at the pour time.

Results of the temperature measurement indicate a total freezing time from 5 to 7 min, depending on the pouring temperatures. Pour temperatures ranging from 2450 F to 2520 F were recorded in the tests.

An initial computer run has been made to check out the mathematical model as rescaled for correlation with the above-mentioned plant experiments. On the basis of preliminary indications from the experimental data, it was decided that the casting wall thickness at the time separation occurred was in the neighborhood of 0.63 in. and this value has been adopted for the first calculations.

The first computer run was made to check the operation of the rescaled program and to examine the stability and convergence of the solution using various time increments. It was found that a time interval of 5 sec between successive temperature calculations gave nonoscillatory temperature values after the pour was completed. During the pour, a 1-sec time interval was used, and it was found that the interface behavior was erratic for a short period during which the wall attempted to remelt due to the superheat remaining in the liquid metal. This condition can be partially rectified by using shorter time increments during the pour and subsequent solidification of the casting-wall cells.

The first over-all solidification time calculated was 14.3 min, while a preliminary analysis of the experimental data indicates that the experimental value is in the range of 5 to 7 min.

E. SPRAY DEPOSITION OF CALCIUM METAL ON NICKEL OR INCONEL

R. M. Evans

This is the fifth monthly report on a feasibility study to determine if calcium coatings can be successfully put on nickel by arc-spraying methods.

In a review of the progress of the work so far, it was decided to continue experimentation to establish the following:

(1) The causes of the black/white vapor in spray chamber

(2) The kind of spraying that will give the best results and least vapor

(3) The effect of the presence of vapor in the chamber on coating adherence

(4) The effect of changes in material preparation and operating conditions to minimize the black/white vapor.

Answers to these questions should lead to better conclusions about the feasibility of the process. Experimental work has been resumed.

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

The transformation and volatility of UO_2 during oxidation can be prevented by adding 60 mole per cent of an additive such as La_2O_3 or Y_2O_3. In an attempt to reduce the amount of additive current work has been directed toward the partial substitution of a divalent additive such as CaO for La_2O_3 or Y_2O_3 as the case may be. Results to date indicate that 20 to 25 mole per cent of CaO is needed to accomplish stabilization. Additional work is being done to verify these initial results. Although a die of satisfactory design has been constructed for the high-pressure high-temperature reaction studies, some difficulties have been encountered in obtaining accurate temperature measurements. Effort is being directed toward improving the heater gradients as well as obtaining more reliable thermocouples.

Postirradiation examination of two capsules containing zirconium-2 w/o uranium alloy hydride has been initiated. The calculated burnups of these specimens was about 20 a/o of the uranium. The initial examination of these specimens by stereomacroscope was encouraging in that there appeared to be no visible damage. Analyses of the dosimeter wires is in progress and measurements of physical dimensions, density determinations, and metallographic examination will be completed during the next report period.

The cold irradiations of Type 347 stainless steel in a high-flux position at the ETR is continuing, although the reactor did not operate at full power during May. It is estimated that the sum of the partial-power runs was 4.5 days of full-power operation. The second ETR core-filler piece has been shipped to the ETR for the "gamma-heated" capsule experiments and three additional nonlead capsules are being assembled.

The niobium-base alloys being studied as possible alternate cladding material for future EBR cores have melted and hot rolled. Metallographic examination and chemical analyses have also been completed and the data are being accumulated. The material will be cold rolled, and tensile studies will be initiated as soon as possible. Corrosion testing of commercial-purity niobium-base alloys in 680 F water has been continued. The binary alloy containing 45 a/o zirconium and the ternary containing 28 a/o titanium and 5 a/o chromium continue to exhibit tightly adherent tarnish films after 168 days of exposure to 680 F water. A ternary alloy containing 11.2 a/o titanium and 3.2 a/o molybdenum cold rolled to an 18 per cent reduction has been tested in creep at 1200 F under a stress of 30,000 psi. After an initial (elastic) on-load deformation of 0.17 per cent, this sample showed substantially no deformation in 138 hr.

Friction studies of molybdenum sliding against itself, dry and sodium-lubricated, are continuing in an attempt to better understand the phenomena involved in operating devices required for systems containing alkali metals. To date results indicate higher friction between surfaces coated with MoO_3 than that between surfaces coated with MoO_2. Sodium lubrication reduces the friction still further.

Valence Effects of Oxide Additions to Uranium Dioxide

W. B. Wilson, A. F. Gerds, and C. M. Schwartz

An investigation is being made of the effect of oxide additions to uranium oxide in preventing the transformation and volatility associated with the formation of U_3O_8 during oxidation. Previous research has shown that this may be accomplished with UO_2 containing 60 mole per cent of an additive such as La_2O_3. Current work has been directed toward reduction of the large amount of additive required for stabilization through partial substitution for La_2O_3 of a divalent additive such as CaO.

Improved methods of chemical analysis to distinguish between La_2O_3 and CaO have disclosed that in excess of 50 per cent of the CaO additive was lost from the materials during preparation and sintering of the solid solutions. This heavy loss of CaO would possibly result in materials which were inferior from the standpoint of stabilization, since sufficient additive may not have been present.

In contrast to the materials from which the CaO losses occurred, an earlier sample of UO_2-20 mole per cent La_2O_3-20 mole per cent CaO prepared by a different fabrication procedure exhibited superior oxidation characteristics. The possibility that the different fabrication technique resulted in less CaO loss from the solid solution during preparation suggests that the earlier fabrication method should be used for the next series of preparations.

A final series of samples is being prepared, using the older technique of preparing the binary solid solutions of UO_2-La_2O_3 and UO_2-CaO first. The binaries are then reacted to form the ternary UO_2-La_2O_3-CaO composition. The compositions which appear to be most suitable based on previous data are (in mole per cent):

(1) UO_2-20 La_2O_3-20 CaO

(2) UO_2-25 La_2O_3-25 CaO

(3) UO_2-20 Y_2O_3-20 CaO

(4) UO_2-25 Y_2O_3-25 CaO .

These materials are being prepared and will be evaluated in air oxidation at 1750 C. Chemical and structural analyses will be obtained before and after oxidation testing and volatility will be evaluated by weight-change analysis.

F-3

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

The purpose of this project is to investigate the effects of combined high pressure and high temperature on solids. Work was continued to improve the measurement and control of temperature in the high-pressure die, concurrent with an investigation of the effects of pressure and temperature on the uranium-oxygen system (U_3O_8). The major difficulties encountered with temperature measurement have been due to gradients in the heater and to shorting and disruption of thermocouples introduced into the high-pressure system.

Considerable improvement has been obtained in the gradient problem by modifying the sample chamber to incorporate a sleeve of high-thermal-conductivity material (e. g. , BeO) around the heater. A major problem, however, is still present in obtaining reliable thermocouples. Work will be directed toward improved temperature measurement and studies of the uranium-oxygen system under pressure will continue.

Fueled Zirconium Hydride Moderator

G. E. Lamale, A. K. Hopkins, and H. H. Krause

Postirradiation studies are in progress on the specimens of hydrided zirconium-2 w/o uranium which were irradiated in the MTR. All other experimental work on the project has been completed.

Structure and Pressure-Composition-Temperature Studies

The hydrogen-absorption isotherms and the high-temperature X-ray diffraction studies have been completed.

Radiation-Damage Studies

The postirradiation examination of the two capsules, BMI-20-1 and BMI-20-2, containing specimens of hydrided zirconium-2 w/o uranium, has begun at the Battelle Hot Cell Facility. Both capsules were irradiated at the MTR to burnups of approximately 20 a/o of the uranium.

Both capsules have been opened and all specimens removed. Examination with the stereomacrograph showed that the specimens did not suffer any visible radiation damage. Slight discoloration of the surface of some of the samples, which appears to be oxide, was noted, although a hydrogen atmosphere was maintained in the capsules during irradiation. Photographs of the samples were taken.

Dosimeter wires, which were located alongside the specimens and close to their surface, have been cut in lengths which correspond closely to the temperature-monitored specimens. Consequently, accurate dosimetry will be possible. Measurements of physical dimensions and densities, as well as metallographic and burnup studies, are planned for the coming month to complete the postirradiation study and evaluation.

Irradiation-Surveillance Program on Type 347 Stainless Steel

F. R. Shober, J. E. Howes, Jr., and J. F. Lagedrost

An irradiation-surveillance program is being conducted on AISI Type 347 stainless steel in support of the KAPL C-33 loop in the ETR. Changes in the mechanical properties as affected by exposure to a fast-neutron flux (neutrons having energies greater than 1 Mev) at 120 and 600 F are being investigated. The tensile, cyclic strain fatigue, and impact properties are to be measured after each period of exposure representing 6 months of service in the F-10 position. Data are available on the mechanical properties of Type 347 stainless after exposure to 3.74×10^{21} n per cm^2 and represent the starting point for this investigation. It is expected that the total fast-neutron exposure will be approximately 1.2 to 1.6×10^{22} n per cm^2.

Eight "cold" capsules are being irradiated in the K-8 and the L-8 positions and have been placed to take advantage of the higher flux of each position. The insertion of fuel into the L-7 and M-7 positions has increased the flux density by approximately 40 per cent in the vicinity of the capsules. The ETR did not operate at full power during May, and very little exposure has been accumulated. The sum of the partial-power runs for May is equivalent to 4.5 days at full power. The total nvt accumulated to May 20, 1959, for each capsule is shown in Table F-1 and has been calculated on the basis of 4.5 days at full power.

TABLE F-1. CUMULATIVE FAST FLUX ON "COLD" CAPSULES OF THE TYPE 347 STAINLESS STEEL IRRADIATION-DAMAGE PROGRAM

Capsule	Capsule Type	Approximate Removal Date[(a)]	Approximate Exposure at Time of Removal[(b)], 10^{22} nvt	Total Exposure as of May 20, 1959, 10^{21} nvt	
				Top	Bottom
BMI-24-2	Tensile and fatigue	Jan., 1962	1.31	1.920	2.728
BMI-24-4	Tensile and fatigue	Jan., 1963	1.78	0.969	2.074
BMI-24-6	Tensile and fatigue	June, 1961	1.08	2.806	2.101
BMI-24-8	Tensile and fatigue	June, 1962	1.54	1.105	2.125
BMI-24-10	Tensile and fatigue	Jan., 1961	0.84	2.188	2.599
BMI-24-12	Tensile and fatigue	June, 1960	0.61	2.971	2.291
BMI-24-14	Impact	June, 1962	1.54	2.688	2.752
BMI-24-16	Impact	June, 1960	0.61	2.725	2.651

(a) Based on 6-month lead on loop, plus 2 months for tests.

(b) Based on maximum fast flux at tube of 1.7×10^{14} nv for 8-month periods, 6-month lead time and 2-month time for examination.

Nickel dosimeters have been placed in the four faces and the center of the core-filler piece at the beginning of Cycle 15 and will remain there until a burnup of approximately 5000 megawatt-days has been accumulated. This should be equivalent to approximately 2 cycles at full power (175 megawatts). The dosimeter in the center hole will be analyzed at the ETR, and the others will be returned to Battelle for analysis.

A second four-hole core-filler piece has been fabricated at Battelle and shipped to the ETR. It is planned to use this core-filler piece in the "hot" capsule experiment. Thermocouple-lead Capsule BMI-24-19 is to be used in an attempt to correlate temperature during irradiation with a known gamma flux. It is expected that the temperature of the "hot" nonlead capsules during irradiation can be estimated if the gamma flux is known. Insertion of the lead capsule will be made at such time as it is assured that a reasonable total exposure time can be guaranteed before the capsule need be discharged.

Three nonlead capsules containing tensile and cyclic-strain-fatigue specimens are being assembled. The specimens from these capsules will be given a postirradiation anneal at 600 F after an exposure time comparable to specimens tested in the as-irradiated condition at process-water temperature. A comparison of data from these specimens should give some indication of the effect irradiation at elevated temperatures on their mechanical properties.

Development of Niobium-Base Alloys

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

An evaluation of several niobium-base alloys for possible application as alternate cladding material in future EBR cores is being made. The alloys being studied are niobium-1 w/o chromium, niobium-2 w/o chromium, niobium-4.5 w/o zirconium, niobium-10 w/o tantalum-2 w/o chromium, niobium-20 w/o titanium-1.5 w/o chromium, niobium-40 w/o titanium-10 w/o aluminum, unalloyed niobium and a comparison alloy of vanadium-10 w/o titanium-1 w/o niobium (an alloy currently being considered as an alternate EBR cladding material).

Melting, evaluations of hot and cold fabricability, metallographic examinations, chemical analyses, and a short heat-treatment study have been finished.

Cold rolling of the hot-forged slabs resulted in edge cracking; therefore, the slabs have been encapsulated in stainless steel cans and evacuated, and will be annealed at 1050 C for 1/2 hr. The cracked material will then be removed and cold rolling will be continued.

Tensile specimens of each alloy will be prepared for tests at 650 and 800 C. The thermal conductivity will be determined on the three alloys having the highest tensile strength at 800 C. In the meantime, as soon as sheet material has been obtained, sheet-to-sheet weldability of the cold-fabricable alloys will be studied.

F-6

Development of Corrosion-Resistant Niobium Alloys

D. J. Maykuth, W. D. Klopp, R. I. Jaffee, W. E. Berry,
and F. W. Fink

The development and evaluation of niobium-base alloys for possible service in pressurized-water reactors is being continued. Mechanical properties and corrosion data are being accumulated on high-purity base alloys which were chosen on the basis of hot-water-corrosion screening tests conducted on commercial-purity niobium-base alloys.

Corrosion testing of the commercial-purity niobium-base alloys in 680 F water has been continued. Total weight changes after 168 days of exposure are presented in Table F-2. The 45 a/o zirconium binary and the 28 a/o titanium-6 a/o chromium ternary continue to exhibit tightly adherent tarnish films. All other alloys are covered with fairly adherent gray films except the tungsten and low-molybdenum alloys, which are losing weight.

Results after 14 days of exposure in 750 F 1500-psi steam are also presented in Table F-2 for selected duplicate specimens from the commercial-base alloys. To date all these specimens exhibit adherent films.

Corrosion results for consumable-electrode-melted high-purity base alloys are summarized in Table F-3. After exposure times ranging up to 84 days in 600 and 680 F water and 750 F steam, all unalloyed niobium samples are either beginning to flake and lose weight or have completely oxidized. The 7.18 a/o molybdenum alloy failed by cracking after 70 days of exposure in 750 F steam, while all other specimens in all test conditions continue to exhibit adherent films. The reaction rates for the 12.58 a/o vanadium alloy continue to follow the cubic rate law, while those of all the other alloys more nearly approach a parabolic rate.

The evaluation of the corrosion behavior of melts from several 50-g ingots has been started. Weight changes after 14 days of exposure in 680 F water are presented in Table F-4. The corrosion product on the 1 a/o zirconium alloy is flaking and the alloy is losing weight. All other specimens possess adherent films, but exhibit fairly high weight gains which approach 1 mg per cm^2 at alloy contents below 5 a/o.

A sample of the ternary niobium-11.2 a/o titanium-3.2 a/o molybdenum alloy was cold rolled to an 18 per cent reduction and tested in creep at 1200 F under a stress of 30,000 psi, a stress level 35 per cent in excess of the 1200 F yield strength of this alloy. After an initial (elastic) on-load deformation of 0.17 per cent, this sample showed substantially no creep deformation in a 138-hr exposure period. These results confirm earlier creep data which indicated that cold working of this material, and probably of other niobium-base alloys, would substantially improve the 1200 F creep resistance.

Melting of the thirteen 50-g niobium-base alloys listed in Table F-5 was finished to complete the preparation of all of the new screening-alloy compositions. Excepting Ingots 47, 52, and 55, all alloys were fabricated by alternate processes of cold rolling and vacuum annealing. During the initial cold rolling, the 5 a/o iron alloy cracked severely. Consequently, this ingot was remelted and scheduled for hot rolling in

TABLE F-2. CORROSION DATA FOR COMMERCIAL-PURITY NIOBIUM-BASE ALLOYS

Alloy Content (Balance Niobium), a/o	Cross Section[(a)], barns per atom	Total Weight Change[(b)], mg per cm^2	
		After 168 Days in 680 F Water	After 14 Days in 750 F 1500-PSI Steam
Unalloyed Nb	1.15	(c)	--
10.5 Zr	1.05	0.63	0.15
26.1 Zr	0.91	0.18	--
35.7 Zr	0.81	0.70	--
45.7 Zr	0.72	0.70	--
1.08 W	1.2	-2.07	0.66
4.67 W	2.0	-24.2	--
9.56 W	2.9	(d)	--
2.45 Mo	1.18	-3.64	0.35
5.20 Mo	1.21	-0.33	0.30
7.40 Mo	1.25	0.65	--
4.42 V	1.31	0.39	0.35
6.59 V	1.41	0.70	0.25
8.93 V	1.50	0.57	0.25
10.7 V	1.62	0.83	--
13.7 V	1.80	0.48	--
24.2 V	2.10	0.04	--
4.90 Fe	1.2	0.14	0.73
9.41 Ti	1.6	0.64	0.21
18.8 Ti	2.1	0.43	--
24.3 Ti	2.3	0.52	--
30.5 Ti	2.6	0.40	--
33.8 Ti	2.8	0.55	--
12.0 Ti-0.5 Cr	1.54	0.61	--
20.2 Ti-2.1 Cr	2.11	0.32	--
28.2 Ti-6.1 Cr	2.44	0.24	--
12.0 Ti-4.2 Mo	1.69	0.66	--
17.4 Ti-6.2 Mo	2.00	0.47	--
23.1 Ti-7.8 Mo	2.24	0.49	--
10.4 Ti-5.0 V	1.72	0.25	--
16.1 Ti-8.4 V	2.14	0.40	--
22.6 Ti-11.0 V	2.67	0.52	--

(a) Estimated graphically.

(b) A minus sign indicates sample has lost weight.

(c) Completely oxidized after 42 days of exposure.

(d) Specimen cracked after 14 days of exposure.

TABLE F-3. CORROSION DATA FOR ALLOYS PREPARED FROM HIGH-PURITY NIOBIUM BY CONSUMABLE-ELECTRODE ARC MELTING

Alloy Content (Balance Niobium), a/o	Total Weight Change, mg per cm^2		
	In Water		In Steam at
	600 F	680 F	750 F
	84 Days of Exposure		
Unalloyed, commercial Nb	0.66	(a)	(b)
Unalloyed, high-purity Nb	0.53	0.97	-12.0
7.18 Mo	0.23	0.62	(c)
12.58 V	0.12	0.32	0.62
	56 Days of Exposure		
46.8 Zr-5.06 Ti	0.12	0.63	1.24
18.8 Ti-8.7 Mo	0.16	0.26	0.42
11.2 Ti-3.2 Mo	0.24	0.29	0.61

(a) Completely oxidized after 56 days of exposure.
(b) Completely oxidized after 28 days of exposure.
(c) Cracked after 70 days of exposure.

TABLE F-4. CORROSION DATA AFTER 14 DAYS OF EXPOSURE IN 680 F WATER FOR ROCKING-HEARTH ARC-MELTED ALLOYS PREPARED FROM HIGH-PURITY NIOBIUM

Nominal Alloy Content (Balance Niobium), a/o	Total Weight Change, mg per cm^2
Unalloyed Nb	1.02
1 Zr	-7.75
2.5 Ti	0.59
10.0 Ti	0.34
25.0 Ti	0.28
1 Cr	0.98
5 Cr	0.65
2.5 V	0.59

TABLE F-5. COMPOSITIONS, THERMAL-NEUTRON-ABSORPTION CROSS SECTIONS, AND HARDNESSES OF 50-G HIGH-PURITY NIOBIUM-BASE ALLOYS

Alloy	Nominal Alloy Content (Balance Niobium), a/o	Cross-Section, barns per atom	As-Cast Hardness, VHN
N56	100 Nb	1.15	93
N45	1 Fe	1.16	89
N46	5 Fe	1.23	116
N47	10 Fe	1.28	238
N48	10 Cr(a)	1.34	84
N16	2.5 V(b)	1.22	106
N49	2.5 V-2.5 Ti	1.36	115
N50	2.5 V-2.5 Mo	1.29	127
N51	2.5 V-2.5 Fe	1.28	136
N52	2.5 V-2.5 Ni	1.34	177
N53	2.5 V-2.5 Cr(a)	1.30	106
N54	2.5 V-2.5 Al(a)	1.22	105
N55	2.5 V-2.5 Zr	1.22	149

(a) High weight losses in melting indicate part or all of the chromium or aluminum added to these alloys may have been lost during melting.

(b) Earlier data reported here for comparison purposes.

stainless steel with Ingots 47, 52, and 55. The balance of these alloys finished to good quality 45-mil-thick strip. Samples of each alloy, as annealed, were submitted for corrosion testing in 680 F water.

Double consumable-electrode arc melting was completed on a niobium-45 a/o zirconium-3 a/o vanadium alloy. Half-inch-thick slabs of this ingot, which had an average as-cast hardness of 258 BHN, were then clad in evacuated stainless steel packs for attempted rolling at 1800 F. While the pack rolled easily to 0.125-in.-thick strip, the alloy fragmented severely and cannot be considered fabricable under these conditions.

Rubbing Surfaces in Sodium Environments

J. W. Kissel, C. M. Allen, and W. A. Glaeser

Studies are being conducted in an effort to understand the phenomena involved in operating various devices which are required for systems containing molten alkali metals. This program has been investigating friction and wear of rubbing surfaces in these environments. The goal for this program is to establish design parameters for rubbing components which must operate in these unusual environments. The current effort involves friction-versus-temperature measurements for molybdenum sliding against itself, dry and sodium lubricated. The specimen geometry (ball on a plate) and sliding speed are designed to assure nonhydrodynamic friction. To date, in the renewed program, friction recordings have been obtained for 87 experiments performed in vacuum or argon gas and for 175 experiments with sodium present between the sliding specimens.

The "stick-slip" friction records obtained from these experiments have been interpreted in terms of average coefficient of kinetic friction. For dry sliding the coefficient varied between about 0.5 and 1.2 over a temperature range from 80 to 1300 F. Except for the fairly consistent high friction found at high temperatures, no definite trend was established because of the large variability among the several runs made at each temperature. Following overnight conditioning of the specimens at 1000 F in a very good vacuum, the coefficient of friction varied between 0.3 and 0.4 over the same large temperature range, and the spread among runs made at each temperature was only about one-sixth as great. In the case of the sodium-lubricated runs, the friction records demonstrated that an even greater decrease in the kinetic friction can result from simply adding molten sodium – even at temperatures below 400 F. These low values for the coefficient of kinetic friction, between 0.1 and 0.4, continued even after the sodium had been evaporated from the rubbing surfaces at high temperature. Deposition of this evaporated sodium in the form of finely divided particles on all relatively cool surfaces within the apparatus provided very effective "gettering" action for gaseous impurities during succeeding friction runs. As a result, perfectly clean and shiny molten sodium surfaces could be maintained indefinitely. A second addition of a larger amount of molten sodium completely immersed the flat specimen, but this did not appreciably alter the coefficient of friction. The result was the same even after sufficient air was admitted to the system to produce a voluminous oxide surface film on the pool of sodium. Subsequent heating and cooling cycles again produced the long-lasting oxide-free shiny sodium surface which was observed previously.

The presence of a layer of MoO_3 on lapped and cleaned specimens was indicated by electron diffraction. This reduced to MoO_2 when heated in a vacuum of 5 x 10^{-6} mm of mercury. The friction runs indicate higher friction between the surfaces coated by the higher oxide. The addition of sodium to the test surfaces produces a similar reduction in friction. The mechanism of the latter effect is being further investigated. The results of differential thermal analysis, to discover whether a reaction occurs between MoO_2 and sodium, were confused by "wetting" of the stainless steel container walls. This experiment will be repeated with the "wetting" reaction included in the analysis.

STUDIES OF ALLOY FUELS

R. F. Dickerson

Binary alloys of niobium and 10, 20, 30, 40, 50, and 60 w/o uranium are being exposed to air at 572, 662, and 752 F. All of the alloys exhibit a black adherent film and consequently good resistance to corrosion at 572 F. At 662 and 752 F a brown spalling film forms during 40 hr of testing. After 4 weeks of exposure to 600 and 680 F water, all of the alloys exhibit corrosion behavior comparable with unalloyed niobium. Tests are being conducted in 750 F steam, in 600 F CO_2, and in molten NaK. Tensile testing at room temperature and 1600 F is in progress.

Recrystallization temperatures for binary thorium-uranium alloys are being determined, and thermal-analyses data have been obtained from thorium-uranium-zirconium and thorium-uranium-zirconium-niobium alloys.

Development of Niobium-Uranium Alloys

J. A. DeMastry, F. R. Shober, and R. F. Dickerson

The potential of alloys of the niobium-rich portion of the uranium-niobium constitution diagram as high-temperature reactor fuels has not been realized. Use has not been made of these alloys due to the lack of information concerning their fabricability, their mechanical and physical properties, and their compatibility with various environments. Since prior work has indicated that properties may be influenced by impurity content, the effect of impurities (oxygen and zirconium) on the above-mentioned properties is being investigated.

Three grades of niobium were used in alloy preparation: one containing 0.7 w/o zirconium and 600 ppm oxygen, a second containing 0.17 w/o zirconium and 700 ppm oxygen, and a third containing less than 200 ppm zirconium and less than 300 ppm oxygen. The niobium-10 and -20 w/o uranium alloys have been successfully fabricated into slab material.

Fabrication of alloys with greater than 20 w/o uranium has not been successful to date. Additional alloys have been prepared and will be fabricated when a high-temperature furnace under construction has been completed. There has been no evidence to indicate that the purity of the base niobium has any effect on the fabrication characteristics of the alloys studied.

Table F-6 shows the results of oxidation tests on the uranium-niobium alloys at 575, 662, and 752 F. This study is incomplete. All alloys exhibit good corrosion resistance at 572 F; at this temperature, a black adherent film is formed. In all cases a brown spalling oxide is formed at 662 and 752 F during 40 hr of testing. At the present time, no appreciable effect due to use of different types of niobium as base melting stock has been observed.

TABLE F-6. CORROSION DATA FOR URANIUM NIOBIUM ALLOYS IN AIR[(a)]

Nominal Alloy Content (Balance Uranium), w/o	Impurity Content		Specimen Condition	Total Weight Change, mg per cm^2			
	Oxygen, ppm	Zirconium, w/o		After 67 to 886 Hr		After 100 Hr	
				Hr	572 F	662 F	752 F
40 Nb	600	0.74	As cast	67	0.47	1.82	22.2
	700	0.17	As cast	67	0.15	2.31	26.2
	300	0.02	As cast	67	0.18	2.81	30.4
50 Nb	600	0.74	As cast	67	0.04	3.74	26.7
	700	0.17	As cast	67	0.10	11.00	24.0
	300	0.02	As cast	67	0.04	13.1	29.6
60 Nb	600	0.74	As cast	67	0.00	9.0	30.2
	700	0.17	As cast	719	0.32	10.0	27.6
	300	0.02	As cast	719	1.17	8.9	37.1
					.31		
70 Nb	600	0.74	As cast	719	--	--	--
	700	0.17	Fabricated	--	--	--	--
	300	0.02	As cast	719	0.41	--	--
80 Nb	600	0.74	Fabricated	--	--	2.6	--
	700	0.17	Fabricated	886	1.3	29.2	18.2
	300	0.02	Fabricated	886	0.66	25.5	175
90 Nb	600	0.74	Fabricated	886	0.82	4.5	324
	700	0.17	Fabricated	--	--	1.6	--
	300	0.02	Fabricated	--	--	--	--

(a) Average of two specimens.

Table F-7 lists 2- and 4-week corrosion data for uranium-niobium alloys in 600 and 680 F water. In 600 F water, all of the alloys are exhibiting corrosion behavior comparable with unalloyed niobium. All alloys are gaining weight and show a black adherent film. In 680 F water, most of the alloys showed weight losses after 4 weeks of exposure, indicating that spalling of the oxidized metal is occurring. These behaviors at 600 and 680 F follow the general pattern for the corrosion of unalloyed niobium in water. No effect of different impurity levels in the niobium on the corrosion properties has been noted.

TABLE F-7. CORROSION DATA FOR URANIUM-NIOBIUM ALLOYS IN WATER AFTER 14 AND 28 DAYS[(a)]

Alloy Content (Balance Uranium), w/o	Impurity Content		Specimen Condition	Total Weight Change, mg per cm^2			
	Oxygen, ppm	Zirconium, w/o		In 600 F Water		In 680 F Water	
				14 Days	28 Days	14 Days	28 Days
40 Nb	600	0.74	As cast	0.35	0.42	-24	-31.5
	700	0.17	As cast	0.36	0.48	0.26	-2.4
	300	0.02	As cast	0.24	0.36	-0.02	-1.15
50 Nb	600	0.74	As cast	0.31	0.47	0.47	0.29
	700	0.17	As cast	0.33	0.46	0.39	0.40
	300	0.02	As cast	0.31	0.46	0.27	0.00
60 Nb	600	0.74	As cast	0.64	0.83	0.75	1.00
	700	0.17	As cast	0.33	0.50	0.43	0.30
	300	0.02	As cast	0.28	0.40	0.65	0.80
70 Nb	600	0.74	As cast	0.43	0.53	1.19	0.38
	700	0.17	Fabricated	0.33	0.42	0.88	0.43
	300	0.02	As cast	0.45	0.64	0.70	0.68
80 Nb	600	0.74	Fabricated	0.25	0.42	0.48	0.61
	700	0.17	Fabricated	0.39	0.49	0.68	1.05
	300	0.02	Fabricated	0.25	0.33	0.38	0.55
90 Nb	600	0.74	Fabricated	0.21	0.28	0.32	0.28
	700	0.17	Fabricated	0.31	0.48	-1.52	-3.66
	300	0.02	Fabricated	0.35	0.53	-14.3	-23.8
100 Nb	131	0.05	Fabricated	1.73	2.30	0.79	1.00
	500	0.03	Fabricated	1.95	3.50	0.41	-1.36

(a) Average of two specimens.

Table F-8 shows the results of corrosion testing of uranium-niobium alloys in 750 steam for 14 and 21 days. All specimens are losing weight due to spalling from the surface.

Table F-9 shows the results of exposure for a few of the uranium-niobium alloys in carbon dioxide at 600 F. This work is incomplete, and additional specimens are on test. Corrosion testing of specimens in NaK is currently being conducted, but no results have been obtained.

All tensile specimens have been machined and will be tested at room temperature and 1600 F.

Results have not been obtained from thermal-conductivity, electrical-resistivity, and thermal-expansion measurements.

Construction of a high-temperature furnace for future fabrication studies is continuing.

TABLE F-8. CORROSION DATA FOR URANIUM-NIOBIUM ALLOYS IN 750 F STEAM

Alloy Content (Balance Uranium), w/o	Impurity Content		Specimen Condition	Total Weight Change, mg per cm^2	
	Oxygen, ppm	Zirconium, w/o		After 14 Days	After 21 Days
40 Nb	600	0.74	As cast	-8.22	-11.1
	700	0.17	As cast	-0.49	-8.60
	300	0.02	As cast	-4.32	-6.87
50 Nb	600	0.74	As cast	-3.84	-6.90
	700	0.17	As cast	-1.54	-3.52
	300	0.02	As cast	-3.95	-6.47
60 Nb	600	0.74	As cast	-0.03	-0.28
	700	0.17	As cast	0.00	-0.33
	300	0.02	As cast	0.73	0.81
70 Nb	600	0.74	As cast	-1.88	-2.24
	700	0.17	Fabricated	-2.79	-3.14
	300	0.02	As cast	-4.20	-4.98
80 Nb	600	0.74	Fabricated	0.63	-0.43
	700	0.17	Fabricated	1.28	-3.87
	300	0.02	Fabricated	0.76	0.27
90 Nb	600	0.74	Fabricated	0.49	0.24
	700	0.17	Fabricated	-5.59	-7.59
	300	0.02	Fabricated	-13.9	-18.5
100 Nb	131	0.05	Fabricated	--	-3.22
	500	0.03	Fabricated	--	-8.05

TABLE F-9. CORROSION DATA FOR URANIUM-NIOBIUM ALLOYS IN CO_2 AT 600 F

Alloy Content (Balance Uranium) w/o	Impurity Content		Specimen Condition	Total Weight Change in 100 Hr, mg per cm^2
	Oxygen, ppm	Zirconium, w/o		
40	600	0.74	As cast	0.34
40	300	0.02	As cast	0.18
80	600	0.74	Fabricated	0.43
90	600	0.74	Fabricated	0.20
90	700	0.17	Fabricated	0.28
90	300	0.02	Fabricated	0.28

F-15

Development of Thorium-Uranium Alloys

M. S. Farkas, A. A. Bauer, and R. F. Dickerson

Improvement of the corrosion resistance and of the irradiation behavior of thorium-uranium alloys is being sought by means of alloying and control of processing variables. Ternary and quaternary additions of molybdenum, niobium, and zirconium have been made for the purpose of stabilizing the gamma-uranium phase and for strengthening the thorium matrix. Several fabrication variables are being studied to determine their effect on the size and distribution of uranium particles within the thorium binary alloys.

Studies of cast binary thorium-uranium alloys have shown that uranium-particle distribution is random in arc-melted alloys, whereas induction-melted alloys show higher concentrations of uranium in the grain boundaries. This difference is due to the different cooling rates encountered in the melting process. The fast cooling during arc melting was simulated by water quenching induction-melted alloys from 1000 C. The structures thus obtained were similar to those of the arc-melted alloys.

Recrystallization of thorium-5, -10, -15, and -20 w/o uranium alloys, cold reduced 90 per cent, begins within 2.5 min at a temperature of 700 C. Additional information is being obtained.

Thermal-analysis data obtained from thorium-uranium-zirconium and thorium-uranium-zirconium-niobium alloys is presented in Table F-10. Tentative phase changes indicated by these data are also given.

TABLE F-10. THERMAL-ANALYSIS DATA OBTAINED FOR THORIUM-URANIUM-ZIRCONIUM AND THORIUM-URANIUM-ZIRCONIUM-NIOBIUM ALLOYS

	Transformation Temperature in Thorium-Base Alloy[a] Shown, C								
Phase Transformation	10 U-10 Zr	20 U-20 Zr	25 U-25 Zr	15 U-15 Zr-3 Nb	10 U-5 Zr-2 Nb	20 U-10 Zr-4 Nb	10 U-10 Zr-2 Nb	15 U-7.5 Zr-3 Nb	20 U-20 Zr-4 Nb
$\alpha Th+UZr_2 \rightarrow \alpha Th+UZr_2+\gamma U-Zr$	550	552	549	529	--	--	--	--	--
$\alpha Th+UZr_2+\alpha U-Zr \rightarrow \alpha Th+\gamma U-Zr$	583	585	587	564	--	--	--	--	--
$\alpha Th+\gamma UZr \rightarrow \alpha Th+UZr+\gamma Th$	963	966	990	1017	1085	1072	1017	1068	1016
$\alpha Th+\gamma UZr+\beta Th \rightarrow \beta Th$	1045	999	1002	1030	1145	1102	1064	1088	1030
$\beta Th \rightarrow \beta Th+liquid$	1165	1200	1199	1178	1224	1185	1187	1201	1179
$\beta Th+liquid \rightarrow liquid$	1183	1262	1239	1194	1263	1227	1212	1237	1195

(a) Compositions are given in weight per cent.

F-16

FISSION-GAS RELEASE FROM REFRACTORY FUELS

J. Melehan, D. A. Vaughan, R. Barnes,
S. D. Beck, and F. A. Rough

Preparations for the study of fission-gas release from UO_2 under irradiation are being made. Supporting experiments on characterization of different types of UO_2 and on diffusion in fused single crystals of UO_2 having known geometry are reported. Construction of the apparatus for in-pile study is expected to be complete about August 1, 1959.

Diffusion in UO_2

Specimens of single-crystal UO_2 are being prepared for study of fission-gas diffusion coefficients by low-temperature irradiation and subsequent heating to collect the fission gases. Two groups of single crystals are being considered. One is from a lot of fused single crystals obtained from the Norton Company. The other was supplied by the Mallinckrodt Chemical Works and was prepared by the Verneuil process. Single-crystal material was chosen for this study, of course, because the geometry and the model of release is not in question as it is in porous sintered material. Because of nitride impurities, vacuum and hydrogen heat treatments and combinations of both have been used in purifying the Norton crystals. Hydrogen heat treatment at 1500 C removes the original impurity but apparently introduces a second unidentified contaminant. Vacuum heating does not completely remove the nitrides; however, results of a 2-hr anneal at 1800 C are somewhat encouraging. The contaminant was removed from a surface layer about 3 to 5 mils thick and partial purification and agglomeration occurred in regions farther from the specimen surface. Estimates based on available diffusion data suggest that a purified zone 5 mils thick should be sufficient for the proposed diffusion studies. Attempts will be made to duplicate the results of the vacuum heat treatment.

The thin-plate and spherical specimen geometries are being considered. Dimensions and surface-condition specifications and methods of preparation have been established. Briefly, the plate specimens are being prepared as follows. Large fused crystals are sliced into plates 70 mils thick. Subsequent grinding and polishing produce specimens with dimensions of 0.250 by 0.250 by 0.050 in. All surfaces are finished with Linde B aluminum oxide. Final polishing is continued until etching reveals no subsurface polishing artifacts. The polished specimens will be heat treated to remove the nitride contaminant (a sufficient number of control specimens will be retained for comparison with microstructures of heat-treated and irradiated specimens). Final etching before irradiation will remove any surface contamination or tarnishing of the oxide which might result from the heat treatment. Similar techniques will be used for the preparation of spherical specimens about 0.2 in. in diameter. Several plates and spheres have been prepared to the required size and surface condition; these will be used in trial experiments to test the operation of postirradiation heating and gas-trapping equipment.

Plans for preparation of samples of the specimens of the MCW Verneuil crystals are being prepared. At the moment, activities are confined to study of techniques of preparing small spherical samples.

Characterization of Sintered UO_2 and Model of Gas Release

Results are reported this month primarily of efforts to develop additional techniques for study and characterization of UO_2 materials. Techniques investigated included optical and infrared transmission of thin sections. Thin specimens, about 40 μ thick, were polished to a reflecting surface on both sides and examined in reflected or transmitted light. Reflected light distinguished between voids and inclusions, while transmitted light showed the distribution of these imperfections throughout the body of the UO_2.

Examination of the Norton fused UO_2 revealed numerous defects, even in a large piece which had very little uranium nitride on its surface. These defects, when examined by reflected light, were identified as inclusions, apparently of the same phase as that identified as U_2N_3 on outer surfaces of the piece. In the thin sections examined, only one void was observed at the surface. This would suggest that very few of the numerous defects observed in the 40-μ sections are voids. The larger inclusions were angular and about 4 μ across. Many smaller inclusions were visible. Defects in the surface due to specimen preparation were easily distinguished from voids or inclusions. These surface defects were present as fine scratches or chips which result in conchoidal fractures, rather than the regular shapes which were generally observed in the case of inclusions or voids.

Two types of the MCW crystals have been examined by the techniques employed on the Norton UO_2; one type is needle shaped while the other is partially spheroidal, with a radial growth of needles. The needle type has an oxygen/uranium ratio of 2.0 while the radial crystals have an oxygen/uranium ratio of 2.05. Neither of these contains a second phase that can be detected by X-ray diffraction or optically in the polished section. The radial-type crystals contain many angular voids, but are not transparent to visible light in thin sections. In the case of the needle-type MCW crystals, thin sections are transparent. These contain no visible voids or inclusions. There were no defects in these needle-type crystals that were not attributed to polishing or to gross slip induced during preparation of the thin sections. The MCW needle-type UO_2 crystals would appear to contain the least defects of any material examined for the fission-gas release program.

In order to compare the above material with the more practical fuel material, sintered bodies of 90 and 98 per cent of theoretical density have been examined in thin sections. The larger voids of about 2 μ in diameter in the 90 per cent dense body appear to be located in the grain boundaries. In the 98 per cent dense body, a large fraction of the large grain-boundary voids were not observed. However, clusters of smaller voids are visible within the grains of the latter specimen. More small inclusions are visible under reflected light in the 98 per cent dense sintered body. Although the voids in the sintered bodies are smaller than the inclusions in the Norton fused UO_2, there are many more voids.

Two specimens of uranium dioxide powder ($UO_{2.02}$ and $UO_{2.08}$) were examined for their transmission of infrared over the wavelength range 2 to 15 μ. The analysis of the spectra of these samples indicates that both compositions are completely transparent. It is interesting that the thin sections of single-crystal specimens which had an oxygen/uranium ratio of 2.01 were transparent to visible red light at a wavelength of about 0.7 μ. However, a specimen with an oxygen/uranium ratio of 2.05 did not transmit visible light. These results would indicate an absorption band between 0.7 and 2 μ.

Preparation for In-Pile Study

Effort has continued on the design and construction of an apparatus for study of the effects of neutron irradiation upon gas release from sintered UO_2. The apparatus is to be installed in a beam tube of the Battelle Research Reactor in August.

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

Fabrication techniques are being developed for the preparation of cermet fuel elements containing 60 to 90 volume per cent of fuel. Recent studies have been concerned with the fabrication and cladding of dispersions consisting of 80 volume per cent of UO_2 and 20 volume per cent of molybdenum or chromium. The powder mixtures are cold pressed and then clad and densified by either gas-pressure bonding or hot-press forging.

The gas-pressure-bonding method is being utilized for the bonding of niobium and molybdenum fuel elements and subassemblies. Strong metallurgical bonds have been achieved; however, bonding is usually accompanied by a loss of ductility. Results of tests conducted during this report period indicate that this loss of ductility can be minimized by improved surface preparation, reduction of bonding temperature, proper assembly, and control of impurities in the helium autoclave atmosphere.

Dispersion fuels consisting of 24 w/o UN or UC dispersed in stainless steel and clad with stainless have been irradiated at temperatures of from 1500 to 1700 F to estimated burnups of 2 to 12 a/o of the uranium. These specimens are now being evaluated to determine more accurately the average temperature of irradiation, the uranium burnup, and dimensional and structural changes.

Studies are being conducted to establish the mechanism and kinetics of solid-state bonding of the type involved in the gas-pressure-bonding process. The bond interface produced during hot-pressure bonding resembles a grain boundary under metallographic observation; however, during heat treatment, it behaves differently from a grain boundary.

F-19

Fabrication of Cermet Fuel Elements

S. J. Paprocki, D. L. Keller, G. W. Cunningham,
and D. E. Kizer

Evaluation of methods of producing cermets of 90 per cent of theoretical density or better containing 60 to 90 volume per cent fuel are being made on the basis of microstructures and physical and mechanical properties. Techniques being evaluated for producing dense cermets are hot-press forging and pressure bonding. Thermal-conductivity measurements are being made on cermets of various fuel loadings.

Green-pressed pellets 1 in. in diameter, containing 80 volume per cent UO_2 dispersed in molybdenum and in chromium, have been fitted into stainless steel tubes and sealed. The assemblies were then pressure bonded for 3 hr at 2300 F under a helium gas pressure of 10,000 psi. Densities in both cases were increased from 72 per cent of theoretical to 93.6 per cent of theoretical for the chromium matrix and 86.7 per cent of theoretical for the molybdenum matrix. The stainless cladding is being machined from each cermet in order to utilize the specimens for obtaining thermal-conductivity data.

In addition, 1-in.-diameter green-pressed pellets of 80 volume per cent UO_2-chromium have been sealed in stainless steel frames with 1/8-in. cover plates. The packs were hot-press forged at 1900 F to reductions ranging from 10 to 40 per cent in thickness. Cores reduced less than 30 per cent in thickness were free from surface cracks, while those reduced 30 per cent or more in thickness had visible surface cracks. Density measurements have not yet been completed.

Thermal-conductivity measurements have been made on a rod of 80 volume per cent UO_2-Type 302B stainless steel cermet of 93.8 per cent of theoretical density. The cermet was produced by hydrostatic pressing the green-pressed mixture for 3 hr at 2300 F under a helium gas pressure of 10,000 psi. Values of 0.063, 0.069, and 0.093 w/(cm)(C) were obtained at 100, 400, and 750 C, respectively. Electrical-resistivity values of 3.1, 2.7, and 2.0×10^{-3} ohm-cm were measured at 100, 400, and 650 C, respectively.

Pellets of 80 volume per cent UO_2 dispersed in molybdenum and in chromium are being prepared for hot-press forging at temperature from 2000 through 2200 F. In addition, green 1-in.-square cores of 80 volume per cent UO_2-molybdenum are being prepared for pressure bonding.

Gas-Pressure Bonding of Molybdenum- and Niobium-Clad Fuel Elements

S. J. Paprocki, E. S. Hodge, R. W. Getz,
and P. J. Gripshover

The gas-pressure bonding technique is being studied as a possible method for cladding ceramic and cermet fuels with molybdenum or niobium. High-temperature strengths and favorable nuclear properties make these metals desirable cladding materials for fuel elements.

Surface-preparation techniques are being investigated for the bonding of niobium at the lowest possible temperature to minimize loss in ductility of the niobium during the bonding cycle. Specimens bonded at 2100 F at 10,000 psi for 3 hr exhibited adequate flow and diffusion with these bonding parameters; however, evidence of contamination that prevented complete grain growth was observed along the original bond interface. Attempts are being made to further reduce this observed contamination by surface preparation. Evaluations of specimens treated in a saturated sodium hydroxide solution or a chromic acid solution have shown that both surface preparations result in residual contamination on the bonding surfaces that prevents complete grain growth across the original bond interface. Nevertheless, bend and peel tests indicate that these bonds have excellent strength. Specimens are presently being evaluated to determine the merits of machining, belt abrading, and pickling in a nitric-hydrofluoric acid solution as surface-preparation treatments.

A number of materials are being investigated as barriers between the stainless steel pressure-bonding container and the niobium to prevent contamination of the niobium by diffusion during bonding. Complete evaluation of graphite-coated spacers has shown that a considerable amount of carbon diffuses into the niobium specimen at the higher bonding temperatures. TiNamel plates coated with flame-sprayed aluminum oxide or aluminum oxide with titanium oxide additions, or hard lime window glass between the specimen and the container, have been used as spacer materials, and metallographic evaluations are being made of these materials.

In an effort to minimize the loss of ductility during bonding, the stainless steel cans and spacers used to contain the specimen are outgassed at 2100 F in vacuum prior to assembly. In addition, modifications in the autoclave-loading technique have permitted the use of a number of Zircaloy plates to getter the autoclave gas. Specimens bonded under these conditions have exhibited better ductility than any previously bonded specimens.

Several molybdenum specimens were canned in vacuum-degassed Type 304 stainless steel containers and bonded for 3 hr at 2300 F and 10,000 psi. Each of these specimens consisted of two metal plates cut from molybdenum which had been cold rolled with a 4-to-1 reduction. The surfaces were either shaped to various roughness values, belt abraded, pickled in chromic acid followed by a sodium hydroxide solution treatment, or pickled in nitric acid under kerosene. Preliminary evaluations indicated the specimens were more ductile than specimens bonded at higher temperatures. Metallographic examinations showed that the specimens prepared with nitric acid under kerosene were consistently bonded, with greater than 50 per cent grain growth across the original interface. Evaluation of the belt-abraded and the machined specimens has shown that some residual contamination remained after surface conditioning. Further investigations will be conducted with these surface preparations in an effort to obtain improved bonds.

Spacers used in the current molybdenum specimens, between the molybdenum and the bonding container, included graphite-coated molybdenum, flame-sprayed alumina on TiNamel plates, and hard lime glass. Chemical analysis of specimens next to graphite-coated spacers has indicated that excessive carbon pickup had occurred. No apparent reaction of molybdenum with either flame-sprayed alumina or hard lime glass was observed metallographically. However, the flame-sprayed coating particles embedded in the specimens during bonding, which indicates lapping of the coatings may be required to minimize such grit pickup. Of the spacer materials investigated to date, hard lime glass appears the most promising; however, additional investigations are needed to produce ductile molybdenum specimens.

The Irradiation of UC- and UN-Stainless Steel Dispersion-Type Fuel Elements

D. L. Keller and A. W. Hare

The high-temperature irradiation behavior of dispersion fuel elements consisting of 24 w/o UN or UC dispersed in stainless steel and clad with stainless steel is being evaluated in this program.

Twelve test specimens, six containing UN as fuel and six containing UC, have been irradiated in three NaK-filled capsules in the MTR to estimated burnups of 2, 6, and 12 a/o of the uranium. Two specimens of each type of fuel were contained in each capsule. The specimens are about 1-1/2 in. long by 11/16 in. wide by 47 mils thick. The stainless steel cladding is approximately 8 mils thick.

The postirradiation density and dimension measurements have been completed on all twelve specimens. Currently in progress are metallographic examinations of each type of fuel at the three burnup levels. Radiochemical analyses are also being run on two specimens from each burnup level. Dosimeter wires which were adjacent to the specimens during irradiation are being analyzed. The burnup data, dosimetry data, and temperature data will be compared and evaluated to determine the average burnup per specimen.

When all of the temperature, burnup, and metallography data are available, the results will be compared with dispersion fuel elements of UO_2 of comparable fuel loading and burnup.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

The solid-state bonding of metals by application of heat and pressure is being studied in an attempt to establish the mechanism and kinetics of the process. Most of the specimens used in the investigation have been machined from copper plate. However, iron, nickel, and zirconium specimens have also been prepared.

There has been some difficulty in evaluating specimens made from electrolytic tough-pitch copper because of the presence of copper-Cu_2O eutectic. Although it is planned to use OFHC copper, material has not yet been received for specimen preparation. However, excellent specimens have been prepared for these studies by melting electrolytic tough-pitch copper in a hydrogen atmosphere, cold rolling to plate, machining, and annealing at 1400 F in hydrogen before use. Specimens prepared in this manner show no evidence of oxides or other secondary phases, and no indication of impurities at the bond line can be detected in pressure-bonded specimens.

Two specimens of the hydrogen-cast copper have been pressure bonded by hot-pressing techniques and are being used in annealing studies. One specimen was prepared by bonding an 8-rms surface of a plate annealed at 1400 F to a 500-rms surface of a plate previously cold worked approximately 50 per cent. The specimen was bonded by pressing at 10,000 psi, heating to 1100 F, and repressing at 10,000 psi. The second specimen was prepared by heating to 1100 F two plates previously annealed at 1400 F and pressing at 10,000 psi. In both cases after pressing there was no evidence of porosity at the bond line, and the bond line very closely resembled an ordinary grain boundary at magnifications up to 1500X. Although the width of the bond line varied, it was in general about equal to the width of the widest grain boundaries. In a few places, the bond line could not be detected at 1500X except for small dots which could have been pores.

Sections of these specimens are being annealed at 1900 F for various times in order to gain information on the nature of the bond line. Examination of the specimens after a 2-hr anneal at 1900 F has been completed. After the anneal, the bond line still resembled grain boundaries, but there was evidence of porosity at some points along the bond line. While the porosity was negligible in the specimen annealed before bonding, a few pores could be detected at low magnifications (100X) in the specimen bonded by pressing a cold-worked plate on an annealed plate. It is interesting to note that although the bond line appears to be similar to a grain boundary, it does not behave in a similar manner. The bond line apparently does not move or tend to straighten out during the anneal, even though the average grain size is increased by a factor of 10.

Copper specimens are also being annealed in liquid bismuth to determine whether diffusion is more rapid along the bond line than along grain boundaries. Penetration of the bismuth in a specimen annealed 26 hr at 800 F was not sufficient to allow comparative measurements. Additional specimens are being prepared for these diffusion studies.

FF. FUEL-CYCLE PROGRAM STUDIES

GAS-PRESSURE BONDING OF CERAMIC, CERMET, AND DISPERSION FUEL ELEMENTS

S. J. Paprocki, D. L. Keller, E. S. Hodge,
C. B. Boyer, J. B. Fox, and D. E. Kizer

The objective of this program will be to refine and further develop the gas-pressure-bonding process for the simultaneous densifying and cladding, with stainless steel, of ceramic, cermet, and dispersion fuels. Primary emphasis will be placed on the reduction of fuel-element manufacturing cost while maintaining or improving the quality of the fabricated elements.

Initial efforts in this study will be directed toward the development of uranium dioxide fuel material clad with Type 304 stainless steel. The work will include fabrication development for several basic fuel-element designs. The general results that are obtained with the ceramic uranium dioxide fuel element should also be applicable to the other fuels being considered. A small effort will be directed to the development of uranium dioxide cermets and dispersion systems.

Investigations are being conducted to determine the effects of particle size, density, porosity, stoichiometry, microstructure, and preparation of various types of uranium dioxide powders on the initial cold-pressed density. It would be desirable from a cost standpoint to be able to eliminate the sintering step and use directly cold-pressed cores of a high density in the gas-pressure-bonding process.

Five types of uranium dioxide powders have been tentatively selected for the preliminary phases of this program: Mallinckrodt ceramic, high-fired, high-density, and spherical types, and Nuclear Equipment and Materials Company high-fired type.

The effects of various binders in achieving high cold-pressed densities are being studied. Preliminary results indicate that a binder consisting of paraffin in benzene or Ceremul C should help to attain a high green density. However, sizing of the powders is also important. Compacts 10 through 15, as shown in Table FF-1, were prepared with minus 325-mesh powder. In some cases, the minus 325-mesh size was achieved through ball milling powder of a coarser size. Compacts prepared with this ball-milled minus 325-mesh UO_2 powder exhibited higher densities in practically all cases. This trend gives a basis for investigation of ball milling for extended periods. At present, batches of ceramic-grade UO_2 powder are being treated by ball milling for various lengths of time. The oxygen-to-uranium ratio will be checked analytically after these tests to determine if an increase of this ratio will result in an increased density. In Table FF-1, Compacts 11 through 15 were used as a basis for checking various binders. In all cases, the powder was minus 325 mesh. In an earlier study, compacts made with these binders had green densities which were in a range of 70 per cent of theoretical or better.

Compacts 1 through 9 in Table FF-1 show the results of compacting under the various conditions which gave a high green theoretical density. As shown in the table, high densities can be achieved without binders when minus 325-mesh powder is

TABLE FF-1. GREEN COMPACTING OF UO_2 POWDERS UNDER VARIOUS CONDITIONS

Compact	UO_2 Mesh Size	Type of UO_2	Binder Material	h/d Ratio	Compacting Pressure, tsi	Green Density G per Cm^3	Green Density Per Cent of Theoretical
1	-100	High fired	Polyvinyl alcohol	0.25 (plate)	50	6.41	58.49
2	-100	High fired	Polyvinyl alcohol	Ditto	50	6.80	62.04
3	-100	High fired	Polyvinyl alcohol	Ditto	50	6.76	61.68
4	75 per cent -100 +325 25 per cent -325	High fired	Polyvinyl alcohol	Ditto	50	6.69	61.04
5	75 per cent -100 +325 25 per cent -325	High fired	Polyvinyl alcohol	Ditto	40	6.70	61.13
6	-100	High fired	Slurry of camphor and alcohol, air dried	Ditto	40	7.63	69.62
7	-100	High fired	Slurry of stearic acid - alcohol, air dried	Ditto	40	7.78	70.99
8	-100	High fired	Slurry of camphor and alcohol, air dried	Ditto	30	7.77	70.89
9	-100	High fired	Polyvinyl alcohol	Ditto	50	6.60	60.22
10	-325	High fired	No binder	0.70 (pellet)	20	7.58	69.00
11	-325	High fired	2 w/o paraffin/alcohol	0.66 (pellet)	20	7.70	70.30
12	-325	High fired	0.5 w/o MgO	0.66 (pellet)	50	7.82	71.40
13	-325	High fired	2 w/o urea	0.39 (pellet)	20	8.15	74.2
14	-325	High fired	2 w/o Ceremul C	0.35 (pellet)	20	8.35	76.0
15	-325	High fired	2 w/o paraffin/ benezene	0.33 (pellet)	20	9.34	85.0

employed. However, in pellets prepared previously with coarser powder, the average theoretical density for green-compacted pellets using no binder was approximately 45 to 55 per cent of theoretical.

The controlled densification of ceramic uranium dioxide fuel cores by gas-pressure bonding is being studied. Uranium dioxide cores have been highly densified by this technique as shown in preliminary studies. By proper selection of oxide cores and gas-pressure bonding parameters, it is believed that it will be possible to achieve almost any desired density.

Investigations into the effect of the bonding parameters of time, temperature, and pressure on the bonds obtained with Type 304 stainless steel are being conducted. By this study, a range of optimum gas-pressure-bonding conditions for obtaining acceptable stainless steel bonds will be established. Round specimens consisting of two 1/2-in. diameter by 1-3/4-in.-long Type 304 stainless steel rods assembled into a 0.020-in.-wall stainless steel tube are being utilized. The rods and tubes are being placed through a standard wash cycle after being pickled in 10 volume per cent nitric acid-2 volume per cent hydrofluoric acid aqueous solution prior to assembly. The tubes are sealed and then evacuated to a pressure of 5×10^{-3} mm of mercury, or less, prior to the sealing of the evacuation tube. After the gas-pressure-bonding cycle, each bonded assembly is machined into a standard 1/4-in.-round tensile test specimen with the bond interface located midway in the gage length. These specimens are introduced into a screw-type tensile machine in such a manner that a load is applied along the specimen axis, and the applied load is gradually increased until the specimen breaks. This test gives the ultimate strength of the pressure-bonded Type 304 stainless steel and gives an excellent test for the strength of the bond. Specimens gas-pressure bonded at 2100 F and 10,000 psi for periods of 1-1/2 hr and 3 hr have been tested and examined by metallography. No breaks occurred at either of the bonds. The metallographic study revealed slight contamination at the bond interface, but complete grain growth was observed. The ultimate strength of these two specimens was 83,500 psi as compared with 90,900 psi for the initial material. Additional specimens are being bonded at lower temperatures and pressures.

Compacts 2, 5, and 8 given in Table FF-1 were placed into a flat-plate swastika-type container for pressure bonding. These compacts were utilized to check the design of the pressure-bonding container to be used for flat plates in the initial study, as well as the methods of loading and degassing the compacts. After assembly of the compacts into the containers, they were degassed through the evacuation tube at 1112 F until a constant vacuum was obtained. The containers were then evacuated to a pressure of 5×10^{-3} mm of mercury, or less, for a period of 12 hr and the evacuation tubes sealed. The specimens were pressure bonded at 2100 F for 2 hr at 10,000 psi. All containers appeared well deformed, and they are being measured at the present time to obtain the final densification of each compact.

FF-4

DEVELOPMENT OF URANIUM CARBIDE-TYPE FUEL MATERIALS

F. A. Rough and W. Chubb

Uranium carbides are being developed as a portion of the AEC Fuel-Cycle Development Program. This type of fuel alloy appears to have considerable potential as a low-cost, high-density, refractory, irradiation-resistant, and easily reprocessable fuel for a variety of nuclear reactors. A major obstacle to the development of uranium carbides as reactor fuels was recently overcome by the discovery of techniques for casting of molten uranium carbides into geometric forms. This discovery has enabled the preparation of massive pieces of uranium carbide, and has not only indicated a serious lack of information on the properties of carbide, but also has provided the means for obtaining reliable data.

It is planned to obtain data on uranium carbides in five general areas: (1) investigation of powder-metallurgical techniques for preparation of carbide powders and compacts, (2) development of improved techniques for melting and casting of uranium carbide slugs of controlled size and quality, (3) determination of the physical and mechanical properties of uranium carbides and compositional variations thereof, (4) measurement of diffusion rates of uranium and carbon in uranium monocarbide, and (5) examination of the nature of the processes of neutron-irradiation damage in uranium carbides.

In an initial investigation of the effect of impurities on uranium monocarbide, it was observed that traces of silicon interfere with the casting qualities of the alloy. In measurements of the specimen prepared to date, the mean density of 18 ingots containing 4.8 w/o carbon was 13.56 g per cm^3, while the mean density of 17 ingots containing 5.0 w/o carbon was 13.48 g per cm^3. Electrical resistivity measurements on these same materials indicated that the resistivity of uranium carbide is of the order of 38 μ-ohm-cm with a trend toward higher values for 4.8 w/o carbon. Measurements of the maximum transverse breaking stress of carbide materials produced values up to 24,000 psi. It appears that alloys containing 5.0 w/o carbon are somewhat weaker than alloys containing 4.8 w/o carbon, and that less than 800 ppm of certain impurities are very harmful to strength.

Preliminary studies of diffusion in the UC-UC_2 system show that interdiffusion rates are very rapid at 1980 C (D equal to about 2×10^{-6} cm^2 per sec) and that appreciable diffusion occurs at 1600 C (D equal to about 4×10^{-8} cm^2 per sec).

In powder-metallurgical studies, a sample of minus 100-mesh UC powder was cold pressed at 40 tsi and sintered to 90 per cent of theoretical density in 2 hr at 1800 C. No reaction was observed under the sintering conditions between the pellet and the tantalum tray used to contain it.

Two additional projects are in various stages of planning. A study of the mechanism of irradiation damage to uranium carbides is in the planning stage. That portion of the research having to do with improved melting and casting techniques for carbide materials is expected to be activated in July.

FF-5

Fabrication of UC by Powder Processes

S. J. Paprocki, D. L. Keller, and D. E. Kizer

Several methods of fabricating UC of high density by powder-metallurgy techniques will be investigated in this program. The materials will be evaluated both on the basis of quality and on the basis of projected costs of large-quantity fabrication.

Several pellets of UC powder have been compacted at 40 tsi and sintered at 1500 C and 1800 C. The UC powder for these experiments was prepared by crushing a UC arc-melt button containing between 4.8 and 5.0 w/o carbon. These studies will provide basic information on the sintering and densification characteristics of one type of UC. Seven-gram samples of various particle sizes were weighed and several drops of absolute methyl alcohol were added to each as a binder. The mixes were removed from an argon-filled dry box and pressed immediately at 40 tsi. Green densities ranged from 73 per cent of theoretical for the coarser particles (minus 100 plus 140 mesh) to 65 per cent of theoretical for the finer particles (minus 325 mesh). During vacuum sintering, the pellets prepared from the coarser particles of narrow size distribution (minus 100 to plus 270 mesh) tended to break up. Pellets prepared from minus 100-mesh, minus 270 plus 325-mesh, and minus 325-mesh powder held together during sintering; however, no densification occurred at 1500 C after 3 hr.

A pellet prepared from minus 100-mesh UC powder by pressing at 40 tsi increased in density to approximately 90 per cent of the theoretical value on sintering at 1800 C for 2 hr. It was noted that no reaction occurred between the pellet and a tantalum tray under these conditions.

Modifications of a high vacuum-induction sintering furnace have been made in order to conduct reaction studies of UO_2 and U_3O_8 with carbon. Stoichiometric (UC) mixtures of U_3O_8 and carbon are being prepared to determine the feasibility of a reaction to form UC while sintering to a high density. Meanwhile, modification of a Sieverts apparatus for preparing UC powder by reacting uranium metal with methane are being completed.

Physical Characteristics and Properties of Uranium Carbides

W. M. Phillips, E. L. Foster, and R. F. Dickerson

Research on the properties of uranium carbide has as its ultimate goal the definition of properties as a function of variables and the development of specifications for obtaining reproducibility in the production of high-quality carbide materials. The present initial phase of the work is concerned with the effects of impurities, carbon content, and heat treatment on such properties as ease of casting, hardness, density, resistivity, and corrosion resistance.

Hydrogen, iron, nitrogen, oxygen, and tungsten were selected as elements which might contaminate UC during melting and casting. These elements were added to UC castings in amounts from 100 ppm to 800 ppm as shown in Table FF-2. The specimens were melted six times to insure homogeneity. An inert-arc laboratory furnace was used both for melting and for casting into a 3/8-in.-diameter by 1-3/4-in.-long graphite mold. Hot tops were cut from the specimens, and the ends and circumferences were machine ground using a diamond wheel.

Electrical resistivity measurements were taken using a Kelvin double bridge. The resistivity values shown in Table FF-2 show considerable scatter, probably due to inhomogeneity. However, a trend toward higher values for the 4.8 w/o carbon specimens is indicated.

Densities were determined by micrometer measurements and weighing in air. The piping observed in some specimens during radiographic examination would affect both the density and resistivity values. Since the piping is concentrated along the center line it is believed that it would have little effect on the breaking stress listed in Table FF-2. This is based on the fact that the stress was determined for flexural loading using the "three-point method" where the load is supported almost completely by the material on the circumference of the specimen. The disproportionately low values may be attributed to surface imperfections which have a profound effect on brittle materials. The stress listed was calculated from the formula:

$$\text{Maximum stress} = \frac{2.546 \times \text{breaking load} \times \text{span}}{\text{diameter}^3} .$$

A 1/2-in. span was used in the tests.

Only two specimens, 11 and 31, have low densities. Specimen 11 contains 800 ppm of silicon, which was found to make the production of sound castings difficult. Specimen 31 was melted under 1/15 atm of hydrogen. The breaking stress for the hydrogen-contaminated specimen was extremely low, while the silicon addition prevented obtaining a specimen of sufficient length to obtain a flexure-test specimen. Silicon in excess of 300 ppm can be considered detrimental. Additional chemical analyses of the specimen melted in the hydrogen-contaminated atmosphere are in process.

Specimens containing 4.8 w/o carbon had, in general, greater strength than those containing 5.0 w/o carbon. Thus carbon in excess of the stoichiometric ratio lowers the strength of the material.

Uranium Monocarbide Diffusion Studies

W. Chubb and F. A. Rough

The physical behavior of uranium monocarbide during sintering, during irradiation, and under stress at elevated temperatures can be predicted in a qualitative manner from diffusion data. To provide this type of insight into the nature of uranium carbide, a program is in progress to determine diffusion rates in the uranium-carbon system near 4.8 w/o carbon.

TABLE FF-2. EFFECTS OF IMPURITIES ON SOME PROPERTIES OF CAST UC

Specimen	Nominal Carbon Content, w/o	Impurity Content, ppm	Radiographic Results	Density, g per cm^3	Resistivity, μ ohm-cm	Maximum Transverse Breaking Stress, psi
1	4.8	100 Fe	Minor shrink pipe	13.55	39.2	18,470
2	5.0	100 Fe	Minor shrink pipe	13.44	36.7	13,100
3	4.8	300 Fe	Sound	13.67	42.0	21,000
4	5.0	300 Fe	Minor shrink pipe	13.46	39.7	8,900
5	4.8	800 Fe	Sound	13.43	39.1	14,600
6	5.0	800 Fe	Sound	13.47	39.2	7,200
7	4.8	100 Si	Sound	13.54	39.4	24,100
8	5.0	100 Si	Minor shrink pipe	13.47	36.8	3,800
9	4.8	300 Si	Minor shrink pipe	13.49	34.4	13,700
10	5.0	300 Si	Sound	13.26	35.7	4,300
11	4.8	800 Si	Sound	13.09	(a)	(a)
12	5.0	800 Si	Minor shirnk pipe	13.49	42.3	21,000
13	4.8	100 W	Sound	13.80	37.2	18,100
14	5.0	100 W	Sound	13.42	38.0	7,200
15	4.8	300 W	Sound	13.56	44.0	1,100
16	5.0	300 W	Sound	13.49	38.1	9,900
17	4.8	800 W	Minor shrink pipe	13.52	41.0	14,900
18	5.0	800 W	Minor shrink pipe	13.45	37.5	13,600
19	4.8	100 N	Minor shrink pipe	13.49	43.0	17,300
20	5.0	100 N	Major shrink pipe	13.57	36.2	11,800
21	4.8	200 N	Sound	13.45	37.0	23,600
22	5.0	200 N	Sound	13.45	36.3	9,900
23	4.8	500 N	Sound	13.47	--	22,900
24	5.0	500 N	Sound	13.43	37.6	12,300
25	4.8	100 O	Sound	13.56	42.1	12,200
26	5.0	100 O	Sound	13.54	39.3	(b)
27	4.8	100 O	Sound	13.62	40.7	16,600
28	5.0	200 O	Sound	13.67	--	3,900
29	4.8	500 O	Minor shrink pipe	13.62	41.0	20,800
30	5.0	500 O	Sound	13.63	41.2	18,200
31	4.8	H_2(c)	Sound	13.17	40.8	2,400
32	4.8	None	Minor shrink pipe	13.67	41.2	24,200
33	4.8	None	(d)	--	--	--
34	5.0	None	Sound	13.48	35.4	15,300
35	5.0	None	(d)	--	--	--

(a) Specimen was too short for resistivity and flexure tests.

(b) Specimen broke at a low, undetected load.

(c) Melted under 1/5 atm of hydrogen.

(d) Specimens for corrosion testing (duplicates).

Initial attempts to determine diffusion rates have been based on measurement of the growth of the carbide layer between graphite and liquid uranium saturated with carbon. If certain conditions are fulfilled, this experimental technique can produce accurate diffusion-rate data. Analysis of the carbide layers produced by this technique shows that, as expected, the proper conditions do not exist using this particular experimental setup at temperatures below 2000 C; two layers of carbide are formed. In addition, the data produced show that diffusion rates are too rapid above 2000 C for practical determination of rates at these temperatures.

In an experiment of this type a diffusion zone containing a 0.017-in.-thick layer of UC_2 and a 0.040-in.-thick layer of UC was produced in 101 min at 1980 C. Obviously, diffusion rates are somewhat higher in UC than in UC_2. However, for the purposes of calculation, it was assumed that the diffusion rates in these two compounds were the same and that the outer phase boundaries moved as the square root of time. Under these tenuous assumptions, the diffusion coefficient, D, at 1980 C for interdiffusion of uranium and carbon in UC and UC_2 was calculated to be about 2.2×10^{-6} cm^2 per sec. This figure represents some sort of average of a somewhat higher diffusion rate in UC and a somewhat lower diffusion rate in UC_2. It represents very rapid solid-state diffusion in both phases at this temperature.

In an identical experiment at 1600 C, a diffusion zone of layers of UC and UC_2 having a total thickness of 0.008 in. was produced in 120 min. Using the same tenuous assumptions, the diffusion coefficient, C, for interdiffusion of uranium and carbon in UC and UC_2 was calculated to be about 4×10^{-8} cm^2 per sec.

Because this type of approximate data can be used in designing more accurate experiments, additional data will be obtained by this method until supplies can be accumulated for the more accurate determinations. Depleted uranium has been ordered for preparation of uranium monocarbide which can be used as one half of a diffusion couple for determination of the self-diffusion of uranium in uranium monocarbide. Fully enriched uranium metal foil will be sandwiched between two pieces of depleted carbide and diffusion annealed. Diffusion of the uranium-235 will be measured by sectioning, activating the samples obtained by neutron irradiation, and analyzing by gamma counting.

Tracer techniques for the determination of self-diffusion of carbon in uranium monocarbide do not appear practicable because of the health hazard involved in handling a carbide containing quantities of carbon-14. Other possibilities are being considered. The similarity of nitrogen and carbon and of UN and UC suggests that suitable data might be obtained by measuring the rates of interdiffusion of nitrogen and carbon from UN and UC couples.

Irradiation Effects in UC

A. E. Austin and C. M. Schwartz

A study of the effects of neutron irradiation and fission upon the physical properties and structure of uranium carbide has been initiated. The carbides are different from both metals and oxides, and thus represent potentially different fuel materials. They exhibit metallike properties of good electrical and thermal conductivity; on the other hand, they are hard and brittle and have a high melting temperature. Uranium monocarbide possesses a strong covalent type of bonding between uranium and carbon, but some free electrons are available for electrical and thermal conductivity. The higher carbides, U_2C_3 and UC_2, have strong carbon-carbon covalent bonding. Therefore, the carbides partake of some of the properties of both metals and refractory oxides.

The dependence of irradiation effects upon temperature may be expected to be similar to those of other refractory compounds. Thus, at low temperatures, the diffusion of fission products and interstitials may be quite slow, with crystal-lattice strain increasing until fracture occurs. However, at high temperatures there will be appreciable diffusion and possible relief of strain. Some previous observations have indicated that the monocarbide is ductile at very high temperatures. Therefore, some accumulation of fission gases into bubbles may be expected, leading to swelling. Information on the self-diffusion of uranium and carbon will be used in determining probable temperature dependence of fission-atom diffusion.

The immediate program will consist of an effort to evaluate various types of measurements as to their ability to yield the desired information on crystal strain, crystal fragmentation, mobility of fission products, nucleation and growth of gas bubbles, loss of fission gases, and physical expansion caused by neutron irradiation. Conventional hot-cell metallography and measurements of density, physical dimensions, and electrical resistivity can give part of the information. It is planned to obtain data on crystal strain and fragmentation by means of X-ray diffraction. The use of low angle X-ray scattering offers promise for detecting void of bubble formation. Light and electron microscopy will be of use in obtaining evidence of microcracking and larger voids or bubbles. Examination of presently available irradiated arc-cast material will be carried out.

G-1 and G-2

G. FATIGUE STUDIES OF INCONEL AND INOR-8

R. G. Carlson

Fatigue Studies of Inconel

The objectives of this program are to obtain basic fatigue information on Inconel and to establish quantitative relationships for Inconel among the variables of temperature, stress, strain, time, and cyclic frequency. The current phase of the program is concerned with measuring and recording strain in Inconel specimens under elevated-temperature fully alternating stress-cycling conditions.

During May, the stress-strain traces obtained from constant-temperature fatigue tests were analyzed to determine the relationships among the variables of interest. The information obtained is now being readied for inclusion in a topical report summarizing the work of the program.

Fatigue Studies of INOR-8

This program is concerned with the investigation of the temperature and frequency dependence of fatigue properties of INOR-8 alloy.

During May, the rotating-beam fatigue tests of INOR-8 were completed. The data obtained are now being analyzed. A frequency effect was found at 1500 F but not at 1100 F. At 1500 F, specimens cycled at 100 cpm had considerably shorter lives than specimens cycled at identical loads at 600 or 3000 cpm. However, the 600- and 3000-cpm data fell on the same stress-lifetime curves.

The results of this program will be contained in a topical report.

H. PHYSICAL RESEARCH

F. A. Rough

The AEC Division of Research is currently supporting four projects of physical research, two of which are reported below. The other two projects are reported on alternate months.

In the uranium-compound program, emphasis is presently on the preparation by sintering of UC bodies for study of fission-gas release. The results of the initial experiment on migration of hydrogen in zirconium and supporting data on diffusion coefficients of hydrogen in beta zirconium are reported.

Thermal Migration of Hydrogen in Zirconium

J. W. Droege, W. M. Albrecht, W. D. Goode,
and H. H. Krause

Preliminary experiments have been conducted with the apparatus designed for the study of the migration of hydrogen in zirconium under the influence of a thermal gradient. Additional diffusion coefficients for hydrogen in beta zirconium have been determined as a function of the hydrogen concentration.

Thermal Diffusion

Necessary preliminary information has been obtained from the operation of the first diffusion cell to be fabricated. This cell consists of a disk of zirconium hydride separated by stainless steel foil from two copper blocks, the whole being pressure bonded in a stainless steel cylinder. The copper blocks serve to maintain a constant temperature on each side of the zirconium hydride. Holes are drilled through the copper blocks and steel foil to give access to the faces of the zirconium hydride so that the hydrogen pressure can be measured on each side. Stainless steel thermocouple wells are inserted from each side to a point near the copper-steel interface so that the temperature of each face can be determined. A heater is provided to raise the temperature of either end of the cell, establishing a temperature gradient across the zirconium hydride. The diffusion cell is operated inside a tube furnace, through which argon or an argon-hydrogen mixture can be passed.

The equilibrium pressures of the zirconium-hydrogen system are known as a function of temperature and composition. Therefore, when a steady state has been reached, the concentration of hydrogen at the two faces can be found from the observed pressure and temperature. However, with this first cell, the bonding of the hydride to the stainless steel jacket was not gastight, so that the hydrogen could leak from one side of the cell to the other. Consequently, no valid thermal-diffusion data could be obtained from this cell. Nevertheless, the operational characteristics of the system could be determined. When the cell was heated by the tube furnace, but no thermal gradient was applied to the zirconium hydride, equilibrium pressures were obtained quickly. When a thermal gradient of 10 to 20 C was imposed on the hydride, a higher pressure was observed on the high-temperature side, but the steady state was approached slowly. The temperature in the copper blocks was found to be constant on the high side, but dropped off on the low side.

At the conclusion of these experiments the diffusion cell was sectioned and examined. The bonding between the stainless steel cylinder and the units inside was found to be unsatisfactory. As a result of the experience with the first cell, several modifications are anticipated for the future. Larger and more precisely drilled holes will be made in the copper blocks. The gradient heating system will be changed to obtain more uniform heating on the low-temperature side. The pressure-bonding conditions will also be changed to insure gastight seals around the zirconium hydride disk.

Diffusion Coefficients

Several diffusion coefficients for hydrogen in beta zirconium at 650 to 850 C using 0.063- and 0.065-cm-thick disks have been reported previously. These data indicated a possible decrease in the value of the diffusion coefficient with increasing hydrogen concentration. Further experiments have been made at 700 to 850 C using 0.085- and 0.089-cm-thick disks. The experimental conditions and diffusion coefficients are given in Table H-1. For comparison, the data previously reported are also given. On the basis of the results obtained thus far, there appears to be very little, if any, variation of the diffusion coefficient with hydrogen concentration in beta zirconium. Further permeation experiments using disks of other thicknesses will be made, and the temperature range of 400 to 700 C will also be examined.

TABLE H-1. SUMMARY OF RESULTS FROM DETERMINATION OF DIFFUSION COEFFICIENTS FOR HYDROGEN IN BETA ZIRCONIUM

Temperature, C	Average Hydrogen Concentration, a/o	Pressures on Membrane, mm of mercury		Diffusion Coefficient, 10^{-6} cm^2 per sec
		P_1	P_2	
650(a)	23	0.9	0.4	0.60
650(a)	32	1.5	0.7	0.47
700(a)	17.5	1.4	0.8	3.0
700	25.3	2.1	1.8	3.0
700(a)	31.5	4.5	1.4	2.2
750(a)	15.2	2.3	1.5	13
750	21.9	3.8	3.1	8.3
750(a)	29.2	7.9	4.5	9.6
800	9.5	2.0	1.3	41
800(a)	20.1	8.4	5.2	31
800	20.2	6.8	6.4	19
800(a)	27.8	14.5	10.4	21
850	19.4	13.3	12.1	75
850(a)	30.3	31.1	29.9	90

(a) Data previously reported.

H-3

Preparation and Properties of Refractory Uranium Compounds

A. B. Tripler, Jr., M. Jack Snyder,
and W. H. Duckworth

In this work, a correlation between fission-product release and certain physical properties of some sintered uranium compounds is being sought. Physical properties such as porosity, grain size, and bulk density are varied by varying initial particle size, compacting pressure, temperature of sintering, and time at temperature. The sintered compacts will be subjected to short-time low-temperature low-level irradiation in the Battelle Research Reactor, after which fission-product release will be studied.

Table H-2 shows the experimental design and the available property data for uranium monocarbide. Conditions 1, 2, 3, 4, 7, 11, and 15 are being investigated initially. They will provide a wide variation in physical properties for possible correlation with fission-product retention. Whether the remaining conditions will be investigated will depend on the outcome of the fission-product-retention studies on the initial specimens.

TABLE H-2. EXPERIMENTAL DESIGN AND RESULTS OF PREPARATION AND CHARACTERIZATION FOR UC

			Sintering Conditions		Properties of Material After Sintering		
Condition	Initial Particle Size, μ	Forming Pressure, 10^3 psi	Time, hr	Temperature, F	Average Grain Size, μ	Average Void Volume[(a)], per cent	Bulk Density, per cent of theoretical
1	6.2	50	1	1600	22.0	24.4	75.5
2	14.7	50	1	1600	43.0	33.5	73.3
3	6.2	100	1	1600	22.0	28.8	73.3
4	14.7	100	1	1600	43.0	29.3	75.9
5	6.2	50	3	1600	--	--	--
6	14.7	50	3	1600	--	--	--
7	6.2	100	3	1600	--	--	81.6
8	14.7	100	3	1600	--	--	--
9	6.2	50	1	1800	--	--	--
10	14.7	50	1	1800	--	--	--
11	6.2	100	1	1800	--	--	91.0
12	14.7	100	1	1800	--	--	--
13	6.2	50	3	1800	--	--	--
14	14.7	50	3	1800	--	--	--
15	6.2	100	3	1800	--	--	--
16	14.7	100	3	1800	--	--	--

(a) Estimated by metallographic means.

For Conditions 1, 2, 3, and 4 of Table H-2, the average grain size correlates with initial particle size. Forming pressure had no significant effect on sintered grain size. The percentage of voids is not entirely consistent with forming pressure nor with initial particle size. The bulk density, given in terms of per cent of theoretical density, shows little variation.

H-4

Cylindrical specimens of uranium monocarbide, sintered according to Conditions 7 and 11 in Table H-2, had cores composed of relatively small-grained material enclosed in rings having much larger grains. The grain boundaries associated with the outer larger-grained material appeared to be uranium rich. This phenomenon was observed in previous work and was reported in BMI-1313. It was believed at that time that the phenomenon was related to a deficiency of carbon in the starting material; however, in the current work a slight excess of carbon was observed by chemical analysis. The grain-size determinations and the estimation of percentage of voids for these latter specimens are not available at this writing. The higher bulk-density values may be due in part to the uranium-rich grain boundaries in the outer rim.

The preparation of the uranium nitride powder has been completed and the material is being processed into two portions with different particle sizes.

Attempts will be made to determine the conditions causing the large-grained rings in the UC specimens, and sintering experiments will begin with UN.

I. SOLID HOMOGENEOUS FUELED REACTORS

D. N. Sunderman and W. H. Goldthwaite

Research in support of the development and evaluation of spherical fueled-graphite fuel elements for the Pebble-Bed Reactor program is in progress at Battelle. Candidate fuel elements, 1-1/2-in. in diameter, prepared by several vendors and varying in composition and method of manufacture, are being evaluated by laboratory tests and irradiations designed to obtain information on their structural integrity and fission-product-retention characteristics.

LABORATORY EVALUATIONS OF FUELED-GRAPHITE SPHERES

J. F. Lynch, T. S. Elleman, W. C. Riley, and W. H. Duckworth

Fueled-graphite spheres are being evaluated with respect to selected thermal and mechanical properties. Other physical-property measurements are made as needed.

During May, two types of coated fueled-graphite spheres were tested to determine the concentration of uranium in the surface coatings. Measurement of alpha activity was used as a criterion of uranium concentration.

Admixture-type spheres with a SiC coating had alpha activities ranging from 25,000 to 140,000 alpha counts per min, which indicated surface uranium in amounts of 5 to 30 mg. The surface of the sphere was washed and scrubbed, causing a decrease of about 30 per cent in alpha activity. Further investigation of uranium content in the coating is being conducted.

The other type of spherical-fuel elements exhibited negligible alpha activities. These spheres have an unfuelded graphite shell 1/4 in. thick beneath the SiC-silicon coating and a fueled core 1 in. in diameter.

Coated spheres of both types were subjected to thermal-cycling tests. Spheres supported in a basket were alternately raised into a furnace and lowered into a cool zone below the furnace. All tests were conducted in an argon atmosphere. The cycle consisted of 20 min in a hot zone at about 2000 F and 15 min in a cool zone which lowered the temperature of test spheres to about 200 F. After three successive cycles, the test spheres were subjected to the hot-fluid test to indicate any failure of the surface coating. No failure was detected in either type of coated sphere after nine cycles.

I-2

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

D. N. Sunderman

The release of fission products from fueled-graphite spheres of various compositions is being studied both by postirradiation heat-treatment experiments (dynamic activation) and by in-pile sweep-gas experiments (dynamic irradiation). The dynamic-activation technique serves to screen materials of potential value as a reactor fuel. The most promising materials are advanced to the dynamic-irradiation experiments.

Dynamic-Activation Experiments

R. Lieberman, D. N. Sunderman, and M. Pobereskin

During May work began on a new series of postirradiation evaluations of prototype spheres. The program will differ from previous work in that several modifications of the experimental conditions of evaluation will be made. These include:

(1) The temperature lag in bringing the spheres to test temperatures will be reduced. This will be accomplished by induction heating.

(2) The spheres will be heated up to 2500 F.

(3) The radiation exposure of the test spheres will be increased to achieve higher sensitivity.

(4) The fission gas released will be continuously monitored by measuring the activity of the refrigerated charcoal trap during heating of the specimen.

Experimental work during May consisted of measurement of the temperature differences between the outside and center of a test sphere during rapid heating to operating temperatures of 1000 and 1500 F. A thermocouple was sealed into a hole drilled to the center of a graphite-UO_2 sphere. The outside temperature measurement controlled the heating cycles to duplicate previous experiments measuring fission-gas release. Helium was passed through the system at the rate of 3 liters per hr. The sphere in its quartz container was placed in a preheated furnace.

The results of the temperature measurements are shown in Tables I-1 and I-2.

TABLE I-1. TEMPERATURE MEASUREMENTS OF SURFACE AND CENTER OF GRAPHITE-UO_2 SPHERE HEATED TO 1000 F

Time After Startup, min	Temperature, F	
	Center	Outside
0	75	75
5	540	600
8	750	800
12	980	1000
13	1000	1000
14	1020	1000
18	1020	1000
27	1020	1000
27	Sphere removed from furnace	
29	820	520
30	700	380
37	340	160

TABLE I-2. TEMPERATURE MEASUREMENTS OF SURFACE AND CENTER OF GRAPHITE-UO_2 SPHERE HEATED TO 1500 F

Time After Startup, min	Temperature, F	
	Center	Outside
0	90	90
2	580	760
4	890	950
8	1190	1180
12	1320	1280
16	1370	1350
20	1410	1370
30	1500	1470
43	1520	1500
48	1520	1500
58	1520	1500
58	Sphere removed from furnace	
59	1310	850
61	870	550
63	680	390
68	380	210

Work was begun to identify long-lived nonvolatile radioisotopes present in a discarded gas-storage tank from Capsule SP-2.

I-4

Dynamic-Irradiation Experiments

D. B. Hamilton, G. E. Raines, and W. H. Goldthwaite

The irradiation of the second sweep-gas capsule (SP-3) was initiated on May 6, 1959, in the BRR. Estimated specimen-surface temperatures of 1300 to 1590 F have been maintained throughout full-power operation. The specimen loading for this experiment was as discussed in BMI-1340, two silicon carbide-coated spheres and two coated with a siliconized-silicon carbide. High activity levels in the carrier-gas line prevented sampling of the gas from the silicon carbide-coated specimen.

On May 8, 1959, a sample of the carrier gas was obtained from the siliconized-silicon carbide-coated specimens. The fractional release for two fission-gas isotopes is given in Table I-3.

TABLE I-3. FISSION-GAS-RELEASE DATA FOR SILICONIZED SILICON CARBIDE-COATED SPECIMENS

	Fractional Release[(a)]	
Isotope	At 2 Megawatts (May 8, 1959)	At 100 Kilowatts (May 18, 1959)
Kr^{85m}	2×10^{-7}	8×10^{-5}
Xe^{135}	3×10^{-8}	3×10^{-6}

(a) Fractional release is defined as the ratio of atoms released per second to atoms being produced per second.

After operation as a static capsule with no gas flow for 3 days, the activity level of the carrier-gas lines was too high to allow sampling even after an initial flush period. To reduce activity levels the capsule was removed from the reactor on May 15, 1959. A low-power run was made on May 18, at 100 kw with a specimen temperature of 175 F. A gas sample indicated an increase in fractional release by a factor of 100 to 400 over the previous sample, as shown in Table I-3.

Revision of the gas systems is under way in order that fractional release rates may be measured regardless of their magnitude.

Additional experiments are being planned; these include a high flux-high burnup static capsule, a high-temperature (~ 2000 F) static capsule to determine the extent of conversion of uranium dioxide to uranium carbides, and another sweep gas capsule with specimens having improved coatings. These irradiations will be conducted during the next fiscal year.

I-5 and I-6

POSTIRRADIATION EXAMINATIONS OF FUELED-GRAPHITE SPHERES

R. J. Burian and J. E. Gates

The postirradiation examinations planned for the fuel elements will include nondestructive examinations and destructive testing. The former tests include visual examination, dry-weight and physical-dimension measurements, and immersion in a hot-oil bath for detection of cracks when the specimens are coated. The destructive tests which will be performed will include compression tests (while the specimen is immersed in a hot-oil bath if the specimen is coated) and drop-weight impact tests using the hot-oil-bath technique to determine coating failure. Postirradiation metallography will be employed in some instances to study, for example, the conversion of UO_2 to the carbide.

J-1

J. PROBLEMS ASSOCIATED WITH THE RECOVERY OF SPENT REACTOR FUEL ELEMENTS

CORROSION STUDIES

C. L. Peterson, P. D. Miller, O. M. Stewart, J. D. Jackson
W. M. Stiegelmeyer, W. C. Baytos, T. E. Snoddy, and F. W. Fink

The evaluation of materials of construction for several processes for the recovery of spent nuclear reactor fuel elements is being conducted as a program of assistance to the Chemical Technology Division of ORNL. A topical report on the evaluation of materials for the hydrofluorinator of the Fluoride-Volatility process will be issued soon. Corrosion studies concerning the aqueous processes are drawing to a close. These will be summarized in another topical report also in preparation.

Two years of exposure of Type 347 stainless steel to chloride-contaminated Purex extraction systems has produced only negligible attack. Bench-scale dissolutions of Consolidated Edison fuel pins in a Ni-o-nel dissolver by a Sulfex-Thorex route and in a titanium dissolver by a Darex-Thorex operation have not resulted in serious corrosion problems. Metallographic studies of these dissolvers remain to be completed before the extent of localized attack can be determined.

The Darex Process

In the Darex process, fuel elements with stainless steel as a cladding or diluent are dissolved in dilute aqua regia. After chloride removal, the solution is adjusted and the uranium content recovered by solvent extraction.

The investigations of the Darex process have been terminated. At present, no other studies of this system are contemplated. The final results of a 2-year exposure of Type 347 stainless steel specimens to chloride-contaminated Purex acid systems are summarized below.

Chloride-Contaminated Purex Extraction Systems

Specimens of Type 347 stainless steel were exposed to Purex extraction solutions containing chloride contaminations such as might occur during extraction of uranium from Darex solutions. Two types of aqueous solutions were used; both were 2 M HNO_3. One was high in stainless steel ions and low in uranium and the other was high in uranium and low in stainless steel. The aqueous solution was mixed with an organic phase in the ratio of 2 to 3, the organic phase consisting of one part, by volume, of tributyl phosphate to two parts of Apco 125 (sulfuric acid-treated kerosene fraction). Chloride contaminations from 2 ppm to 1.00 w/o in the aqueous phase were studied in separate experiments.

Specimens in the form of disks, coupons stressed to 90 per cent of their yield strength, and coupons bent into the shape of horseshoes were exposed. These were submerged in the aqueous phase of the system contained in long, stoppered tubes. The tubes were kept at room temperature and, during the first few weeks, the specimens were removed daily while the two-phased system was intimately mixed by vigorous shaking.

When no attack developed during the first few months, the specimens were inspected at 6-month intervals. After 2 years of exposure, the study was discontinued. No sign of attack was observed on any of the specimens except for a slight brightening of the surfaces. The weight changes were insignificant. No cracking or other localized attack could be found even upon a metallographic study of the specimens from the solutions containing the highest chloride contaminations.

Under the conditions of these experiments, there is no indication that even large chloride contaminations will cause corrosion of Type 347 stainless steel Purex extraction-system equipment.

The Sulfex-Thorex Process

In the combination Sulfex-Thorex process, stainless-clad fuel elements of thoria may be dejacketed with boiling 6 M H_2SO_4. Following a wash step, the core is dissolved with a boiling solution which is 13 M HNO_3, 0.04 M F^- with added aluminum to mitigate corrosion.

Experiments With Ni-o-nel

A total exposure of 648 hr was completed for Ni-o-nel specimens exposed to boiling 3.5 M H_2SO_4 containing 50 g per liter of dissolved stainless steel. Solutions made by dissolving Type 304 stainless steel rod and others made by addition of iron, chromium, and nickel salts, in the appropriate proportions, were used. Some intergranular attack had developed by the end of this exposure, but the attack was not very severe in either type of solution, all corrosion rates ranging from 0.3 to 0.5 mil per month. No further exposure of these specimens is planned.

Bench Dissolution of Fuel Pins by the Sulfex-Thorex Process

A total of 21 Consolidated Edison fuel pins have been dissolved, one at a time, in the bench-scale Ni-o-nel dissolver by the Sulfex-Thorex route. Most of the difficulties concerned with passivation of the steel cladding and precipitation of salts in the decladding solution have been alleviated by altering the operating techniques. The dissolver does not show serious attack. Ni-o-nel coupons inserted in the dissolver show corrosion rates of about 2 mils per month. As mentioned in BMI-1340, incipient intergranular attack can be seen in the heat-affected areas around some of the weldments. After four more pins have been dissolved, the vessel will be sectioned and examined metallographically to determine the extent of penetration of localized attack.

J-3

The Darex-Thorex Process

The Darex-Thorex process is under consideration as an alternate to Sulfex decladding. Stainless steel cladding would be removed by boiling dilute aqua regia, while core dissolution would be carried out by the Thorex route. Titanium is fairly well established as the material of choice for the Darex dissolver, so the success of the combined Darex-Thorex operation hinges on containing the core-dissolution step in titanium.

Scouting Experiments With Titanium

Welded titanium specimens were exposed to boiling Thorex solutions containing various additions of aluminum or thorium for 1000 hr. Exposure to boiling 13 M HNO_3, 0.04 M F^- alone results in a general attack at a maximum rate of about 7 to 10 mils per month, which occurs in the vapor and which produces a smooth shiny surface. Addition of 0.04 M aluminum to this solution decreases the rate to 1.4 to 1.6 mils per month. A dark coating is formed on the specimens and no localized attack occurs. However, if larger quantities of aluminum are added (0.09, 0.15, or 0.20 M), or, if thorium is also present with the 0.04 M aluminum, as would occur as core dissolution progresses, the attack becomes localized. This occurs especially in the vapor phase and predominantly in the heat-affected areas around weldments.

A Darex decladding step may result in chloride contamination of the Thorex core dissolution. Excessive corrosion of titanium by nitric acid solutions containing both chloride and fluoride contaminations has been reported by other investigators. In order to study the effect of chloride carry-over, plain and welded titanium coupons have been exposed for 835 hr to boiling 13.0 M HNO_3, 0.040 M F^-, 0.10 M Al^{+3}, 100 ppm Cl^- solutions which are renewed weekly. While the corrosion rates, based on weight losses, have been very low (less than 0.3 mil per month), several of the specimens are pitted in isolated spots. Pitting occurs both upon liquid- and vapor-phase exposure and appears to be associated with stressed areas, either those arising from cold work or those associated with heat-affected areas. While this pitting is noticeable, it is not serious, as yet. Similar behavior was not observed in other studies with chloride absent. Exposure of these specimens is being continued.

Bench Dissolution of Fuel Pins by the Darex-Thorex Process

The dissolution of 25 Consolidated Edison fuel pins, one at a time, has been completed in a bench-scale titanium dissolver by a Darex-Thorex route. This has resulted in a total exposure of the vessel for 225 hr to the decladding and core-dissolution solutions. The vessel has been sawed in two and examined. In general, it looks very good. No attack is apparent on the submerged portions, but a localized attack has occurred around some of the weldments in the vapor. This has resulted in a pronounced etching of the weld beads and the heat-affected area surrounding them, but this etch does not seem to have penetrated very far. Coupon-type specimens submerged in the dissolver have gained a few milligrams in weight. Similar specimens inserted in the vapor have a corrosion rate of less than 0.1 mil per month. Presently, metallographic examination of the vessel is under way to determine the extent of penetration of the localized attack.

The Fluoride-Volatility Process

A topical report describing the corrosion studies associated with evaluating materials of construction for the hydrofluorinator of the Fluoride-Volatility process is being prepared. This report will include the results obtained in both the $NaF-ZrF_4$ and NaF-LiF systems. No other evaluations of these systems are planned during this contract period.

STUDY OF THE EFFECT OF IRRADIATION ON CLADDING- AND CORE-DISSOLUTION PROCESSES

R. A. Ewing, D. K. Dieterly, and M. Pobereskin

Preliminary assembly of the glass dissolution equipment for study of the dissolution of Consolidated Edison fuel pins by the Sulfex process was completed during May. Some modifications to reduce the over-all height appeared desirable for hot-cell operation; these are now being made.

Initial dissolution tests with "cold" prototype fuel pins are scheduled to begin after the apparatus is revised and reassembled. A number of prototype fuel pins, irradiated at ORNL, are now cooling; these are scheduled to be shipped to BMI during June. It is not yet certain whether it will be possible to initiate testing of these prior to July 1, 1959.

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

Irradiations of three capsules of uranium-5 w/o carbon specimens and subsequent hot-cell examinations have been essentially completed. These results up to burnups of 6400 MWD/T uranium are generally favorable and are summarized in this report. Additional irradiations up to 15,000 MWD/T uranium are under way. Routine progress on these irradiations is reported.

Irradiation of Uranium Monocarbide

D. Stahl and W. H. Goldthwaite

Four additional capsules of uranium monocarbide specimens are in various stages of irradiation at the MTR. Irradiations up to 15,000 MWD/T are planned as indicated in the schedule in Table K-1.

TABLE K-1. PRESENT IRRADIATION SCHEDULE FOR AI URANIUM MONOCARBIDE BMI-23 SERIES CAPSULES

Capsule(a)	Carbon Content, w/o	Target Uranium Burnup, MWD/T	Cycles Required at 1 x 10^{14} NV	MTR Position	Insertion Cycle	Insertion Date
23-3	5.0	10,000	12	A-28-NE	116	1-12-59
23-4	5.0	15,000	18	A-27-SE	115	12-22-58
23-5	4.6	5,000	6	A-30-NE	122	5-22-59
23-6	4.8	5,000	6	A-30-NE	126	9-21-59

(a) Capsules BMI-23-1 and BMI-23-2, containing 5.0 w/o carbon, were irradiated for one and six cycles and were discharged from the MTR at the end of Cycles 107 and 115, respectively.

Capsule BMI-23-5, containing uranium monocarbide (4.6 w/o carbon) specimens, was inserted into MTR Position A-30-NE during the shutdown for Cycle 122. The capsule is now operating with a central core temperature of about 1370 F in a thermal-neutron flux field estimated from heat-transfer considerations to be about 0.9 x 10^{14} nv; the normal increase in flux during a cycle may increase this temperature to the 1500 F design level.

During the first part of Cycle 122, a general temperature reduction was noted for Capsules BMI-23-3 and BMI-23-4, located in MTR Positions A-28-NE and A-27-SE, respectively. During Cycle 121, BMI-23-3 operated with a core temperature averaging about 1400 F, while for BMI-23-4 the average temperature was 1250 F. However, current averages are about 1300 F for BMI-23-3 and 1150 F for BMI-23-4.

Postirradiation Examination of Uranium Monocarbide

S. Alfant, A. W. Hare, F. A. Rough,
and R. F. Dickerson

A program is currently being conducted to determine the effects of irradiation on uranium monocarbide. To date, three experiments have been completed on capsules containing uranium-5.0 w/o carbon specimens. At this time, essentially all the experimental work has been completed on these capsules.

Table K-2, which contains the preirradiation and postirradiation data for specimens taken from Capsules BMI-23-1 and BMI-23-2, both irradiated in the MTR, illustrates the effect of irradiation on UC at different temperatures and burnups.

An examination of data obtained from specimens irradiated up to 6400 MWD/T indicates that UC behaves much better than uranium or uranium-alloy fuels operating at the same burnup levels and temperatures.

Preparation of UC Pins for Irradiation in the SRE

E. L. Foster and R. F. Dickerson

Specific techniques for the preparation of cast specimens of UC 0.6 in. in diameter have been established. The actual preparation and preirradiation examination of pins for irradiation at the SRE will begin as soon as the enriched uranium is received.

TABLE K-2. IRRADIATION DATA ON UC SPECIMENS FROM CAPSULES BMI-23-1 AND BMI-23-2

	Capsule BMI-23-1				Capsule BMI-23-2			
	Specimen AI-3 (Top)		Specimen AI-2 (Bottom)		Specimen AI-26 (Top)		Specimen AI-37 (Bottom)	
Dimensions								
Preirradiation, in.	0.3746		0.3746		0.3732		0.3737	
Postirradiation, in.	0.3760		0.3757		0.3769		0.3733	
Change, per cent	0.4		0.3		0.9		0.9	
Densities								
Preirradiation, g per cm^3	13.368		13.383		13.3439		13.428	
Postirradiation, g per cm^3	13.27 and 13.25[a]		13.0452		13.26 and 13.09[a]		13.19 and 13.18[a]	
Change, per cent	0.7 and 0.9[a]		2.5		0.6 and 2.0[a]		2.5 and 2.5[a]	
Temperature								
Minimum, F								
Surface	640		520		625		510	
Core	1100		700		920		710	
Maximum, F								
Surface	900		660		1075		895	
Core	1600		1100		1830		1475	
Mean, F								
Surface	770		540		916		701	
Core	1330				1376			
Burnup of Uranium	a/o	MWD/T	a/o	MWD/T	a/o	MWD/T	a/o	MWD/T
Estimated	0.196	1420	0.213	1590	0.625	4600	0.765	5700
Dosimetry	0.156	1160	0.195	1460	0.625	4600	0.850	6400
Chemical Analysis	Not available as yet				Not available as yet			
Fission-Gas Release (Krypton-85)								
Estimated Total Produced, cm^3	5.9 x 10-2				20.9 x 10^{-2}			
Estimated Released by Recoil, cm^3	2.5 x 10^{-5}				8.8 x 10^{-5}			
Actual Released, cm^3	2.4 x 10^{-5}				9.7 and 24.0 x 10^{-5}			

[a] Two pieces of specimen were checked.

L. TANTALUM AND TANTALUM-ALLOYS STUDIES

J. H. Stang

Studies of tantalum and tantalum alloys being conducted under the auspices of the LAMPRE development program at Los Alamos Scientific Laboratory are reported in this section. During May, the activity in the container-materials research consisted of the preparation of another series of binary tantalum-base alloys and initial metallurgical studies of their characteristics; as before, selected samples will be subjected to fuel-mixture corrosion exposures at LASL. In the study of irradiation damage to tantalum, encapsulated tensile specimens were inserted in the MTR in mid-May; the fabrication of control specimens for laboratory evaluations is well under way.

Development of Container Materials for LAMPRE Applications

D. C. Drennen, M. E. Langston, C. J. Slunder, and J. G. Dunleavy

The third series of binary tantalum-base alloy buttons has been prepared without difficulty by tungsten-electrode arc melting under a partial pressure of helium. This series comprises the following nominal additions:

0.025, 0.050, 0.100, 1.50, 3.00, and 6.00 w/o hafnium
0.025, 0.050, and 0.100 w/o titanium
0.025, 0.050, and 0.100 w/o thorium
3.00 w/o tungsten (zone refined)
0.025, 0.050, 0.100, and 0.200 w/o yttrium
0.025, 0.050, 0.100, 1.50, and 3.00 w/o zirconium

Radiographic inspection has shown that the arc-cast buttons are sound.

The buttons are being processed to 0.030-in. strip specimens which will be subjected to the usual sequence of metallurgical screening evaluations (fabricability, purity, microstructural characteristics, etc.). Samples of these materials having acceptable properties will be submitted to LASL for plutonium-alloy compatibility experiments.

Effect of Irradiation on Tantalum

J. A. DeMastry, F. R. Shober, F. A. Rough,
and R. F. Dickerson

Two capsules, each containing six sheet tantalum tensile specimens in an environment of NaK, were inserted in Position L-53 at the MTR during the shutdown for Cycle 122 (May 18). The capsules, designated as BMI-25-1 and BMI-25-2, are to be exposed to obtain approximately 7 x 10^{20} and 1.4 x 10^{21} nvt, respectively. These exposures have been calculated to be necessary for the transmutation of 1.5 and 3.0 w/o tantalum to tungsten. It is estimated on the basis of reactor-quoted fluxes and capsule flux-perturbation characteristics that the effective thermal flux in BMI-25-1 is in the range of 1.0 to 1-1/2 x 10^{14} nv; at this dosage, four reactor cycles will be required for its irradiation. In the case of BMI-25-2, the corresponding flux ranges from 1.5 to 2.0 x 10^{14} nv and seven cycles will be required.

Nickel-cobalt and pure nickel dosimeter wires are present in these capsules for determination of thermal- and fast-neutron flux. Position L-53 has a relatively high ratio of thermal to fast flux (reportedly, about 20 to 1). This will minimize fast-neutron damage in the tantalum specimens; however, the conversion of tantalum to tungsten will occur and property changes owing to this conversion can be evaluated.

Tantalum-1.5 and -3.0 w/o tungsten alloys have been arc melted and will be rolled to sheet. Tensile specimens are to be prepared from this sheet and will be used as laboratory controls to obtain baseline data for the evaluation of the irradiated material.

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

Pressure-bonding studies are continuing on the fabrication of UO_2-containing fuel plates. In previous work on this subject, difficulties have been experienced with lack of bonding when graphite-coated cores were used. It appeared that the graphite was being smeared on the surfaces to be bonded, giving incomplete bonding in those places. More successful methods of coating UO_2 to prevent reaction between it and Zircaloy-2 without interfering with bonding have been found. In these methods the UO_2 is coated with pyrolytic carbon or with chromium. In the most recent work using these barrier materials, excellent bonds have been produced.

Pressure Bonding of Zircaloy-2-Clad Fuel Elements Containing Compartmented Oxide Fuel Plates

S. J. Paprocki

A Zircaloy-2-clad compartmented flat-plate fuel element containing uranium dioxide cores is being considered for the PWR Core 2. The gas-pressure-bonding technique is being investigated as a possible method of fabricating these compartmented fuel plates. In this process the cores and Zircaloy strip components are assembled together to form the compartmented plate. This assembly is then fusion edge welded to form an envelope that is evacuated, sealed, and pressure bonded at 1550 F for 4 hr at 10,000 psi, using helium as a pressure medium. An alternate method, involving the bonding of the assembled fuel plate in a protective can, has also been investigated; however, the former method has been considered to be more suitable for this type of system. An additional heat treatment after bonding of 5 min at 1850 F in a salt bath has been utilized to improve grain growth along the Zircaloy-to-Zircaloy interface. The specimens now being evaluated indicate that this treatment may not be necessary.

Previous studies have indicated that graphite-coated uranium dioxide cores that were carefully buffed would not react with the Zircaloy during the pressure-bonding or subsequent heat-treatment cycle. It was difficult, however, to eliminate the possibility of mechanically contaminating the Zircaloy surfaces during assembly of the coated cores into the fuel-plate receptacle formed from the flexible strip components. Any of the Zircaloy surfaces that were contaminated with graphite would not bond metallurgically during the gas-pressure-bonding operation. It was shown that graphite in thickness of 200 A would prohibit bonding; consequently, it was desirable to investigate other methods of coating that did not require such careful handling. An investigation was initiated to evaluate pyrolitic carbon and chromium coatings. Studies with buffed-graphite coatings were continued to produce a base line for comparison of core-to-cladding reaction and bonding.

Coating of Uranium Dioxide Cores With Pyrolytic Carbon

E. H. Layer, J. D. Foti, and E. Wintucky

A large number of pyrolytic carbon coated cores have been prepared by the cracking of methane on uranium dioxide platelets at 1830 F. It has been determined that this coating is hard and adherent and is several microinches in thickness. The coatings can now be consistently produced with proper control of the cracking atmospheres. These coatings are also effective in preventing core-to-cladding reaction, as will be discussed in a later section.

Smear and scrape tests are being used for qualitative evaluation of coating adherence. These tests have indicated that, with proper control of the cracking atmosphere so that oxygen is not introduced, it is possible to produce a very adherent coating. Cores coated in this manner have been assembled without contamination of the Zircaloy surfaces.

Additional cores are being coated in this manner. A large number of pressure bonded elements incorporating those cores will be metallographically examined to determine the effectiveness of the coating.

Coating of Uranium Dioxide Cores by Vapor Deposition of Chromium

E. H. Layer, J. D. Foti, and E. Wintucky

The coating of uranium dioxide cores with thin coats of chromium by vapor deposition in a large batch operation is being investigated. Preliminary results indicate this is a feasible method of producing coatings of chromium of 15 to 100 μin. thick on uranium dioxide platelets. The coatings produced are firm and adherent and can be produced to a desired thickness. An upper limit has not been established for the thickness of chromium that can be deposited in this manner; however, thicker coatings formed by multiple-layer deposition are as firm and as adherent as single-layer coatings.

The coatings are applied by a two-step process in a rotating jig. Four sides are coated first and then, subsequent to rejigging, the remaining two sides are coated. A plot of the resistance measured across the face of a coated core has been plotted as a function of the thickness of the coat. This curve shows that a definite relationship exists between resistance and thickness; consequently, the resistance measurement has potential value as a method of control during production of coated cores for pressure bonding. Preliminary results indicate this measurement may be a quick method of estimating coating thickness and an indication of coating integrity.

A large number of cores will now be coated to determine the consistency of the coating in preventing reaction during pressure bonding. The economics of this process compared to other coating techniques that might be used on uranium dioxide cores will be investigated.

Chromium Coating of UO_2 Cores

N. D. Veigel, E. A. Beidler,
and J. M. Blocher, Jr.

Samples of UO_2 cores coated with 5-20 μ of chromium by hydrogen reduction of CrI_2 were submitted for pressure-bonding tests.

Pressure-Bonding Studies of Zircaloy-2-Clad PWR-Type Fuel Plates

S. J. Paprocki, E. S. Hodge,
D. C. Carmichael, and P. J. Gripshover

Investigations are being continued on the preparation of Zircaloy-clad flat-plate fuel elements containing compartmented uranium dioxide fuel by gas-pressure bonding. The use of barrier layers of buffed graphite, crystalline carbon, and vacuum-evaporated chromium on the cores to prevent core-to-cladding reaction is being studied. The elements being bonded for these studies are approximately 3.5 in. wide by 8 in. long by 0.150 in. thick, and consist of a 0.100-in.-thick receptacle plate and two 0.025-in.-thick cladding plates. Each of the elements contains 32 cores which have dimensions of 0.100 by 0.250 by 1.500 in. and is separated into 32 individual compartments by 0.035-in.-wide Zircaloy ribs. The compartmentalized receptacle plate is composed completely of strip components which have been surface finished by machine belt abrasion. After bonding, the elements are sectioned in half longitudinally; one section is evaluated as bonded and the other section is heat treated at 1850 F for 5 min in a salt bath prior to evaluation.

Nine elements were initially pressure bonded to evaluate barrier coatings of graphite and crystalline carbon on the cores and the effect of intentional void space. Six of these specimens contained cores with sprayed-and-buffed coatings of graphite in the amount of 5 mg of graphite per in.^{2} of core surface. Three of these six were fusion edge welded by techniques developed to produce an evacuated, gastight assembly, and three were assembled between graphite-coated stainless steel shims and Ti-Namel spacers into a protective container which was evacuated and sealed. Three of the initial nine elements contained cores which had been coated with crystalline carbon by the methane-decomposition process and were prepared by the edge-welding technique. The crystalline-carbon coatings were 1 to 5 μin. thick. Evaluation of these specimens has been based on metallographic examination, bend tests, and burst tests. Corrosion tests of purposely defected compartments are in progress. The results of the bend tests and burst tests revealed that the Zircaloy cladding of the bonded elements was ductile. Metallographic examination showed that both the crystalline-carbon and graphite coatings had prevented core-to-cladding reaction in the as-bonded specimens and in the specimens which were heat treated at 1850 F for 5 min. Examination of the Zircaloy-to-Zircaloy bonds disclosed contamination along the bond interface which prevented grain growth across the interface in some areas of both the as-bonded and the heat-treated specimens. Burst tests of compartments of these nine elements resulted, in some cases, in failure of the Zircaloy-to-Zircaloy bonds at high pressures, rather than rupture of the cladding. This evidence of weak bonds was associated with the observed contamination of the interfaces, which could have originated from several sources. The bonding surfaces may have been contaminated with graphite or carbon from the core coatings during assembly or handling prior to pressure bonding. Difficulty had been experienced during final sealing of this group of specimens and oxidation of the Zircaloy surfaces could have been caused by overheating. Also, during fabrication of the cladding components, contamination may have been introduced which was not removed during cleaning prior to assembly.

Detailed analyses were made of the effect of intentional void space contained in these nine elements. Two types of void space were included in the elements. Some elements were assembled using cores from which various controlled amounts of material had been ground from the corners to simulate chipped cores; others contained various amounts of void space at the ends of the longitudinal rows of cores to simulate poor dimensional control. The sizes of the simulated chips were 10, 25, 50, and 75 mils as a side of an equilateral triangle; the amounts of longitudinal void space were 10, 20, and 35 mils. Flow of the cladding plates into void areas was considerably greater in the edge-welded elements than in the elements bonded in containers, which had the benefit of the Ti-Namel spacers. However, both types of elements were satisfactory since only the defects of the largest size produced significant amounts of cladding depression and thinning, and in no case did flow produce thinning of the cladding to rupture. These results indicated that significantly less flow of the cladding occurred in this series of specimens which had Zircaloy ribs between each of the cores as compared to earlier gas-pressure-bonded specimens which did not have individually compartmentalized cores.

Three additional edge-welded elements containing cores with sprayed and buffed coatings of graphite and one similar element containing crystalline-carbon-coated cores were pressure bonded. Improved cleaning techniques for the Zircaloy components and an improved sealing procedure were incorporated in the preparation of these elements. The specimens were carefully handled in an attempt to prevent possible contamination of the bonds by graphite or carbon from the core coatings. Metallographic examination of the bonds in these as-bonded and heat-treated specimens revealed that there was no contamination of the interfaces and that grain growth had occurred across the original bond interface in all areas. Extensive burst tests of the compartments resulted in all cases in failure of the cladding at high pressures and deflections rather than failure of the bonds. Thus, the cause for the contaminated bonds in the earlier specimens was not specifically determined, but it was eliminated. As in the earlier specimens, core-cladding reaction had been minimized by the buffed-graphite coatings and crystalline-carbon coatings.

The use of chromium coatings on the UO_2 cores to prevent core-to-cladding reaction has been investigated by pressure bonding three elements incorporating vacuum-evaporated chromium coatings with thicknesses in several ranges from 12 to 100 μin. The improved cleaning and sealing techniques were used. Preliminary metallographic evaluation of portions of these specimens indicated no apparent reaction between the core and cladding in the as-bonded specimens and only slight reaction in the heat-treated specimens. No zones of uranium reaction products were observed at the interface. Very narrow bands of equiaxed grains in the prior-beta structure of the Zircaloy in the heat-treated specimens were observed adjacent to the barrier layer and core; these may be zones of a zirconium-chromium phase rich in oxygen. The corrosion behavior of the interface will be determined by tests of purposely defected compartments in 680 F water. Preliminary metallographic examination of the bonds in these specimens revealed no interface contamination and a high degree of grain growth across the original interface. Burst tests of the compartments of the elements with chromium-coated cores produced no bond failures; rupture of the cladding occurred in all cases at high pressures after large amounts of deflection.

Elements containing stainless steel cores were bonded at the same time as the second group of specimens with buffed-graphite and crystalline-carbon coatings and the group of specimens containing the chromium-coated cores. This was done so that the source of any contamination which might appear in the bonds could be established by comparison as being associated with the oxide core or its coatings, or with a variable in the bonding process. Metallographic examination of these specimens with the stainless steel cores showed that the bonds were uncontaminated and had good grain growth; however, the results of these control specimens were not directly applicable, since the specimens containing the various coatings also showed excellent, uncontaminated bonds.

Additional pressure-bonded elements will be prepared containing cores coated with crystalline carbon, buffed graphite, and evaporated chromium. The results of these studies should indicate the feasibility of using each of the coatings in pressure-bonded elements. The emphasis in this series of specimens will be on the as-bonded condition without the additional 1850 F heat treatment.

N-1

N. DEVELOPMENTS FOR THE MGCR

W. C. Riley and W. H. Goldthwaite

Research on reactor-core materials in support of the MGCR program is in progress at Battelle. Programs on the development and evaluation of UO_2 dispersions in BeO and UC and UC_2 dispersions in graphite, and on the cladding of UO_2 particles with BeO, are in progress. The evaluations include laboratory tests, examinations, and measurements, neutron-activation screening studies of comparative fission-gas-release characteristics, detailed neutron-activation studies of promising developments, in-pile fission-product-release studies, and static capsule irradiations to high burnups.

A study of the diffusion of fission products through fuel-element cladding materials has just been initiated.

Potential carbon-transport, oxidation, and carburization problems in an unclad-graphite-moderated helium-cooled reactor and the effect of radiation on these reactions are being studied by high-temperature capsule irradiations of graphite and metals in small thermal-convection loops.

FABRICATION AND CHARACTERIZATION OF FUEL MATERIALS

A. B. Tripler, Jr.

Current work on fabrication development of fuel materials is directed mainly toward increasing the density of the matrix. Samples containing 20 volume per cent UO_2 of 150-μ particle size dispersed in BeO have been fabricated up to 98 per cent of theoretical density. The effect of certain fabrication variables on strength and apparent density of UC-graphite and UC_2-graphite specimens are being studied. Fueled ceramic specimens have been prepared for high-exposure irradiation in the MTR and for neutron-activation testing.

UO_2 Dispersions in BeO

A. K. Smalley, W. C. Riley, and W. H. Duckworth

Densely sintered matrices of BeO containing uniformly dispersed UO_2 particles are being investigated for fuel-element core applications.

During May, additional experimental pellets containing 20 volume per cent of 30 per cent enriched UO_2 were prepared for MTR irradiation. The UO_2 was made from metal at Battelle by ignition and subsequent reduction. Bulk densities of 24 pellets ranged from 92.0 to 95.2 per cent of the calculated theoretical density of the mixture, and their average bulk density was 93.8 per cent of theoretical.

A laboratory process for preparing dense and uniform dispersion-type BeO-UO_2 pellets containing about 20 volume per cent of UO_2 has been developed. A description of the process follows:

(1) Davison ceramic-grade natural UO_2 was ball milled for 2 hr in alcohol, and then dried.

(2) The UO_2 powder was treated with 3 w/o of polyvinyl alcohol and a few per cent of tempering water. The mixture was worked almost to dryness on a laboratory roll mixer, and then dried completely at 120 F.

(3) The dried material was then crushed, ground, and screened, retaining the minus 80 plus 120-mesh (minus 177 plus 125 μ) fraction.

(4) The PVA-bonded UO_2 particles in the above size range were washed with C_2H_5OH, dried, and then incorporated into a slurry of Brush LOH-grade BeO, 3 w/o of beeswax (BeO basis), and CCl_4. The mixture was worked briefly on the roll mixer, using a 40-mil clearance between the rolls. It was then stripped from the rolls, granulated through a 20-mesh sieve while still moist with CCl_4, and allowed to dry.

(5) The granular mixture was compacted at 20,000 psi in a steel mold to form pellets measuring about 0.3 in. in diameter by 0.3 in. long.

(6) The pellets were sintered by heating for 1 hr at 2800 F in flowing hydrogen.

Pellets made as described above had bulk densities in the range of 97.5 to 98.5 per cent of theoretical. Metallographic examinations indicated that the UO_2 particles were dispersed uniformly throughout the BeO matrix and that the matrix was free of UO_2 dust.

Work in progress includes preparation of BeO-UO_2 pellets for neutron-activation testing. The over-all porosity of the pellets and the crystal size of the BeO phase will be varied, and the effects of these parameters on fission-gas release will be explored.

In future work, Brush UOX-grade BeO will be investigated as a matrix for UO_2 particles.

UC and UC_2 Dispersions in Graphite

W. A. Hedden, A. B. Tripler, Jr., W. C. Riley,
and W. H. Duckworth

The objective of this research is to develop improved graphite fuel-element cores containing dispersed UC or UC_2 in an amount equivalent to 20 volume per cent of UO_2.

Last month, it was reported that a study was in progress on the effect of variations in binder content. Specimens, consisting of small cylindrical compacts, were made from mixes of UC_2 particles, carbon or graphite filler, and varying amounts of coal-tar pitch. The compacts were formed using pressures of 8, 10, or 12 tsi and baked at a temperature of about 2300 F in an inert atmosphere.

Three different filler carbons were used: (1) AGOT graphite, minus 325 mesh; (2) AGOT graphite, 10 w/o minus 140 plus 200 mesh, 40 w/o minus 200 plus 325 mesh, and 50 w/o minus 325 mesh; (3) nongraphitized petroleum coke, 90 w/o minus 200 mesh. The fuel content was 18 to 20 volume per cent in all compacts.

The bulk densities and compressive strengths of the baked compacts indicated that the optimum binder content was about 16 w/o for the mixes containing either of the AGOT graphite fillers and about 14 w/o for the mixes containing the petroleum-coke filler.

In future work, it is planned to investigate minor additives for aiding densification.

Cladding of UO_2 Particles With BeO

A. K. Smalley, W. C. Riley, and W. H. Duckworth

A process for cladding UO_2 particles with a densely sintered shell of BeO is being developed.

Current work is directed toward preparation of spherical particles of fine-grained sinterable UO_2 in the size range of 100 to 500 μ. Spherical particles are desirable because the BeO claddings will be applied by a tumbling process, and uniform claddings of controlled thickness can be applied by this process only to spherical grains. Methods being investigated for preparing the particles are pelletizing, prilling, and spray drying.

Concurrently, experiments are in progress on developing low-porosity BeO cladding, using nonspherical UO_2 particles.

No results from the experimental work are available as yet.

Preliminary Characterization by Neutron Activation

P. Gluck and M. Pobereskin

Preliminary calculation of activities, soon after irradiation and after a cooling-off period, were performed for UC-graphite, UC_2-graphite, and BeO-UO_2 specimens.

An aluminum capsule designed to hold the specimens and dosimeter wires was constructed. A furnace in which individually encapsulated specimens will be annealed at a temperature of 1800 F was secured.

The materials required for specimen encapsulation are on hand. Specimens will be available in June at which time irradiation for screening purposes will be initiated.

STUDIES OF FISSION-GAS RELEASE FROM FUEL MATERIALS

P. Schall

A program on fission-gas release from MGCR candidate fuel materials was initiated during the past month. The program includes gas-release measurements by postirradiation heat treatment of low-burnup specimens (neutron-activation studies) and dynamic measurements made during irradiation (sweep-gas capsule studies) on materials judged to be promising in the neutron-activation studies.

Initial efforts have been devoted to design and procurement of the necessary equipment and to the development of experimental design and data-evaluation techniques.

Detailed Neutron-Activation Studies

P. Gluck, D. N. Sunderman, and M. Pobereskin

During this month calculations were performed leading to the design of an apparatus for the study of fission-gas release from neutron-activated specimens. As designed, the system will be flexible; it will permit heat treatment of irradiated fuel specimens at controlled temperatures up to 2800 F in a rhodium-wire-wound furnace. Should it become desirable, later in the program, to study gas release from specimens to temperatures above 3000 F, the furnace could be readily replaced by an induction-heated section. A closely controlled flow of purified helium carrier gas over a range of 1 to 100 liters per hr will be available. Because of the relatively high moisture and oxygen contents in the available helium, a purification train was designed in which the oxygen and moisture will be reduced to levels below 1 ppm. The impurity level will be monitored continuously.

At present it is expected that the fission gases will be monitored continuously using a proportional flow counter which will be designed next month. In conjunction with the continuous monitor, traps will be used to collect the fission products. The traps are so designed that they fit into the well of a scintillation counter, are easy to fill with activated charcoal, and hold a good vacuum. An added feature of the system is that traps can be changed and degassed while the system is operating. Preliminary experiments on this system are expected to be initiated in the latter part of June or early in July.

N-5

In-Pile Studies

N. E. Miller, P. Gluck, and G. E. Raines

The objective of these studies is to determine the fission-gas release from prospective fuel-element specimens during irradiation under conditions of surface temperatures and heat fluxes comparable to those expected in service. The unclad fuel specimen (0.22 in. in OD and up to 2 in. long) will be contained, with minimum clearances, in a cylinder within an irradiation capsule. During irradiation, helium will flow through the fuel-containing cylinder to carry the released fission gas out to an analytical train for elemental determinations. From preliminary design calculations it appears feasible to operate the fuel over a range of surface heat fluxes from 150,000 to 400,000 Btu/(ft^2)(hr) and surface temperatures from 1500 to 2000 F when irradiated in the BRR core.

The major effort on the program thus far has been directed toward the design of the sweep-gas system which can operate continuously with minimum hazards and minimum personnel time. The major items for this system have been ordered and should be delivered in June.

The design of the gas-sampling system associated with the in-pile apparatus is largely completed. Components for this system were ordered and are available for assembly. This apparatus will be assembled simultaneously with the out-of-pile equipment and should be completed in June. It is expected to have the first sweep-gas capsule ready for irradiation by October 1.

Experimental Design and Evaluation

A. E. Mace and S. D. Beck

A study to develop an experimental design for the in-pile gas-release experiments is in progress. The objective is to develop a design to determine the effect of surface heat flux and temperature, in the range indicated in the previous section, on gas release.

To facilitate separation of real differences due to variation of these parameters from those attributable to experimental error it is desirable to develop a simple operational model based on consideration of the expected effect of each of the variables.

The model then serves as the basis for establishing the spacing of the levels for each of the variables and the sequence in which experiments, under various combinations of the variables, are to be conducted.

The general plan for the experimental design has been outlined and detailed development of the design will be made as soon as details of the experimental apparatus are decided upon.

N-6

HIGH-BURNUP IRRADIATION EFFECTS IN FUEL MATERIALS

J. E. Gates, C. V. Weaver, and R. F. Dickerson

The irradiation of a capsule containing compacts of (1) UO_2 in a BeO matrix, (2) UC in a graphite matrix, and (3) UC_2 in a graphite matrix is continuing in the BRR. The objective of the irradiation is to study the stability of these fueled ceramics under conditions simulating those in the MGCR.

Three pairs of specimens are included in the capsule, which is equipped with thermocouples and electrical heaters. A specimen consists of four pellets of a given type of fueled ceramic in a stainless steel tube. The capsule has been operating satisfactorily, with four specimens maintained at surface temperatures slightly over 1500 F, one between 1400 and 1500 F, and one at about 1300 F. The expected burnup at the time of discharge from the reactor (June 1, 1959) is 1.0 to 1.5 per cent of the uranium-235. The postirradiation examination will be initiated early in June and should be completed by June 30.

Specimens containing compacts of the various fuel materials are now being fabricated for loading into the next three irradiation capsules. These capsules will be irradiated in the MTR under conditions of temperature and heat flux similar to those of the BRR capsule. It is planned to irradiate one capsule to a burnup of 12 per cent of the uranium-235, and two to a burnup of 20 per cent of the uranium-235. It is presently planned to ship these three capsules to the MTR on June 20.

DIFFUSION OF FISSION PRODUCTS IN CLADDING MATERIALS

S. G. Epstein, A. A. Bauer, and R. F. Dickerson

A program to measure the diffusion of fission products in cladding materials has been initiated. The cladding being considered in this project is "A" Nickel.

The investigation will consist of impregnating foils of "A" Nickel with fission products by irradiation recoil from uranium or with rare gases by cathodic bombardment. Two foils will be bonded together at their exposed faces, and then the couples will be heated to promote diffusion. It is planned to analyze for rare gases released from the couples. Analyses for other fission products will be made by machining layers from the recoil-impregnated foils and analyzing these layers by radiochemical techniques. Diffusion coefficients will be calculated for several elements.

A preliminary study is being conducted to determine whether cathodic bombardment is effective in introducing argon into the metal lattice, and to determine if the gas precipitates as bubbles. Two Type 304 stainless steel plates were bombarded for 2 hr with argon at a 200-μ pressure and across an applied potential of 450 v. They were then bonded together at their exposed surfaces under a pressure of about 35,000 psi at a temperature of about 500 C. The couple was heated in a vacuum furnace to 800 C and held at temperature for 2 hr to allow gaseous diffusion. Examination under an electron microscope of a section of the couple near the interface indicated the presence of minute pits, approximately 1000 A in diameter, which are believed to be argon bubbles. To substantiate the fact that the argon actually entered the steel lattice, an "A" Nickel specimen will be bombarded with argon and, after swabbing the surface to remove adsorbed argon, will be either heated in a capsule or chemically dissolved. Any argon present in the nickel lattice will be driven off and detected by a mass spectrograph.

CARBON-TRANSPORT CORROSION STUDIES

N. E. Miller and W. S. Diethorn

A study is being made of potential carbon-transport, metal-oxidation, and metal-carburization problems which may be associated with an unclad-graphite-moderated helium-cooled reactor system.

The GA2-2 capsule containing the four quartz tori (described in previous reports) was irradiated in the BRR core for one cycle, 240 hr at 2 megawatts. During this time the specimens in the hot and cold legs of each torus were at 1500 ± 10 F and 960 ± 10 F, respectively. The examination of the specimens is scheduled to begin in the hot cell in June.

The components for two similar capsules are being made. These will be assembled and irradiated in the next fiscal year.

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor Flow Studies

L. J. Flanigan and H. R. Hazard

Air-flow studies in a quarter-scale model of the Partially-Enriched Gas-Cooled Power Reactor are being conducted at Battelle to provide design data for the prototype. Previously reported work includes construction of the plastic-model parts, the model core, and the parts for the test setup.

In May, assembly of the model core and work on the test setup, at Battelle, were completed. Fabrication of the plastic-model parts by a vendor was continued. As was reported last month, delivery of the parts was delayed. Delivery to Battelle is now scheduled for early in June, 1959.

In June, the model will be assembled and flow studies started. It is anticipated that tests can begin a few days after delivery of the plastic-model parts.

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

The work described in this section is being performed to assist Alco Products in the development of fuel, absorber, and suppressor materials for the SM-2.

Irradiation-test specimens have been prepared for seven capsules scheduled for insertion into the ETR. It is planned to irradiate the capsules in an estimated thermal-neutron flux of 3×10^{14} nv to an average total uranium burnup of 37 per cent and a peak burnup of 77 per cent. The capsules are instrumented with dosimeters, thermocouples, and auxiliary heaters.

Development of Fuel Materials

S. J. Paprocki, D. L. Keller, G. W. Cunningham, J. B. Fox,
D. E. Lozier, W. M. Pardue, J. M. Fackelmann, and D. L. Chase

Techniques are being investigated for use in the fabrication of fuel elements containing cores of 26 w/o UO_2 and 1.1 w/o ZrB_2 dispersed in a prealloyed Type 347 stainless matrix. As an alternate procedure, plates containing the boron in the form of NbB_2 are also being developed. Although the preparation of irradiation specimens is being emphasized at the present time, studies are also being conducted to (1) develop fabrication techniques for preparing full-size elements, (2) establish specifications for ZrB_2, (3) establish reproducible analytical techniques for boron, and (4) determine physical and mechanical properties of fabricated fuel sheet.

SM-2 irradiation specimens have been prepared for the first seven capsule loadings. Table P-1 identifies each type of specimen as to type of fuel, poison, and matrix being used, and a listing of specimens to be contained in each capsule is shown in Table P-2. The detailed fabrication procedures for the irradiation have been previously reported with the exception of the procedure used for specimens with an elemental matrix. These specimens are fabricated by the method listed in Table P-3.

The specimens are fabricated in groups of five of each type. Two are placed in capsules for irradiation, the remaining three are used for metallographic examination and boron analyses. After the packs are cold rolled and annealed, the plates are radiographed and the specimens are marked and sheared oversize. A second radiograph is made and used as a guide to file the specimens to final dimensions of 0.562 + 0.013, - 0.000 by 1.506 + 0.010 in. A 0.031-in.-diameter hole is then drilled in the top of the specimens equidistant from either edge and 0.054 in. from the top, and at the bottom 0.025-in.-diameter holes are drilled 0.10 in. from either edge and 0.018 in. from the bottom. File marks for postirradiation identification of the specimens are then made on the edge of the specimens. At this point, each specimen is ready to be leak tested by boiling for three 1/2-hr periods in solutions of 50-50 nitric acid and water. Each solution is analyzed for uranium content. An increase in uranium content in the second and third pickles or an excessive amount in all three pickles indicates a faulty specimen.

TABLE P-1. SM-2 IRRADIATION SPECIMENS

Specimen Type	Fuel	Poison(a)	Matrix	Remarks
2	26 w/o hydrate UO_2	0.12 w/o B_4C	Prealloyed	--
3	26 w/o hydrate UO_2	None	Prealloyed	--
4	26 w/o spherical UO_2	None	Prealloyed	--
5	26 w/o hydrate UO_2	1.1 w/o ZrB_2	Prealloyed	--
6	26 w/o spherical UO_2	1.1 w/o ZrB_2	Prealloyed	--
7	26 w/o spherical UO_2	0.5 w/o ZrB_2	Prealloyed	Boron content, one-half loading
8	26 w/o spherical UO_2	1.1 w/o ZrB_2	Prealloyed	Double-size specimens containing Eu_2O_3 suppressors
9	26 w/o hydrate UO_2	1.3 w/o NbB_2	Prealloyed	--
10	26 w/o spherical UO_2	1.1 w/o ZrB_2	Prealloyed	--
11	26 w/o hydrate UO_2	1.3 w/o NbB_2	Prealloyed	--
12	26 w/o spherical UO_2	1.1 w/o ZrB_2	Prealloyed	Green compact hot rolled directly
13	26 w/o spherical UO_2	1.3 w/o NbB_2	Prealloyed	--
15	34 w/o UN	2.0 w/o ZrB_2	Elemental(b)	UN loading equivalent to 38.5 w/o UO_2
16	38.5 w/o hydrate UO_2	2.0 w/o ZrB_2	Elemental(b)	--
17	36.5 w/o spherical UO_2	2.0 w/o ZrB_2	Elemental(b)	--

(a) Actual w/o poison varies according to boron analysis of powder used.

(b) Dependent upon appearance of UO_2 particles, prealloyed-matrix specimens may be substituted.

TABLE P-2. SPECIMEN CONTENT OF CAPSULES 1 THROUGH 7(a)

Capsule 1	Capsule 2	Capsule 3	Capsule 4	Capsule 5	Capsule 6	Capsule 7
3-2	2-1	2-0	10-0	10-5	5-0	5-5
4-4	7-2	7-1	11-1	11-0	6-1	6-0
5-5	12-0	12-3	13-2	13-1	7-2	7-1
6-3	3-4	4-5	15-3	15-2	9-3	9-2
8-1	8-5	5-2	16-4	16-3	12-4	12-3
		6-4	17-5	17-4	13-5	13-4

(a) The first number indicates the specimen type as listed in Table P-1. The second number indicates the number of identification notches in the specimens.

TABLE P-3. FABRICATION OF IRRADIATION SPECIMENS CONTAINING BORON-COMPOUND DISPERSIONS IN ELEMENTAL MATRICES

A. Preparation of core compacts

1. Screen and blend powders
   - (a) Screen minus 325-mesh metal powders to prepare mix of 18 w/o Lunex chromium, 9 w/o carbonyl nickel, and 73 w/o electrolytic iron (low oxygen)
   - (b) Blend dry powders 1 hr in V-type mixer
   - (c) Add fuel (38.4 w/o UO_2 or 34.1 w/o UN, fully enriched) minus 100 plus 200-mesh powder and blend 1 hr
   - (d) Add 1/2 w/o methyl alcohol-camphor solution and blend 1 hr
2. Cold press at 50 tsi
3. Sinter for 2 hr at 2000 F in vacuum
4. Coin at 50 tsi

B. Preparation of billets

1. Machine picture frames to size and shear cover plates from 0.040-in. sheet
2. Clean all frames and cover plates
   - (a) Vapor degrease
   - (b) Scrub with steel wool in 180 F Alconox-water solution
   - (c) Rinse in 180 F water
   - (d) Rinse in cold water
   - (e) Repeat Steps (c) and (d) and pat dry with paper towels
3. Insert cores and evacuation stem and weld the assembled billets in an argon-filled dry box
4. Leak test packs
5. Evacuate at 500 F
6. Forge the stem closed

C. Roll

1. Hot roll at 2000 F from hydrogen muffle
   - (a) Preheat 45 min at 2000 F
   - (b) Reduce 40 per cent in thickness on the first pass and 20 per cent in thickness on all remaining passes
   - (c) Reheat between all passes
   - (d) Flip and reverse after each pass

TABLE P-3. (Continued)

2. Anneal 2 hr at 2300 F after hot rolling

3. Pickle to remove scale

   (a) Immerse for 5 min in a 180 F bath of 2 parts HF, 10 parts HNO_3, and 88 parts water

   (b) Rinse thoroughly in water and dry

4. Cold reduce 15 per cent in thickness to give a total reduction of 7.5 to 1 in thickness

5. Anneal 1 hr at 2050 F in hydrogen and cool in cold zone

A final radiograph and identification photographs complete the fabrication and identification of the specimens. No specimens to date have failed the leak test.

The dimensions previously stated are the same for all specimens with the exception of Type 8. These specimens are 1 in. longer since they contain a 2-in. core; one-half of which is the reference fuel core material and the other half a suppressor-material dispersion of 17 w/o Eu_2O_3 in an elemental stainless steel matrix.

In the listing of specimens, the only cores which contain elemental matrices are those with high fuel loadings. The elemental matrix was chosen in this case because it was believed that an elemental matrix would be necessary to prevent excessive stringering and fracturing of UO_2 particles. However, it appears that, if spherical UO_2 is used, it may be possible to fabricate high-loading plates with prealloyed matrices and maintain good integrity and distribution of the UO_2. Also, from a production standpoint, it would be advantageous to use prealloyed powders since, as shown in Table P-3, careful temperature control is required to place nickel in solution without reacting it with ZrB_2 and to place the chromium in solution without reacting the ZrB_2 with nickel-rich areas.

Specimens have been prepared to determine the effect of carbon and silicon on the stability of ZrB_2 in a stainless matrix but these have not yet been fully evaluated. However, examination of specimens consisting of 1 w/o ZrB_2 dispersed in Type 318 prealloyed powders with less than 1 and up to 2.5 w/o silicon indicates that in the low-silicon powders the ZrB_2 is completely oxidized but that in the high-silicon powders, while little or no oxidation occurs, an unidentified precipitate can be detected in the matrix. Chemical analyses for boron have not been completed for these specimens.

In the fabrication studies, transverse tensile tests (pulled in the thickness direction) are being used as an aid in evaluating process variables. The strength of the core after the first hot pass has been determined for plates rolled at 2200 F, and the results are listed in Table P-4. Similar tests will be run on plates with the same variation in the first pass but with identical fabrication to the final size. In the specimens listed in Table P-4, all fractures occurred in the core, indicating that the core-to-cladding bond strength is quite high after the first pass. Generally, specimens made with the same materials and rolled to an 8-to-1 reduction, including 20 per cent cold work, do not have transverse tensile strengths higher than 18,000 psi. Specimens are also being prepared to show the effect of total reduction and total cold work on the core strength.

Techniques for boron analysis are being evaluated and the reliability of Battelle results are being established. As shown in Tables P-5 and P-6, the analytical procedures established at Battelle yield consistent results; however, discrepancies of analyses obtained at Lucius Pilkin and Battelle for identical specimens have not been resolved.

Table P-6 shows results obtained on synthetic standards carried through the procedure with various groups of samples. A wide range of boron has been measured – from 50 μg to approximately 13 mg. The deviation of the results from the amount of boron added are recorded in terms of per cent of the amount present. The average deviation was then calculated by adding these and dividing by the number of standards. Of the 22 results reported, only 4 deviations are greater than ±5 per cent, and 13 results show a negative deviation while 9 results show a positive deviation.

TABLE P-4. RESULTS OF TRANSVERSE TENSILE TESTS ON FUEL PLATES[a] AFTER A SINGLE REDUCTION BY ROLLING AT 2200 F TO A CORE THICKNESS OF 100 MILS

Reduction First Pass, per cent	Total Reduction, ratio	Transverse Tensile Strength, psi
10	1.1:1	23,200
10	1.1:1	21,400
20	1.3:1	22,400
20	1.3:1	21,900
30	1.4:1	24,800
40	1.7:1	26,500
40	1.7:1	27,700
50	2.0:1	28,300
50	2.0:1	21,700

(a) Cladding: Type 347 stainless steel.
Core: 1.2 w/o minus 200 plus 270-mesh ZrB_2, 28 w/o minus 100 plus 200-mesh high-fired UO_2, balance minus 325-mesh prealloyed Type 347 stainless powder.

TABLE P-5. COMPARISON OF MICRO-TITRATION AND COLORIMETRIC BORON RESULTS

Analytical Laboratory Number	Specimen	Boron Content, w/o By Titration	 By Colorimetry
60876	ZrUP-2	0.087	0.075
60877	MoUP-2	0.093	0.086
61306	B-1068	0.015	0.012
61307	B-1069	0.157	0.142
61308	B-1070	0.102	0.095
61309	B-1071	0.167	0.154
61310	B-1072	0.134	0.130
61311	B-1073	0.153	0.151
61312	B-1074	0.127	0.123
61313	B-1075	0.166	0.145
61382	B-1090	0.196	0.175
Master alloy (Oak Ridge)		0.269	0.256
61442	B-1117	0.440	0.460
61796	B-1145	-0.213	0.191
61797	B-1146	0.225	0.217
61798	B-1147	0.113	0.103
61799	B-1148	0.411	0.416
61800	B-1149	0.367	0.375
61801	B-1150	0.362	0.406
61241	B-1054	0.184	0.191
61242	B-1055	0.164	0.163

TABLE P-6. RESULTS OBTAINED ON SYNTHETIC STANDARDS USING THE MICRO-TITRATION METHOD

Description	Boron Analysis, mg	Per Cent Deviation
Blank + 50 μg boron	0.0516	+3.2
2 g 101d[(a)] + 4.95 mg boron	5.03	+1.6
Blank + 4.03 mg boron	4.23	+5.0
3 g 101d[(a)] + 1 g UO_2 + 5.41 mg boron	4.75	-12.2
3 g 101d[(a)] + 1 g UO_2 + 12.90 mg boron	12.64	-2.0
3 g 101d[(a)] + 1 g UO_2 + 12.90 mg boron	13.25	+2.7
3 g 101d[(a)] + 1 g UO_2 + 12.90 mg boron	12.70	-1.5
Blank + 100 μg boron	0.1021	+2.1
Blank + 100 μg boron	0.0933	-6.7
Blank + 100 μg boron	0.0972	-2.8
Blank + 100 μg boron	0.0900	-10.0
Blank + 50 μg boron	0.0516	+3.2
Blank + 50 μg boron	0.0522	+4.4
Blank + 50 μg boron	0.0513	+2.6
2 g 101d[(a)] + 4.95 mg boron	4.86	-1.8
2 g 101d[(a)] + 4.03 mg boron	4.01	-0.5
Blank + 50 μg boron	0.0497	-0.6
Blank + 100 μg boron	0.0934	-6.6
2 g 101d[(a)] + 5.78 mg boron	5.74	-0.7
5 g 101d[(a)] + 15.23 mg boron	15.5	+1.8
Blank + 100 μg boron	0.0980	-2.0
2 g 101d[(a)] + 5.74 mg boron	5.57	-3.0
	Average	3.1

(a) 101d is a Bureau of Standards sample of stainless steel.

P-8

Encapsulation Studies

A. K. Hopkins, W. E. Murr, and J. H. Stang

Early in May, two SM-2 program irradiation capsules of the original group of seven intended for irradiation at ETR were assembled. These two were to be the first of five high-burnup (to 77 per cent total burnup) capsules with the remaining two scheduled to be average burnup (to 37 per cent) systems. Following the assembly of the initial two capsules, the supplementary sheathed resistance wire (Kanthal and Nichrome) heaters incorporated into them were subjected to laboratory tests at full power (2 kw per heater). In these tests, certain overheating difficulties were found in the nickel lead-wire sections of the individual heaters. These difficulties made it necessary to unload the capsules and study the heat-transfer problems involved. In the ensuing period, several steps were taken to improve the situation; the primary improvements consisted of a relatively simple chill-block scheme for conducting heat from the leads directly to the water-contacting outer shell of the capsule and a better copper-to-nickel lead-wire junction. At the end of the month, the over-all revised design of lead-wire-zone components was completed and proved to be satisfactory in bench tests.

Near the end of May, official information was received from the ETR that, because of loop-facility construction and checkouts, the reactor full-power on-stream time during the remainder of this calendar year, will be only a fraction of the normal on-stream time. Since this low duty will prevent the accumulation of fuel burnup at the desired rate; namely, accumulation of 77 per cent burnup by midspring, 1960, consideration is presently being given to possibilities of conducting irradiations at a reactor other than the ETR.

Q. GAS-COOLED REACTOR PROGRAM

D. L. Keller

Studies for Aerojet-General Nucleonics (AGN) directed toward the development of compact gas-cooled reactors are reported in this section. The activities on the various tasks are reported under "Materials Development Program" and "In-Pile-Loop Program'. During the latter part of May criticality studies were initiated on an ML-1 core mock-up. Progress on these studies will be reported next month.

MATERIALS DEVELOPMENT PROGRAM

Two capsules (BMI-27-1 and BMI-27-2), containing solid and annularly loaded UO_2 pins clad in Inconel, are being irradiated at the MTR. Also, dispersion elements of UO_2 and UN in stainless steel are being irradiated in a single capsule at the MTR. Progress on these irradiations is reported.

A fourth capsule is being fabricated for irradiating graphite-UO_2 fuel pins. Capsule-design studies were recently begun on conducting a multicapsule irradiation program at the BRR. Initial results of this later program will be reported next month.

Approximately 60 per cent of the 6000 enriched UO_2 pellets needed for two in-pile loop assemblies and a single flux-plot assembly have been prepared.

Effects of Irradiation

J. H. Saling, J. E. Gates, and R. F. Dickerson

The study of the radiation stability of fuel-element materials for compact gas-cooled reactors includes (1) the evaluation of encapsulated stainless steel-UO_2 specimens, (2) the evaluation of encapsulated fueled-graphite specimens, and (3) the evaluation of in-pile-loop subassemblies. Several irradiation experiments have been completed from each part of the program and the results discussed in earlier reports. Several irradiations are still in progress.

The results of radiochemical burnup analysis on a section which was removed from the axial center of Fuel Plate AI (12-o'clock position) from the GCRE-Mark-1-1-T in-pile-loop subassembly have been completed. The analysis indicated a burnup of 0.6 a/o uranium for this section. This completes the evaluation of fuel materials from this program until additional irradiations are completed.

Q-2

Encapsulation Studies

J. H. Stang, J. C. Smith, P. B. Shumaker, and C. V. Weaver

Capsule BMI-28-1, containing four clad flat-plate specimens (three fueled with 28 w/o UN dispersed in stainless steel and one with 30 w/o UO_2 in stainless; fully enriched uranium in each case) has been irradiated since April 10 at the MTR in a quoted unperturbed thermal-neutron flux peaking at 0.9 x 10^{14} nv (A-40-NE position). Specimen-surface temperatures have been in the 1700 to 1750 F range during the irradiation. According to present plans, the capsule will be discharged June 29 (Cycle 124 shutdown); estimates of fission heat generation from thermocouple data indicate that the burnup at discharge will be approximately 9.6 a/o uranium-235.

Irradiation of the two capsules designated as BMI-27-1 and BMI-27-2 continued at the MTR during May. Each of these capsules contains six Inconel-clad UO_2-fueled (35 per cent enriched) pin-type specimens designed to operate at a nominal cladding temperature of 1700 F. As pointed out in previous reports, Capsule BMI-27-2 was inserted in the A-30-SE position during Cycle 119 shutdown and subsequently operated with the bottom two specimens residing in the 1650 to 1550 F range (surface temperature), the middle two in the 1100 to 1250 F range, and the top two in the 1200 to 1300 F range. During Cycle 122 shutdown, this capsule was lowered 3 in. in the irradiation space in an attempt to equalize the temperatures of the top and bottom specimens. After this relocation, the specimen zone was located between 18 and 32 in. below the lattice. During the startup, an unexpectedly high thermocouple temperature of 1735 F was observed at three-fourths full reactor power and reactor personnel subsequently raised the capsule 11 in. before proceeding to full-power operation; specimen surface temperatures in this adjusted location are ranging from an average of about 1550 F for the bottom pair to about 900 F for the top pair. At present, the high temperature at partial power cannot be accounted for, and the only move now scheduled for the capsule is a return to its initial axial location. Meanwhile, further analyses of the capsule heat-transfer situation are being made to determine the desirability of attempting the 3-in. lowering again.

Capsule BMI-27-1 has been irradiated in the A-7-NW position since Cycle 120. As explained in BMI-1340, it was necessary to locate the specimen zone in this capsule above the flux peak to prevent specimen temperatures higher than the 1700 F originally specified. This positioning has resulted in a steep axial temperature profile where the bottom specimens are in the 1600 to 1400 F range and the top specimens in the 600 to 800 F range. In a sequel to this situation, studies of the feasibility of moving this capsule down in the facility to level out the temperature and burnup characteristics have been carried out. A tentative conclusion is that a move so that the specimen zone spans the flux peak would result in a specimen temperature in the neighborhood of 1850 F and an inner-capsule wall temperature near 1700 F. At this juncture, it would appear that these temperature levels can be tolerated.

Specimens for the fourth high-temperature irradiation capsule (BMI-29-1) in this group are being prepared at Battelle. Six specimens are involved; each is a metal-clad pin containing UO_2 (8 w/o; highly enriched uranium-235) in graphite. Two fuel pins will be clad with Hastelloy X, two with Inconel 702, and two with Carpenter 20 Cb alloy. Like the units discussed above, this capsule will be equipped with heaters and thermocouples and designed so that the specimens are immersed in NaK and heat is transferred across a helium annulus separating an inner and outer container. The specified cladding-surface temperature during irradiation is 1750 ± 50 F. As things now stand, capsule assembly can be started in mid-June and delivery to the MTR can be completed by mid-July.

Fabrication of UO_2 Pellets

H. D. Sheets

About 6000 enriched UO_2 ceramic pellets are being prepared for in-pile testing. Requirements are as follows:

Enrichment, per cent	Outside Diameter, in.	Inside Diameter, in.	Total Length of Pellets Required, in.
25	0.160 ± 0.002	(Solid)	308
50	0.160 ± 0.002	(Solid)	175
25	0.160 ± 0.002	0.048 ± 0.004	618
50	0.160 ± 0.002	0.048 ± 0.004	342

During this period, fabrication of the solid pellets was completed. The yields of acceptable sintered pellets were 89.5 and 93.4 per cent of the 50 and 25 per cent enriched UO_2, respectively. Most of the rejected pellets were oversize. Densities of the sintered pellets, calculated from weights and dimensions, ranged from 10.0 to 10.4 g per cm^3 (91 to 94 per cent of theoretical).

Fabrication of the hollow pellets is under way.

IN-PILE-LOOP PROGRAM

G. A. Francis

Preparation for operation of the two in-pile recirculating gas loops was continued. The Battelle Research Reactor loop was made ready for the start of an irradiation and the Engineering Test Reactor loop installation continued. Details of the activities on the two loops are described in the following paragraphs.

Q-4

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

During the month of May, preparation for the irradiation of a fuel subassembly in the modified loop was completed. This preparation consisted of a recheck of the system and elimination of several significant leaks which had been detected during April.

A fuel subassembly has been received from Aerojet-General Nucleonics and irradiation should begin early in June. The subassembly consists of 19 pins bearing UO_2 pellets. This will be the initial in-pile test of GCRE-I pin-type elements. Present plans call for the irradiation of this element for approximately 1 month for the purpose of obtaining engineering information. Irradiation of the element will be followed by a flux-measuring program and the thermal cycling of a similar unit.

ETR Loop Program

J. V. Baum, W. J. Stevens, and E. O. Fromm

Activity on this program was directed toward the completion of installation of the loop and the completion of construction of loop auxiliary equipment.

At the reactor site, installation and check-out of the mechanical components associated with the loop were continued. The blower cooling coils which were discussed in last month's report have been replaced and additional changes have been made. However, system leakage is still considered excessive. The reactor operator has established an acceptable leak test as one in which there is no measurable pressure decrease from design pressure during a 16-hr period. This is extremely difficult to attain. At present, minor leaks are being detected and repaired.

During this period of cold check, operator training has been initiated. The amount of training has been limited by the necessity for leak checking and the lack of availability of operators. Operators should be available during June, and training should be completed at that time. There has been no change from the July 7 date for in-reactor pipe insertion.

The construction of the attenuator blocks and fuel-element dolly was completed early in May and the parts were shipped to the reactor immediately. With this shipment all equipment designed and built at Battelle is now at the reactor site.

RWD:CRT/all