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PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING AUGUST, 1959

by

Russell W. Dayton
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A. ASSISTANCE TO HAPO

F. R. Shober

The creep properties of annealed and of 15 per cent cold-worked Zircaloy-2 are being studied at elevated temperatures. The long-term tests on the cold-worked material are being concluded. Work pertaining to the effect of a thermal cycle on the creep properties and long-time creep of annealed Zircaloy-2 sheet is being continued. Testing has been completed in the study of the corrosive action of high-temperature water on defected Zircaloy-2-clad uranium specimens.

An investigation to develop a fuel-element leak-detection system has been directed to determination of the rate of dissolution of AgCl and AgBr under dynamic conditions in 180 F flowing water. The solubilities were about a factor of ten less than the solubilities of AgCl and AgBr at 212 F under equilibrium conditions. In the development of a thermal-neutron-flux monitoring system, work has continued on the development of an electrically and mechanically stable ceramic element and on techniques for fabrication of a stable junction between the electrical-heating lead wires and the balancing elements.

Mechanical Properties of Zirconium Alloys

L. P. Rice and J. A. VanEcho

The long-term (10,000 hr or more) creep-property determinations of 15 per cent cold-worked Zircaloy-2 sheet material are being brought to a conclusion. Data concerning the temperature, stresses, and deformations of these tests are shown in Table A-1.

Only one test of this series (7-2-1) has been allowed to go beyond 15,000 hr. Although the total deformation is greater than 1 per cent, there is no indication that third-stage creep has been reached.

Some of the creep rates are so low that it becomes difficult to ascertain the rate very accurately even after 15,000 hr. It also appears that, as the temperature is increased, the slope of creep rate-versus-stress curve becomes greater. This is probably associated with the effect of temperature on relief of the cold working.

Based on the data presently available, the stresses at three test temperatures which will give a minimum creep rate of 0.000005 per cent per hr for the 15 per cent cold-worked Zircaloy-2 are as follows:

Temperature, C	Stress, psi
290	30,500
345	17,500
400	7,500

TABLE A-1. CREEP PROPERTIES OF 15 PER CENT COLD-WORKED ZIRCALOY-2 AT 290, 345, AND 400 C

Specimen	Stress, psi	Minimum Creep Rate, per cent per hr	Deformation, per cent		Test Time, hr
			Final	Load Off	
		290 C (550 F)			
7A-17	37,500	0.00006	1.280	(a)	8,500
7-2-2	35,000	0.000016	0.948	0.636	15,007
7-2-3	30,000	0.000004	0.549	0.289	13,262
7-3-1	25,000	0.000002	0.395	0.189	13,424
7-3-2	20,000	<0.000001	0.291	0.129	13,241
		345 C (650 F)			
7-2-1	25,000	0.000035	1.141	(a)	15,500
7-3-3	20,000	0.000010	0.553	0.385	13,107
7-4-1	15,000	<0.000003	0.342	0.225	12,018
		400 C (750 F)			
7-4-3	15,000	0.00006	1.317	(a)	13,000
7-A-5	12,500	0.00004	0.890	(a)	11,000
7-A-6	10,000	0.000015	0.570	(a)	9,500

(a) Test in progress.

Work on the other two phases of the program is also continuing. This work involves (1) determining the effect of a thermal cycle on the creep properties of both 15 per cent cold-worked and annealed Zircaloy-2 sheet and (2) determining the long-time creep properties of annealed Zircaloy-2 sheet.

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

E. F. Stephan and F. W. Fink

The photographic study of the corrosive action of high-temperature water on defected Zircaloy-2-clad fuel elements was continued.

Two additional elements were studied this month: (1) a tubular specimen to replace a previous one which had a defective weld; (2) a water-quenched rod specimen similar to a specimen already studied (C-8) in which the movie film was not as good as desired. The action time on the duplicate specimens was similar to that previously reported in Table A-1 of BMI-1366.

This completes the present experimental program. A final report is in preparation and the individual films will be combined and reproduced into a single reel.

Development of a Fuel-Element Leak Detector

J. E. Howes, Jr., T. S. Elleman, and D. N. Sunderman

This report summarizes the progress on the development of an isotopic-exchange fuel-element leak-detection system.

The investigations of the past month have been directed toward a determination of the rate of dissolution of AgCl and AgBr under dynamic conditions. Water at 180 F was passed through columns of silver-110-tagged AgCl and AgBr. The silver-110 ions from the dissolved silver halides were separated from the water by a cation-exchange resin. The ion-exchange column was monitored with a scintillation detector to determine the rate of buildup of silver-110 activity and, thus, the rate of silver halide dissolution.

The rate of dissolution of the AgBr and AgCl was determined to be 63 and 550 mg per hr, respectively, at a flow rate of 1 gal per min and a temperature of 180 F. This corresponds to a solubility of 2.78×10^{-7} g per cm^3 for AgBr and 2.42×10^{-6} g per cm^3 for AgCl. These solubilities are about a factor of ten lower than the solubilities of AgBr and AgCl at 212 F under equilibrium conditions.

During the next month, the optimum exchange material will be selected on the basis of the data from solubility experiments. Experiments will then be conducted to determine the effect of flow rate, column size, and particle size on the exchangeability of selected exchange material.

A-4

Thermal-Neutron-Flux Monitoring System

D. R. Grieser, J. W. Lennon, M. S. Snyder, and P. M. Steinback

The objective of this program is to develop a thermal-neutron-flux monitoring system for Hanford reactors. The basic unit for the proposed system is a flux sensor previously developed at Battelle which employs the principle of balancing the temperature of a fission-heated uranium-fueled sensing element and a nearly identical electrically heated balancing element. The electric-power input can then be correlated directly with thermal-neutron flux.

Previous research at Battelle indicated that two major problems were in need of further investigation: the attainment of more reproducibility in (1) desired ceramic element characteristics and (2) stable metal-to-ceramic electrical junctions.

Work has been in progress to develop electrically and mechanically stable ceramic elements with reproducible characteristics. Simultaneously, techniques for fabrication of stable junctions between the electrical-heating lead wires and the balancing elements are under study.

Thus far, ceramic elements have been formed by dust pressing, a convenient method for the shape employed. The dust-pressed element, it has been determined, can be made to conform to the electrical resistivity desired, as measured with low current junctions, and it can withstand thermal stresses adequately for long periods of time in the vicinity of its maximum design operating temperature – provided that its surrounding atmosphere is relatively free of oxygen contamination (the temperature was externally imposed).

Attention is now being given to extrusion as the forming method for forming the ceramic elements. Extrusion is a better production method for the tubular elements contemplated.

Two plasticizers, a starch (Uni-Gum) and polyvinyl alcohol, were tried, and both enabled extrusion of rods. The Uni-Gum rods were stronger after drying.

Preliminary data indicated that the extruded bodies fired to a higher density than did pressed bodies prepared previously using a pressure of 20,000 psi. Also, more $MoSi_2$ was needed in the extruded bodies to compensate for their higher resistivities.

Extruded specimens from one of the mixes, fired at 2900 F in hydrogen, had the required electrical resistivity, but had a slightly higher UO_2 content than necessary by design specifications because of their higher fired bulk density. The composition of this mix was:

Material	Amount, w/o
$MoSi_2$	50.0
Al_2O_3	31.3
UO_2	18.7

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Extrusion experiments will continue and, as satisfactory specimens are produced by this method, they will be subjected to the tests used for the dust-pressed elements. A die for extruding tubes with the dimensions of the prototype element is nearly finished and will be used in further experiments.

A technique for brazing an integral low-resistance junction between the ceramic and the electric leads seems to offer the most promise at present. Tests conducted at high electric power levels (up to 25w) so far indicate good stability of these junctions. In addition to the work toward improved brazed-junction reproducibility, a junction consisting of a flat-surface electric conductor held by spring pressure to a silvered element end face will be investigated further.

Apparatus is nearly ready which will allow the extensive testing of electrically heated balancing elements, either dust-pressed or extruded, and with several types of electrical junctions, for over-all stability under varying conditions of time, electric power, surrounding atmosphere, and ambient temperature. The same apparatus can later be employed for the testing of the prototype flux sensor and its instrumentation.

B-1

DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

N. E. Daniel

An investigation of the temperature and composition of the ternary eutectic in the high-aluminum portion of the aluminum-nickel-uranium system has been completed. The data obtained by differential thermal analyses and metallographic techniques show that the eutectic temperature is 624 ± 2 C and that the composition of the eutectic is approximately aluminum-13 w/o uranium-5.6 w/o nickel. The effects of ternary additions of tin and zirconium on the casting and fabricating characteristics of aluminum-35 w/o uranium alloys are being investigated. Materials are being evaluated with respect to fluidity and castability of the ternary alloys. The cast materials from these studies are being examined by radiographic techniques, metallographic examinations, and chemical analyses.

Aluminum-Uranium-Nickel Eutectic Alloys

V. W. Storhok, A. A. Bauer, and R. F. Dickerson

An investigation to determine the ternary eutectic temperature and composition in the high-aluminum portion of the aluminum-uranium-nickel system has been conducted. As-cast ternary alloys have been studied using differential-thermal-analysis techniques and metallography.

Previously reported differential-thermal-analysis data showed the ternary eutectic temperature to be 624 ± 2 C, or 16 C below the binary eutectic temperatures of both the aluminum-nickel and aluminum-uranium systems. These data have been confirmed by differential thermal analyses of three additional alloys, aluminum-13.1 w/o uranium-5.6 w/o nickel, aluminum-12 w/o uranium-8 w/o nickel, and aluminum-14.2 w/o uranium-6.8 w/o nickel, all of which also exhibited eutectic breaks at 624 C. Additional confirmation of the temperature difference between binary and ternary eutectics was gained by substituting an aluminum-uranium binary eutectic alloy for the graphite neutral specimen normally employed in differential thermal analysis. A difference of 17 C was observed between thermal breaks in the binary and ternary eutectic alloys.

As a result of metallographic examination of the as-cast alloys, the ternary eutectic composition is estimated to be aluminum-13 w/o uranium-5.6 w/o nickel.

This investigation is now completed.

Preparation of Aluminum-Uranium Alloys

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

Interest in aluminum-uranium alloys clad in aluminum for use as fuels in low-temperature water-cooled water-moderated reactors is reflected in the present efforts to prepare more satisfactory fuels of this type. At the present time effort is being directed toward producing hollow cylindrical fuel elements containing uranium concentrations up to 35 w/o uranium and clad inside and outside with aluminum. The contemplated method of fabrication is coextrusion. Improvements in two qualities of the binary alloy are sought. These relate to the soundness and homogeneity of the cast product and its fabrication characteristics. Previous experience has shown that 3 w/o additions of tin and zirconium will improve the casting quality of the aluminum-uranium alloy. Further, these additions inhibit the UAl_3-UAl_4 transformation and in so doing retain greater quantities of aluminum in the matrix of the alloy, thereby decreasing the extrusion pressures necessary and otherwise enhancing the fabricability of the alloys.

Since it has been established that tin or zirconium will completely inhibit the formation of UAl_4, the immediate objective of the program is to ascertain the minimum quantity of each of these additions necessary for the retention of the UAl_3. Therefore, a series of 3-in.-diameter by 10-in.-long castings containing 0.5 to 3.0 w/o of tin and zirconium is being prepared. Standard fluidity tests utilizing a graphite mold are being run on material from the same heats from which the ingots are being cast.

Subsequent evaluation of these ingots will include radiographic examinations of sections of each ingot, metallographic examinations, chemical analyses, and X-ray diffraction studies of selected portions of the as-cast material. The ingots will be extruded and the products will be evaluated by the previously mentioned techniques as well as by tensile tests, creep tests, and corrosion studies.

C. RADIOISOTOPE AND RADIATION APPLICATIONS

D. N. Sunderman

The program undertaken for the Office of Isotopes Development for research in the fields of radioisotope application and radiation chemistry is continuing. Radioisotope applications under study include the use of radiotracer techniques in quality control and the study of intrinsic tracers for process control. Radiation-chemistry studies are being made of the effect of ionizing radiation upon graft polymerization and the nitration of hydrocarbons.

The radiometric method for the determination of calcium appears to be highly accurate and precise. The standard deviation of a series of eight analyses was ±0.5 per cent. The method is very similar to the one developed earlier on this program for magnesium with a similar time requirement of less than 1 hr. Further study will be made of the effect of interfering ions. The results of neutron-activation analysis for the major constituents in cement have improved but remain semiquantitative. One or two more irradiations will be made to establish limits of error with improved techniques.

The Sherritt-Gordon nickel-refining process is being considered for a demonstration of the use of intrinsic tracers for process control. The sensitivity of scintillation-detection techniques for the copper-64 and iron-59 isotopes is being determined. Process solutions are being obtained from the refinery to allow a mock-up of portions of the process for laboratory study.

Experimentation in graft polymerization is centered around the use of electron paramagnetic resonance to determine the number of active sites in irradiated polymethyl- and polybutylmethacrylate. Improved techniques indicate a value as high as 3×10^{-4} site per monomer unit. Thermal-grafting effects and chemical methods of free-radical measurement are under study.

Nitration experiments continue to determine the degree of oxidation at various temperatures in the absence of radiation energy. Results, as yet incomplete, indicate a very high degree of oxidation due in some part to the use of a stainless steel reaction vessel. The use of a glass vessel and shorter reaction times will be studied during the coming month.

Development of Radioactive-Tracer Quality-Control Systems

C. W. Townley, C. T. Brown, D. N. Sunderman,
and M. Pobereskin

Work on the radiometric analysis of calcium in cement has continued during this report period. A method for calcium analysis has been developed which can be completed in less than 1 hr. The calcium is precipitated as $Ca_3(PO_4)_2$ in alkaline solution using $(NH_4)_2HPO_4$ labeled with phosphorus-32. The $(NH_4)_2HPO_4$ solution is radioassayed before and after the precipitation of the calcium, and the decrease in the activity serves as a measurement of the phosphate removed from solution.

C-2

Standard solutions of $(NH_4)_2HPO_4$ and $Ca(NO_3)_2$ were used in the development of the procedure. The phosphate solution contained 4.000 mg of $(NH_4)_2HPO_4$ per ml and enough phosphorus-32 in phosphate form to give initial counting rates on the order of 6000 to 8000 counts per min. The calcium solution contained 1.190 mg of calcium per ml, or 1.666 mg of CaO per ml when calculated on this basis.

The analytical procedure used was as follows:

(1) Add 30 ml of water to 10 ml of phosphate solution (40.00 mg of $(NH_4)_2HPO_4$).

(2) Flush and fill the counting chamber with the solution by drawing the solution through a coarse glass frit into the glass chamber containing the dip-tube counter (Type 1B85, Victoreen Instrument Company).

(3) Radioassay the solution.

(4) Add 10 ml of calcium solution (16.66 mg of CaO) and 2 drops of concentrated NH_4OH (approximately 0.1 ml).

(5) After 15 min of stirring repeat Steps (2) and (3).

The measured radioactivity level is corrected for background, and the radioactivity after precipitation is corrected for dilution due to the addition of the calcium solution. A sample calculation follows:

Initial net radioactivity	6668 counts per min
Final net radioactivity	2305 counts per min
Initial $(NH_4)_2HPO_4$	40.00 mg
Final $(NH_4)_2HPO_4$	40 x 2305/6668 = 13.82 mg
$(NH_4)_2HPO_4$ reacted	26.18 mg

$$CaO = 26.18 \times \frac{3 \times \text{mol. wt CaO } (56.1)}{2 \times \text{mol. wt } (NH_4)_2HPO_4 \text{ } (132)} = 16.67 \text{ mg} .$$

The results of eight calcium determinations are given in Table C-1. The accuracy and precision of the method are within 0.5 per cent at these CaO concentrations. The calcium determination was carried out with CaO concentrations ranging from 3 mg per 50 ml to 40 mg per 50 ml. The accuracy of the results was very good above approximately 12 mg per 50 ml. Below this concentration the results were low.

TABLE C-1. PRECISION AND ACCURACY OF EIGHT CaO DETERMINATIONS

Determination	CaO, mg	Deviation From Average, mg	Calculated CaO, mg	Per Cent Error
1	16.63	-0.04	16.66	0.18
2	16.60	-0.07	16.66	0.36
3	16.81	+0.14	16.66	0.90
4	16.80	+0.13	16.66	0.84
5	16.63	-0.04	16.66	0.18
6	16.60	-0.07	16.66	0.36
7	16.67	None	16.66	0.06
8	16.63	-0.04	16.66	0.18
Average values	16.67	±0.07	--	--
Standard deviation	--	±0.08	--	--

Stirring times shorter than 15 min were tried, but the results were not very good. For the best accuracy, 15 min of stirring was found to be necessary.

Studies were made of the optimum amount of NH_4OH necessary. Without the addition of the NH_4OH the results were as much as 27 per cent low. The hydroxide shifts the phosphate equilibrium in solution toward the PO_4^{-3} ion and aids in complete precipitation of the $Ca_3(PO_4)_2$. Volumes of concentrated NH_4OH ranging from 1 drop to 1 ml were tried. The best results were obtained using 0.1 to 0.2 ml (2 to 5 drops) of concentrated NH_4OH. Quantities of NH_4OH greater than 0.2 ml had very little effect on the accuracy of the results, however. The 0.1 to 0.2 ml of concentrated NH_4OH gave good results over a CaO concentration range of 13 to 40 mg per 50 ml.

During the next report period the effect of interfering ions on the calcium determination will be studied. Work will commence on the development of a procedure for iron and aluminum. An EDTA titration will be tried for these two ions, using $Ag^{110}IO_3$ as the endpoint for the aluminum titration and $Y_2^{91}(C_2O_4)_3$ for the iron titration.

During the past month the neutron-activation-analysis data have been re-evaluated in an effort to improve the accuracy of the results. A more precise method of measuring the areas under the gamma photopeaks has been employed in which a base line is drawn parallel with the x-axis from the leading edge of the gamma photopeak to the base point directly below the maximum value of the peak. This area is then measured and doubled to obtain the correct peak area. The above method applies only to the photopeak of highest energy in any given gamma spectrum.

This method has proven especially valuable in the calcium determination. The calcium-49 photopeak at 3.09 Mev is immediately preceded by two intense pair-production peaks. The gamma spectrum in this region is considerably raised above the background level, rendering useless the usual method of area measurement, i.e., drawing the base line of the photopeak at an angle so it coincides with the apparent increase in the background level.

The corrected analyses for the nine samples analyzed are summarized in Table C-2. The original analyses reported in the July monthly report (BMI-1366) are tabulated for comparison.

TABLE C-2. ACTIVATION ANALYSIS OF CEMENT RAW MATERIALS

	Analysis of Constituent, w/o											
	Al_2O_3			CaO			Mn_2O_3			Na_2O		
	Experimental			Experimental			Experimental			Experimental		
Sample	Original	Corrected	Calculated	Original	Corrected	Calculated	Original	Corrected	Calculated	Original	Corrected	Calculated
Limestone 8768	(a)	(a)	3.8	(a)	(a)	34.3	0.024	0.024	--	0.090	0.080	--
Clay 7440	(a)	(a)	8.7	(a)	(a)	4.0	0.036	0.036	--	0.70	0.76	--
Slag 19459	(a)	(a)	12.8	(a)	(a)	44.9	0.10	0.085	0.23	0.090	0.16	0.12
Limestone 4983	0.33	0.29	0.6	80.0	49.9	53.4	0.001	0.001	--	0.021	0.015	--
Limestone 7604	2.13	1.81	1.7	76.2	46.6	49.6	0.080	0.080	--	0.070	0.070	--
Clay 6956	15.0	9.42	7.7	(b)	(b)	18.3	0.023	0.023	--	0.56	0.50	--
Shale 8772	13.5	14.1	16.2	(b)	(b)	3.9	0.020	0.019	--	0.24	0.19	--
Shale 8773	9.89	9.80	10.8	(b)	(b)	Trace	0.011	0.011	--	0.32	0.23	--
Sand 4299	4.35	4.33	0.8	(b)	(b)	0.6	0.53	0.56	--	0.20	0.12	--

(a) Malfunction of hundred-channel analyzer prevented the analysis of calcium and aluminum in this case.

(b) Calcium-49 activity was too low for analysis.

It will be noted that the Mn_2O_3 and Na_2O results are one-half the values reported in the July monthly report. This correction was due to an error in the concentration determinations in the $MnSO_4$ and $NaNO_3$ standards.

During the next report period one more activation analysis will be run. The effect of improved dosimetry and measurement of photopeak areas will be determined. A limit of accuracy will be placed on the method.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, H. B. Brugger, J. F. Kircher,
and M. Pobereskin

During the past month development of a radiotracer control system for the copper-stripping operation of the ammonia-leach nickel-refining process has continued. The development of more sensitive methods of measuring low levels of copper-64 activity in solution or of concentrating the copper activity is being studied. The methods being investigated for concentrating the copper-64 activity are:

(1) Absorption on a chelating ion-exchange resin

(2) Precipitation with an adsorption on a carrier for copper

(3) Isotopic exchange with inactive CuS.

In order to simulate actual process conditions more closely in laboratory experiments, samples from Sherritt-Gordon nickel-refinery process streams are being obtained. These samples will consist of the solution entering the copper-strip step in the main circuit and the solution entering the iron-removal step, as well as both the copper-free and the iron-free solutions. Samples of the copper-strip filter cake and iron filter cake will also be sent.

The experimental analysis of the exchange between the copper in the solution and that in the copper sulfide precipitate is nearly completed. The final experiments will be done using process solution from the Sherritt-Gordon plant. Results of the study so far indicate that a fairly rapid exchange takes place between the copper in solution and copper in the precipitated sulfide. This effect will, therefore, have to be taken into consideration in a precipitation-filtration operation where the specific activity of copper in the solution differs from the specific activity of the copper in the precipitate. Such a condition may result when solution containing freshly added copper-64 tracer, from which CuS has been precipitated, is passed through a filter containing a thick cake of CuS in which the copper-64 has decayed for several hours.

Experimental study has begun on radiotracer control of the iron-removal operation in the Sherritt-Gordon mixed cobalt-nickel circuit. A standard sample of iron-59 isotope has been ordered for laboratory evaluation of detection efficiency and counting techniques. Construction of equipment to simulate conditions in the plant is also under way. The research for a suitable substitute for iron-59 as a radiotracer in this process is being continued.

Next month effort will be concentrated on study of iron removal from the mixed-metals circuit in the nickel-refining process. Work will continue on development of more sensitive methods for measuring copper-64 activity in solution. An evaluation of possible contamination problems and radiation hazards will also be initiated.

Graft-Polymerization Studies

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

During this month free-radical studies utilizing the Varian electron-paramagnetic-resonance apparatus, were continued. Several attempts to determine hyperfine structure of the spectra of irradiated polymethyl- and polybutylmethacrylate proved unsuccessful. The lines were too broad to resolve fully. During this investigation it was found that the microwave cavity was being saturated. A decrease in power resulted in an increase in the apparent number of sites. The highest value of active sites as measured in polymethylmethacrylate by EPR was approximately 3×10^{-4} site per monomer unit.

Further study was made of the extent of thermal grafting occurring along with the irradiation grafting. Steps were taken to reduce the thermal grafting to a minimum by "deactivation" of the polymers by heating under vacuum and by elimination of heating during grafting. Polymethyl- and polybutylmethacrylate have been treated under these conditions to determine the amount of thermal grafting. Results will be available next month and will allow isolation of radiation from nonradiation effects.

The chemical method of free-radical measurement was refined and appears to give good precision. For example, three successive samples of deactivated radiation-polymerized polymethylmethacrylate powder were irradiated in a vacuum of $0.3\ \mu$ to a total dose of 5.75×10^5 r and found to have $9.5 \pm 0.2 \times 10^{-4}$ site per monomer unit. All the polymers previously measured will be repeated with this improved method.

Radiation-polymerized polymethylmethacrylate has been fractioned into relatively narrow molecular-weight fractions. These fractions will be grafted in the usual way. It is hoped that the subsequent fractionation of grafted polymer will result in separation solely on the basis of amount of vinylpyrrolidone grafted.

During the coming months the investigation will proceed along the lines indicated above. Other methacrylates will be included in the study as they are prepared and deactivated. Polyesters with backbone structures ranging from saturated aliphatic and aliphatic with varying amounts of unsaturation to partially aromatic have been prepared. These polymers will be included in the studies as outlined above as soon as this is practicable.

C-7 and C-8

Nitration of Hydrocarbons

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

During August, thermal runs with the nitric acid-cyclohexane system (in the absence of radiation) were made at four different temperatures. Temperatures ranged from ambient to 140 C. Reaction time for each run was about 1 week. The runs were conducted in the stainless steel apparatus.

Chemical analyses of these thermal runs have not been completed as yet. Results to date indicate the formation of nitro paraffins, and possibly nitrate compounds. The yield of oxidation products (primarily adipic acid) increases with temperature. In the thermal run made at the highest temperature (140 C), the reaction mixture appeared to be completely oxidized.

Several ways of reducing oxidation are under consideration. These include the use of a glass vessel rather than a stainless steel vessel, and shorter reaction times. During September, additional thermal runs will be made under these modified conditions.

Dosimetry measurements in the irradiation vessel and the first irradiation run, at 60 C, were completed in August. Results will be reported as soon as they become available.

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

The study concerned with the effect of valence compensation in the stabilization of uranium dioxide has continued. Although weight-change data obtained from specimens containing CaO as well as La_2O_3 or Y_2O_3 showed that the addition of CaO did not produce as stable a material as the binary oxide with 60 mole per cent of either La_2O_3 or Y_2O_3, recent analytical results indicate that all samples contained less than the desired amount of CaO additive. Additional bodies will be prepared to further evaluate the CaO additions. Microbalance work is being conducted on the gamma modification of U_3O_8, produced at various pressures above 16,000 atm, to establish composition.

Three additional capsules containing tensile and cyclic-strain specimens of Type 347 stainless steel have been placed in the ETR during the shutdown for Cycle 19. The specimens from these capsules will be given postirradiation heat treatments prior to testing to supplement data from specimens irradiated cold and at 600 F. The "cold" capsules already in the reactor were exposed to about 6 days at full power during Cycle 18 of the ETR.

Niobium-base alloys, being studied as a possible cladding for EBR application, cracked during cold fabrication. These alloys are being analyzed for impurity pickup and plans are being made to prepare additional alloys. Unalloyed niobium is being corrosion tested in 680 F water and 750 F steam as part of a cooperative program between KAPL, BAPD and Battelle. After 26 days in 680 F water, the specimens were still gaining weight, while those tested in 750 F steam began to flake and lose weight. Of the niobium-base alloys studied, the binary containing 12.6 a/o vanadium exhibited the greatest resistance to deformation at 1200 F and gained only 0.52 mg per cm^2 after 168 hr of exposure to 680 F water. Another screening series of binary alloys which contain additions of cerium, nickel, palladium, and yttrium in amounts up to 5 a/o is being prepared for additional corrosion tests.

The design of the in-pile test device for use in the study of effects of irradiation on the creep properties of Zircaloy-2 is in process. A major problem in this design is the balance of gamma heat and the external heaters with heat flow in the specimen. Material has been obtained for the study of the strain-aging phenomena in Zircaloy-2 and this work will be initiated as soon as possible.

A research program has been initiated for the purpose of developing an analytical technique for monitoring the oxygen concentration in large-scale radioactive sodium systems. The method should have a sensitivity of ±1 ppm of oxygen in the range from 0 to 10 ppm and be amenable to rapid determination. Methods based on mass spectrometry, the sensitivity of certain properties to oxide contamination, polarography, and plugging-indicator techniques are being considered.

F-2

Valence Effects of Oxide Additions to Uranium Dioxide

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

The effects of oxide additions to uranium oxide are being studied with respect to stabilizing the UO_2 fluorite structure under oxidizing and reducing conditions at elevated temperatures. It has been shown in previous work that the addition of about 60 mole per cent of the rare-earth oxides, La_2O_3 or Y_2O_3, to UO_2 preserves the fluorite structure and reduces the tendency of the resulting mixed oxide to volatilize above 1000 C.

Presently, efforts are being made to reduce the relatively large amount of additive required to stabilize UO_2 by substituting CaO for part of the rare-earth oxide additions. The divalent additives are expected to be more effective than the trivalent additives for valence compensation. Solid solutions were prepared of the following mole percentages with La_2O_3 and Y_2O_3 as rare-earth oxides:

UO_2-20 R_2O_3-20 CaO
UO_2-25 R_2O_3-25 CaO

Analyses indicated that all of the samples contained less than the intended percentages of CaO. Weight-change data showed that none of these materials has been as effective as the UO_2-60 mole per cent R_2O_3 compacts in stabilizing the UO_2 under oxidizing conditions at 1760 C.

Because of the loss of CaO in the preparation of the UO_2-R_2O_3-CaO bodies, a further effort will be made to prepare bodies of the intended compositions. These will be tested on the basis of (1) X-ray diffraction structural studies, (2) stability in dry flowing air at 1760 C, and (3) chemical composition. It is expected that the data obtained on these specimens will conclude the work with the ceramic ternary oxides.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

The effect of ultrahigh pressure and high temperature on uranium oxides and on reactions of uranium oxides with mixed oxides is being investigated. In prior work emphasis has been placed on study of the effect of pressure and temperature on U_3O_8. Microbalance work is being conducted on the gamma modification of U_3O_8, produced at various pressures above 16,000 atm, to establish composition. Further work on U_3O_8 will be confined to an attempt to further determine if the cubic U_4O_9 structure can be maintained by high pressure with a composition near that of U_3O_8.

A fully supported version of the new die has been completed and additional calibration work performed. Further work was performed on densification of UO_2 in support of a radiation-effects program. Pressures of 60,000 atm at temperatures above 1000 C were utilized.

Further work will return to the study of the effects of pressure on reactions of uranium oxides with mixed oxides using the new die.

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, F. R. Shober, J. E. Howes, and J. F. Lagedrost

A surveillance program is in progress to determine the effects of fast-neutron irradiation (greater than 1 Mev) on the mechanical properties of AISI Type 347 stainless steel. The program is in support of the KAPL C-33 loop and other loops in the ETR constructed of Type 347 stainless steel. Twenty-one capsules containing tensile, fatigue, or impact specimens make up the program. One group of capsules will be irradiated at process-water temperature (120 F), a second group will be irradiated at approximately 600 F, and a third group will be irradiated at process-water temperature and given postirradiation annealing treatments. All capsules have been fabricated and shipped to the ETR, where they are either being irradiated or are waiting to be inserted in the proper reactor space.

Irradiation-damage data have been reported for Type 347 stainless steel at exposures up to 3.76×10^{21} n per cm^2. The main objective of the surveillance program is to determine mechanical properties of irradiated Type 347 stainless steel after exposures between this level and the exposure level of 1.6×10^{22} n per cm^2 ultimately expected in the KAPL loop. In order that the irradiation program be of use to the loop program, the exposures obtained in the irradiation capsule program will have to lead those of the loop by a minimum of 6 months.

Three capsules containing tensile and cyclic-strain specimens will be irradiated at process-water temperatures and given postirradiation annealing treatments to supplement data from specimens irradiated cold and at 600 F. These capsules, BMI-24-18, BMI-24-20, and BMI-24-22, were placed into the ETR during the shutdown for Cycle 19 in positions K-8-NW, K-8-NE, and L-8-SE, respectively.

Total neutron accumulation for the "cold" capsules (irradiated at process-water temperatures) as of the end of Cycle 18 is given in Table F-1, along with a list of current and proposed irradiation parameters of the other capsules in the surveillance program. The ETR operated an equivalent of about 6.2 days at full power during Cycle 18 and has been in operation at partial power on Cycle 19 since August 15.

The eight capsules designed to operate at a temperature near 600 F have not been inserted in the ETR, since space of suitable quality has not been available.

TABLE F-1. CAPSULES PREPARED FOR THE IRRADIATION-SURVEILLANCE PROGRAM ON TYPE 347 STAINLESS STEEL

Capsule	Type of Specimen in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date[a]	Approximate Exposure at Time of Removal[b], nvt	Total Exposure as of August 15, 1959, nvt		Location	Remarks
					Top	Bottom		
BMI-24-1	Tensile and fatigue	600	January, 1959	1.55×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility for melting
BMI-24-2	Tensile and fatigue	120	January, 1962	1.31×10^{22}	2.364×10^{21}	3.235×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-3	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-4	Tensile and fatigue	120	January, 1963	1.78×10^{22}	1.435×10^{21}	2.598×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-5	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-6	Tensile and fatigue	120	June, 1961	1.08×10^{22}	3.313×10^{21}	2.532×10^{21}	ETR K-8-NE	Being irradiated
BMI-24-7	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-8	Tensile and fatigue	120	June, 1962	1.54×10^{22}	1.629×10^{21}	2.597×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-9	Tensile and fatigue	600	--	--	--	--	ETR	To be irradiated
BMI-24-10	Tensile and fatigue	120	January, 1961	$.84 \times 10^{22}$	2.61×10^{21}	2.920×10^{21}	ETR K-8-SE	Being irradiated
BMI-24-11[c]	Tensile and fatigue	600	--	--	--	--	ETR	Damaged at ETR
BMI-24-12	Tensile and fatigue	120	June, 1960	$.61 \times 10^{22}$	3.524×10^{21}	2.866×10^{21}	ETR L-8-SE	Being irradiated
BMI-24-13	Impact	600	--	--	--	--	ETR	To be irradiated
BMI-24-14	Impact	120	June, 1962	1.54×10^{22}	3.103×10^{21}	3.188×10^{21}	ETR K-8-NW	Being irradiated

TABLE F-1. (Continued)

Capsule	Type of Specimen in Capsule	Proposed Irradiation Temperature, F	Approximate Removal Date[(a)]	Approximate Exposure at Time of Removal[(b)], nvt	Total Exposure as of August 15, 1959, nvt		Location	Remarks
					Top	Bottom		
BMI-24-15	Impact	600	--	--	--	--	ETR	To be irradiated
BMI-24-16	Impact	120	June, 1960	$.61 \times 10^{22}$	3.161×10^{21}	2.964×10^{21}	ETR K-8-NW	Being irradiated
BMI-24-17(c)	Tensile and fatigue	600	October, 1958	3.25×10^{20}	--	--	BMI	Examined at BMI Hot-Cell Facility after high temperature observed
BMI-24-18	Tensile and fatigue	120	--	--	--	--	ETR	To be irradiated for post-irradiation annealing studies
BMI-24-19(c)	Tensile and fatigue	600	--	--	--	--	ETR	Fabricated to replace BMI-24-17
BMI-24-20	Tensile and fatigue	120	--	--	--	--	ETR	To be irradiated for post-irradiation annealing studies
BMI-24-21	Tensile and fatigue	600	--	--	--	--	ETR	Fabricated to replace BMI-24-1
BMI-24-22	Tensile and fatigue	120	--	--	--	--	ETR	To be irradiated for post-irradiation annealing studies

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

(b) Based on maximum fast flux at tube of 1.7×10^{14} nv for 6-month periods.

(c) Thermocouple lead capsule.

F-6

Development of Niobium-Base Alloys

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

Niobium and niobium-base alloys are potentially suitable materials for cladding fuel elements at elevated temperatures. The alloys being studied for possible use as cladding materials in the EBR include 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 an alloy for comparison with the niobium alloys, containing vanadium-10 w/o titanium-1 w/o niobium.

Melting, hot and cold fabrication, metallographic examinations, chemical analyses, and a short heat-treatment study have been completed. The niobium-1 w/o chromium, niobium-4.5 w/o zirconium, niobium-20 w/o titanium-1.5 w/o chromium, and vanadium-10 w/o titanium-1.0 w/o niobium were cold rolled to 0.035-in. sheet. This sheet showed severe edge cracking. However, the center portion of the rolled sheets contained sound material. Sheet tensile and corrosion specimens have been prepared from this center portion of the rolled sheet. The unalloyed niobium and niobium-40 w/o titanium-10 w/o aluminum failed to cold roll.

In order to determine if contamination of the ingots by oxygen occurred, the ingots are being examined for oxygen content by vacuum-fusion methods. The unalloyed niobium showed an increase in oxygen content from 200 to 800 ppm, which might account for its poor fabrication characteristics. However, the fabricated niobium-1 w/o chromium alloy contains only 90 ppm oxygen, which is believed to be too little to appreciably affect fabrication. Analyses of the remaining six alloys have not been completed.

The niobium-2 w/c chromium and niobium-40 w/o tantalum-2 w/o chromium alloys have been encapsulated in evacuated stainless steel packs and are to be rolled at 2000 F.

Mechanical properties of alloys which were fabricated are being determined at 650 and 800 C, and sheet-to-sheet weldability of the cold-fabricable alloys is being studied.

Ingots to replace alloys containing excessive amounts of oxygen will be prepared; and attempts will be made to fabricate these alloys.

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 selected niobium-base alloys for possible service in pressurized-water reactors was continued. Final evaluation of a series of 2-lb ingots for mechanical properties is being completed. This work indicates that a binary 12.6 a/o vanadium alloy has the most outstanding combination of 1200 F creep strength and corrosion resistance to hot water of seven alloys tested. The preparation and evaluation of several series of 50-g screening-alloy ingots is in process to determine whether satisfactory corrosion resistance can be maintained with lower alloy levels in niobium in order to obtain improved fabricability.

Corrosion results obtained to date in 600 and 680 F water and 750 F 1500-psi steam are summarized in Table F-2. Based on exposures ranging up to 196 days, it appears that alloys prepared from commercial-purity niobium are as corrosion resistant as those prepared from high-purity melting stock. However, in the unalloyed condition, the high-purity niobium is the more corrosion resistant. Alloys which appear most corrosion resistant are those possessing adherent tarnish films and low weight gains. Included in this group are ternary alloys containing 28.2 a/o titanium-6.1 a/o chromium and 10 a/o chromium-5 a/o iron, and all alloys containing more than 40 a/o zirconium.

Corrosion tests are under way on unalloyed niobium as part of a cooperative testing program with the Bettis and Knolls Atomic Power Laboratories. All corrosion specimens were taken from the same high-purity base niobium ingot and were prepared for testing by Bettis. Samples have been exposed 28 days to 680 F water and 750 F 1500-psi steam. Those in 680 F water continue to gain weight, while those in 750 F steam began to flake and lose weight after 28 days of exposure. The 1-day intercept and slope of the curves of log-log plots of weight gain versus time for the two test conditions are tabulated below.

Medium	Exposure Time, days	Log-Log Plot of Weight Gain Versus Time: 1-Day Intercept, mg per dm^2	Log-Log Plot of Weight Gain Versus Time: Slope
680 F water	28	26	0.42
750 F 1500-psi steam	21	20	0.70

Corrosion tests are being continued on all specimens except the commercial-purity alloys which were exposed 196 days to 680 F water. These specimens have been removed from test and are being examined for evidence of oxygen contamination, hydrogen absorption, and the nature of the oxide film.

Hardness measurements at temperatures up to 1650 F were obtained on annealed samples of the 2-lb scaleup alloys and are given in Table F-3. These results are in fair agreement with the tensile and creep properties determined earlier at temperatures up to 1380 F on alloys up through NL-7, inclusive. However, the greatest resistance to deformation at 1200 F was not found in the 46.8 a/o zirconium-5.06 a/o titanium alloy as the hardness data in Table F-3 would indicate, but, rather, occurred in the 12.6 a/o vanadium alloy.

The effect of time on the hardness of the 12.6 a/o vanadium and 7.18 a/o molybdenum alloys at 1200 F was determined as shown in Table F-4. For both alloys, the resistance to deformation decreases slightly, at about the same rate, with increasing times up to 2000 sec.

TABLE F-2. SUMMARY OF CORROSION RESULTS OBTAINED ON NIOBIUM ALLOYS EXPOSED IN HIGH-TEMPERATURE WATER AND STEAM

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
			Commercial Niobium, Rocking-Hearth Melts			
Unalloyed Nb	168	-13.3	42(a)	Disintegrated	28(a)	Disintegrated
10.5 Zr	--	--	196(a)	0.67	84	-17.5
26.1 Zr	--	--	196(a)	0.67	--	--
35.7 Zr	--	--	196(a)	0.66	--	--
45.7 Zr	--	--	196(a)	0.55	--	--
1.08 W	--	--	196(a)	-2.60	84	-21.1
4.67 W	--	--	196(a)	-29.3	--	--
9.56 W	--	--	7(a)	Cracked	--	--
2.45 Mo	--	--	196(a)	-7.10	84	-76.6
5.20 Mo	--	--	196(a)	-1.30	84	-26.1
7.40 Mo	--	--	196(a)	0.62	--	--
4.42 V	--	--	196(a)	0.42	84	0.55(b)
6.59 V	--	--	196(a)	0.73	84	1.27
8.93 V	--	--	196(a)	0.59	84	1.02
10.7 V	--	--	196(a)	0.78	--	--
13.7 V	--	--	196(a)	0.50	--	--
24.2 V	--	--	196(a)	--	--	--
4.90 Fe	--	--	196(a)	0.10	84	-6.41
9.41 Ti	--	--	196(a)	0.65	84	1.35
18.8 Ti	--	--	196(a)	0.48	--	--
24.3 Ti	--	--	196(a)	0.52	--	--
30.5 Ti	--	--	196(a)	0.40	--	--
33.8 Ti	--	--	196(a)	0.33	--	--
12.0 Ti-0.5 Cr	--	--	196(a)	0.66	--	--
20.2 Ti-2.1 Cr	--	--	196(a)	0.39	--	--
28.2 Ti-6.1 Cr	--	--	196(a)	0.20	--	--
12.0 Ti-4.2 Mo	--	--	196(a)	0.64	--	--
17.4 Ti-6.2 Mo	--	--	196(a)	0.54	--	--
23.1 Ti-7.8 Mo	--	--	196(a)	0.45	--	--
10.4 Ti-5.0 V	--	--	196(a)	0.56	--	--
16.1 Ti-8.4 V	--	--	196(a)	0.40	--	--
22.6 Ti-11.0 V	--	--	196(a)	0.48	--	--

TABLE F-2. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
			High-Purity Niobium, Consumable-Electrode Melts			
Unalloyed Nb	168	0.75	168	-2.29	168	-44.2
7.18 Mo	168	0.66	168	0.25(b)	70(a)	Cracked
12.6 V	168	0.37	168	0.52	168	0.76
46.8 Zr-5.06 Ti	140	0.30	140	0.78	140	2.66
11.2 Ti-3.2 Mo	140	0.45	140	0.49	140	0.84
18.8 Ti-8.7 Mo	140	0.19	140	0.39	140	0.74
9.9 Zr-9.4 V	56	0.21	56	0.28	56	-1.32
5.7 Zr-11.4 V	56	0.18	56	0.24	56	0.12(b)
9.1 Ti-6.3 Cr	56	0.18	56	0.26	56	-1.19
			High-Purity Niobium, Rocking-Hearth Melts			
Unalloyed Nb	--	--	84	1.43	--	--
Unalloyed Nb	--	--	56	-33.6	--	--
Unalloyed Nb	--	--	56	0.53	--	--
1 Zr	--	--	84	-95.2	--	--
5 Zr	--	--	56	-0.68	--	--
10 Zr	--	--	56	-0.20	--	--
40 Zr	--	--	56	0.41	--	--
65 Zr	--	--	56	0.66	--	--
75 Zr	--	--	56	0.89	--	--
90 Zr	--	--	56	0.59	--	--
2.5 Ti	--	--	84	0.02(b)	--	--
10.0 Ti	--	--	84	0.52	--	--
25.0 Ti	--	--	84	0.32	--	--
1 Cr	--	--	84	1.06	--	--
5 Cr	--	--	84	0.66	--	--
10 Cr	--	--	56	-0.14	--	--
1 Fe	--	--	56	0.26(b)	--	--
10 Zr-5 Ti	--	--	56	0.37	--	--
25 Zr-5 Ti	--	--	56	0.36	--	--
25 Zr-15 Ti	--	--	56	0.49	--	--
25 Zr-25 Ti	--	--	56	0.25	--	--
35 Zr-5 Ti	--	--	56	0.33	--	--
35 Zr-15 Ti	--	--	56	0.33	--	--
45 Zr-5 Ti	--	--	56	0.36	--	--
10 Zr-5 Mo	--	--	56	0.21	--	--
35 Zr-5 Mo	--	--	56	0.38	--	--
45 Zr-5 Mo	--	--	56	0.32	--	--

TABLE F-2. (Continued)

Alloy Addition (Balance Niobium), a/o	600 F Water		680 F Water		750 F Steam	
	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2	Exposure Time, days	Total Weight Change, mg per cm^2
35 Zr-5 Al	--	--	56	0.46	--	--
45 Zr-5 Al	--	--	56	0.32	--	--
10 Zr-5 Cr	--	--	56	0.32	--	--
45 Zr-5 Cr	--	--	56	0.28	--	--
10 Zr-5 Fe	--	--	56	0.24	--	--
2.5 V	--	--	84	0.71	--	--
2.5 V-2.5 Ti	--	--	56	0.51	--	--
2.5 V-2.5 Mo	--	--	56	0.56	--	--
2.5 V-2.5 Fe	--	--	56	0.60	--	--
2.5 V-2.5 Cr	--	--	56	0.44	--	--
2.5 V-2.5 Al	--	--	56	0.67	--	--
5 V-2.5 Zr	--	--	56	0.26	--	--
5 V-25 Zr	--	--	56	0.51(b)	--	--
5 V-35 Zr	--	--	56	0.64	--	--
5 V-45 Zr	--	--	56	0.24	--	--

(a) Off test.

(b) Losing weight.

TABLE F-3. HOT-HARDNESS DATA FOR HIGH-PURITY NIOBIUM-BASE ALLOYS AT VARIOUS TEMPERATURES

Alloy	Alloy Content (Balance Niobium), a/o	Hardness(a) at Temperature Shown, VHN						
		75 F	600 F	900 F	1200 F	1400 F	1650 F	75 F(b)
TP-37	100 Nb(c)	81	78	56	37	26	21	82
NL-2	12.6 V(c)	246	164	155	153	153	149	259
NL-3	7.18 Mo	199	133	133	116	104	89	187
NL-4	46.8 Zr-5.06 Ti	282	199	200	201	186	153	305
NL-7	11.2 Ti-3.2 Mo	178	107	87	80	79	87	189
NL-8	9.9 Zr-9.4 V	277	212	194	186	185	212	291
NL-9	5.7 Zr-11.4 V	275	188	181	186	194	245	293
NL-10	9.1 Ti-6.3 Cr	239	158	148	153	150	146	246

(a) All alloys vacuum annealed 1 hr at 2730 F before testing.
(b) Determined after heating samples to 1650 F.
(c) Values reported previously.

TABLE F-4. EFFECT OF TIME ON THE HOT HARDNESSES OF TWO NIOBIUM-BASE ALLOYS AT 1200 F

Indenter(a) Dwell Time, sec	Hardness(b), VHN	
	Alloy NL-2 (12.6 a/o Vanadium)	Alloy NL-3 (7.18 a/o Molybdenum)
5	162	114
10	147	115
20	164	111
40	157	108
80	155	104
200	158	108
500	153	107
1000	155	110
2000	152	104

(a) Sapphire indenter with 730-g load used in all tests.
(b) Average of at least three indentations using the same dwell time.

Elevated-temperature tensile and creep tests on Alloys NL-8, NL-9, and NL-10 (see compositions in Table F-3) are in process. While it is possible that one or more of these alloys will show greater strengths at 1200 F than the 12.6 a/o vanadium alloy, none of these three alloys showed acceptable room-temperature ductility in tensile or bend testing.

Preparation of an additional series of 50-g screening-alloy ingots is in process. These include binary alloys containing additions of cerium, nickel, palladium, and yttrium, in amounts up to 5 a/o, and a series of ternary and more complex niobium-vanadium-base alloys containing titanium or zirconium plus oxygen or carbon as dispersion strengtheners.

Investigation of the Creep Properties of Zircaloy-2 During Irradiation at Elevated Temperatures

J. H. Stang, A. P. Young, and F. R. Shober

A study of the creep characteristics of Zircaloy-2 at elevated temperatures during exposure to a fast-neutron flux (neutrons having energies greater than 1 Mev) has been initiated. It is generally believed, based on postirradiation data, that irradiation should enhance the creep properties of Zircaloy-2. Arguments, however, can be advanced to show that irradiation may be expected to have adverse influence on the creep properties. This is especially true for the temperature range 500 to 700 C, where Zircaloy-2 has been shown to exhibit some properties of a strain-aged material. It is intended to compare the creep behavior of Zircaloy-2 in a reactor with the creep behavior out-of-reactor using similar test units. Preirradiation and postirradiation measurements of tube diameters are to provide the basis for comparison.

Work has been started on the design of a capsule for the in-pile studies. Heat-transfer calculations will be made to establish a specimen geometry which is conducive to maintaining a uniform test temperature in the specimen gage section during irradiation. One of the major problems is to balance the heat generated by gamma heating in the specimen and the external heaters with heat flow in the specimen. It has been suggested that individual specimens and capsules would be better in this respect than a multiple-gage-section specimen in a single capsule. It is planned, also, to build an out-of-reactor test unit similar to the in-reactor capsule to determine out-of-reactor creep properties of Zircaloy-2. This unit will be based on the in-reactor capsule design. The operation of the out-of-reactor unit will provide a proof test for the in-reactor test equipment. It is expected that design and construction of the out-of-reactor units will take approximately 4 to 6 months.

The initiation of studies of strain-aging phenomena in Zircaloy-2 has been delayed until material can be secured. Zircaloy-2 plate and wire to be tested will be prepared from the same billet, and detailed chemistry and fabrication history of the billet will be noted. This material will be used for all specimens prepared, including tensile, internal-friction, and metallographic specimens for strain-aging studies.

F-13

Determination of Oxygen in Sodium at Concentrations Below 10 PPM

D. R. Grieser, P. M. Steinback, and W. H. Goldthwaite

A research program has been initiated to develop an analytical technique for monitoring the oxygen concentration in large-scale radioactive sodium systems. The method sought is to (1) have a sensitivity of ±1 ppm of oxygen in the range from 0 to 10 ppm and (2) be amenable to rapid and, preferably, continuous determination. Methods of analysis now employed do not appear to satisfy completely both of these specifications. Of the present methods, only one is reported to have the sensitivity desired. This is the vacuum distillation-flame spectroscopy method devised at Argonne National Laboratory. This method, however, does not meet the requirement of rapid analysis. This, of course, does not detract from its usefulness, particularly as a laboratory tool; in fact, vacuum distillation offers a highly refined calibration technique for the analytical experiments that will be conducted as the present research progresses.

In view of the limitation of available methods, emphasis to date has been placed on formulating new analytical approaches, based on various chemical and physical principles. Several concepts have emerged as a result of this effort. The leading of these are described briefly below.

(1) Mass spectrometry. This technique would involve the analysis by a fixed-detector mass spectrometer of ionized sodium vapor obtained by evaporation of single droplets drawn from a large flow system; the concentration of oxide species would be measured directly.

(2) Methods based on a sensitivity of certain properties to oxide contamination. Properties of particular interest include

  (a) The phase change on a reflection of polarized light from the surface of molten sodium or from the surface of a getter metal. Use would be made of polarimetric optical apparatus.

  (b) The formation of an Na_2O precipitate film on a sodium-exposed surface as the temperature of that surface is reduced below the prevailing oxide-saturation level. Again, optical apparatus would be used.

  (c) Surface tension as manifested by changes in sodium drop-formation characteristics from a capillary tube.

  (d) The measurement of electrical-resistance characteristics of sodium.

Other properties have been considered but, at this time, are not regarded as promising, primarily because there is little hope that the required sensitivity to contamination exists and, even if it does, the time and expense involved to establish it might be prohibitive. These include properties such as the following: Hall effect, thermoelectric power, superconductivity, melting-point depression, nuclear magnetic resonance, and radiation emission.

(3) Polarography. This would involve special polarographic techniques with sodium electrodes substituted for the conventional mercury electrodes.

(4) Plugging-indicator techniques. It is likely that significant refinements can be made in existing plugging-indicator techniques, which have been well proven in the 10 to 20-ppm oxide contamination range and beyond. One possibility for improvement lies in plugging-indicator construction based on a small-bore venturi-type orifice with a high-sensitivity electromagnetic flowmeter to detect variation in sodium flow as the orifice temperature is decreased and precipitating Na_2O tends to plug it.

As it now stands, the activity during September will be devoted to final selection of the most promising concepts for experimental investigation, initiation of the design of experimental devices for studies of the sensitivity of selected properties to contamination, and the initiation of the design of a sodium pumping loop in which the oxygen concentration in the sodium can be controlled by gettering techniques.

STUDIES OF ALLOY FUELS

R. F. Dickerson

A study of the gamma-immiscibility loop in the niobium-uranium alloy system has been initiated and it is also planned to determine the effects of oxygen on these composition limits. After 98 days in 600 F water the weight change for all of the alloys is about the same — this is only about four times that undergone by Zircaloy-2 under similar conditions. The results obtained from the 680 F water test are not quite as consistent because of the type of film which is produced.

The study of thorium-uranium alloys is being continued with emphasis on ternary alloys containing zirconium additions. This series will contain larger zirconium additions than those alloys studied to date. It is postulated that increasing the zirconium-to-uranium ratio will increase the high-temperature strength of the alloys and introduce the possibility of improving the corrosion resistance by surrounding thorium with a high-zirconium-uranium alloy. In addition to the alloy studies, the ThC-UC system is being studied.

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Development of Niobium-Uranium Alloys

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

Niobium-rich niobium-uranium alloys have received limited consideration as reactor fuels. This is partly due to a lack of information concerning their mechanical and physical properties, fabrication characteristics, and behavior in various corrosive media. Prior investigations conducted on uranium-rich uranium-niobium alloys have indicated that the properties are affected by the impurity level of the base material. Therefore, the effects of oxygen and zirconium on the properties of niobium are being investigated.

Three different grades of niobium were used in alloy preparation. One grade contains 0.7 w/o zirconium and 600 ppm oxygen, a second contains 0.17 w/o zirconium and 700 ppm oxygen, and a third contains 0.02 w/o zirconium and less than 300 ppm oxygen.

A study of the gamma-immiscibility loop in the niobium-uranium system has been initiated in which the composition limits will be defined by metallographic examination of suitably heat treated samples. The effects of oxygen on these composition limits will also be determined. The materials to be used in the preparation of the niobium-uranium alloys are electrolytic uranium and sintered niobium containing 200 ppm oxygen. In order to study the effects of oxygen on the gamma_1 plus gamma_2 loop, some of the niobium has been exposed to measured amounts of oxygen in a modified Sieverts apparatus. Alloys prepared from this niobium containing 1000 and 3000 ppm oxygen will be compared with alloys prepared from niobium containing 200 ppm oxygen. Alloys of uranium containing 10, 20, 30, 40, 50, 52, 54, 56, and 58 w/o niobium will be arc cast into finger bars. Each composition will be prepared using niobium with all three oxygen contents. Melting of these alloys has been initiated.

The niobium-10 and -20 w/o uranium alloys have been successfully fabricated by forging. No successful fabrication of alloys containing more than 20 w/o uranium has been accomplished. Ingots of the niobium-30 and -40 w/o uranium alloys have been prepared and are to be forged at 3000 F as soon as a furnace now under construction has been completed.

The results of corrosion studies in water are shown in Table F-5 for 98 days of exposure at 600 and 680 F. All alloys being tested in 680 F water have a brown oxide coating and are losing weight. Most of the alloys being tested in 600 F water have also begun to lose weight but still have a black oxide coating.

Compatibility studies of niobium-uranium alloys with NaK at 1600 F are in progress. The specimens will be examined after 500 hr of exposure.

Specimens for creep studies are being prepared. It is planned to determine the creep properties of fabricable niobium-uranium alloys at 1600, 1900, and 2400 F in vacuum. Creep tests of 1000 hr in duration are planned. In addition to these creep tests, short-time stress-rupture studies will be made to determine the 100-hr rupture strength.

TABLE F-5. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN HIGH-TEMPERATURE WATER AFTER 98 DAYS

Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change[(a)], mg per cm^2	
	Oxygen, ppm	Zirconium, w/o		In 600 F Water	In 680 F Water
10 U	600	0.74	Fabricated	0.54	0.11
	700	0.17	Fabricated	0.72	-11.15
	300	0.02	Fabricated	0.78	-39.9
20 U	600	0.74	Fabricated	0.96	0.67
	700	0.17	Fabricated	1.07	0.54
	300	0.02	Fabricated	0.68	0.87
30 U	600	0.74	As cast	0.78	-0.90
	700	0.17	Fabricated	0.48	-1.08
	300	0.02	As cast	1.25	-2.55
40 U	600	0.74	As cast	0.66	1.54
	700	0.17	As cast	0.57	0.51
	300	0.02	As cast	0.67	1.78
50 U	600	0.74	As cast	0.75	-0.72
	700	0.17	As cast	0.68	-0.48
	300	0.02	As cast	0.42	-4.84
60 U	600	0.74	As cast	-0.99	(b)
	700	0.17	As cast	-0.01	-11.3
	300	0.02	As cast	0.34	-7.76
Zircaloy-2	--	--	--	0.20	0.34

(a) Average of two specimens.

(b) Both specimens taken off test.

Specimens for thermal-conductivity, thermal-expansion, and electrical-resistivity measurements have been prepared and are ready to be tested.

Development of Thorium-Uranium Alloys

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

Development of thorium-uranium-base alloys of improved radiation stability and corrosion resistance is the aim of a program currently in progress. The effect of starting materials, casting methods, and fabrication on the size and distribution of uranium-phase particles is being investigated. Additions of niobium and zirconium and zirconium in conjunction with niobium are being studied. Also, preparation of thorium carbide, thorium-uranium carbides, thorium nitride and thorium-uranium nitrides by arc-melting methods is being studied.

Thorium ternary and quaternary compositions previously studied contained 10 to 25 w/o uranium, 10 to 25 w/o zirconium, and, in some cases, 2 to 4 w/o niobium, with zirconium-to-uranium ratios of 1 or less. Also, thorium-10 and -20 w/o uranium alloys with small additions of either niobium or molybdenum were studied. These compositions were chosen from the standpoint of keeping the thorium content high while maintaining a low alloy content for nuclear reasons. Results of 200 C water-corrosion and hot-hardness tests indicate that these alloys are improved, though not greatly, by increased zirconium content. New thorium-base ternary compositions containing 5 to 20 w/o uranium and 10 to 25 w/o zirconium are now being studied. All have a zirconium-to-uranium ratio of greater than 1. It is hoped that some of these new compositions will yield microstructures that consist of thorium grains completely surrounded by a zirconium-uranium envelope, and thereby impart corrosion resistance. In addition, increased zirconium may impart higher hot strength to these alloys.

The new alloy compositions have been prepared by arc and induction melting and have been hot rolled. Metallographic examination of the as-cast alloys is now under way. Investigation of etching techniques for these alloys has shown that an electrolytic etchant of 49 parts methyl alcohol, 49 parts water, and 2 parts hydrofluoric acid is successful when used at 25 v for 2 to 4 sec.

Further investigation of the new alloys will include corrosion testing in 200 C water and heat treatment at various temperatures to determine the effect of heat treatment on uranium particle size, distribution, and phases present. Hot-hardness measurements will be performed at temperatures up to 900 C to ascertain the high-temperature strength of these alloys. In addition, the recrystallization characteristics of several of these alloys will be determined. Results obtained from these alloys will be correlated with those obtained on alloys previously investigated.

Arc-melted buttons of thorium-uranium carbides have been examined further. The as-cast structure of ThC-60 mole per cent UC was found to be single phase, whereas ThC-40 mole per cent UC exhibited a two-phase structure. Inasmuch as the ThC-UC system is reported to exhibit complete solubility, it is suspected that non-stoichiometric conditions existed.

Since these carbides decompose rapidly under atmospheric conditions, especially at high ThC contents, metallographic examination of thorium-uranium carbides may not be possible. Arc-cast bars of thorium-uranium carbides are now being prepared by skull casting arc-melted buttons into graphite molds.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

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

The objective of this project is to study and understand the important causes of fission-gas release from UO_2. At present, a considerable effort continues to be devoted to completion of equipment for continuous release and collection of gas during irradiation in a beam tube of the Battelle Research Reactor. Equipment is also being readied for diffusion studies to be conducted by postirradiation heating. Meanwhile, considerable effort is also being devoted to the preparation and characterization of UO_2 for these studies.

A series of single crystalline samples has been prepared and characterized for experiments to determine whether diffusion of fission gas occurs by a volume diffusion mechanism. Additional single crystals of UO_2 prepared by other methods are being sought. Studies are in progress on the preparation and characterization of sintered UO_2 for study primarily in the in-pile apparatus. In addition, consideration is being given to methods of preparing fused UO_2 powders which can be used in both the in-pile and out-of-pile diffusion studies. In both the sintered UO_2 and powders for in-pile high-temperature study, the particle size or measured surface areas are being selected so that a large diffusion release of fission gas, compared with surface recoil release, can be expected.

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

Cermets containing 60 to 90 volume per cent ceramic fuel are being investigated as potential fuel-element materials. Fabrication techniques are being developed for the preparation and cladding of cermets consisting of UO_2 fuel dispersed in metal matrices of chromium, molybdenum, niobium, and stainless steel. In comparison to ceramic UO_2 fuel, these materials possess superior structural strength, thermal-shock resistance, and thermal conductivity. A very significant favorable property is the increase in thermal conductivity with increase in temperature, in contrast to UO_2 for which the thermal conductivity decreases with temperature.

The gas-pressure-bonding process is being utilized for the bonding of niobium- and molybdenum-base fuel systems. Techniques for the self-bonding of niobium have been developed, and the process is now being adopted for the preparation of niobium-clad compartmented UO_2 fuel elements. Molybdenum has proven to be more difficult to bond, and studies are continuing to develop optimum conditions for the surface preparation and self-bonding of this material.

A study is being conducted to obtain a basic understanding of the kinetics involved in the bonding of materials by use of temperature and pressure. The results of this investigation are being used as a guide for all of the programs utilizing the gas-pressure-bonding process.

Fabrication of Cermet Fuel Elements

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

Methods of producing cermets of 90 per cent of theoretical density or better containing 60 to 90 volume per cent ceramic fuel are being investigated. Evaluations of fabricated cermets are being made on the basis of microstructure and physical and mechanical properties.

Cermets of 80 volume per cent UO_2-chromium have been hot press forged at 1900 F. The green pellets were pressed at 40 tsi in a 1-in.-diameter die from a powder mix of minus 100 plus 140-mesh Mallinckrodt spherical UO_2 and minus 400-mesh chromium powder. Green densities ranged from 75 to 76.2 per cent of theoretical. The green pellets were assembled in 1/4-in.-thick by 2-in.-square Type 304 stainless steel frames prior to welding on the 1/8-in. stainless steel cover plates. The assembled packs were hot press forged from a furnace at 1900 F to obtain reductions in thickness from 36 to 55 per cent, which gave densities of 83.5 to 95.3 per cent of theoretical, respectively. A reduction in thickness of approximately 45 per cent was necessary to obtain a density of 95 per cent of theoretical. The spherical UO_2 powder gave a microstructure superior to cermets previously prepared using Mallinckrodt high-fired UO_2.

In addition, 80 volume per cent UO_2-molybdenum cermets have been hot press forged at 1900 F to obtain reductions in thickness of 30 to 53 per cent. Minus 100 plus 140-mesh Mallinckrodt spherical UO_2 and minus 325-mesh molybdenum powders were used. The powder mix was pressed at 40 tsi in a 1-in.-diameter die and assembled in Type 304 stainless packs 2 in. square and 1/4 in. in thickness prior to welding on the 1/8-in. stainless steel cover plates. The green-pressed pellets ranged from 75 to 76.3 per cent of theoretical density. After heating to 1900 F, a reduction of 30 per cent in thickness resulted in a crack-free cermet having a density of 85.2 per cent of theoretical. Another assembly hot press forged from a furnace at 1900 F to obtain a reduction of 45 per cent in thickness resulted in a cermet density of 94 per cent of theoretical. Cracks were visible on the cermet surface. Reductions of more than 45 per cent in thickness resulted in UO_2-molybdenum cermets which were broken in several pieces when removed from the hot-press-forge frames.

A cermet rod of 80 volume per cent UO_2-molybdenum was prepared by pressure bonding. One-inch-diameter pellets were pressed at 30 tsi from a UO_2-molybdenum powder mix prior to fitting in a stainless steel tube. The powder mix was composed of 80 volume per cent UO_2 (80 w/o being minus 100 plus 140-mesh high-fired UO_2 and the remainder minus 270 mesh) and 20 volume per cent minus 325-mesh molybdenum. Green-pressed pellets ranged from 72.3 to 72.6 per cent of theoretical density. The stainless steel tube-cermet assembly was sealed and evacuated before pressure bonding for 3 hr at 2300 F under a pressure of 10,000 psi. A cermet density of 86.7 per cent of theoretical density was measured. After pressure bonding an additional 3 hr at 2400 F under 10,000 psi, a cermet density of 91.1 per cent of theoretical was obtained. The cermet rod is being prepared for thermal-conductivity and electrical-resistivity measurements.

Thermal-conductivity measurements have been made on cermet rods of 70 and 80 volume per cent UO_2-Type 302B stainless steel and 70 volume per cent UO_2-molybdenum. The cermet rods were fabricated by pressure bonding green-pressed pellets for 3 hr at 2300 F under a pressure of 10,000 psi. The results are shown in Table F-6 and Figure F-1. It is interesting to note that at higher temperatures the thermal conductivity of the molybdenum cermet is nearly seven times higher than the corresponding value for UO_2, while the stainless steel cermet improves the thermal conductivity by a factor of three. As would be expected, the increase in volume per cent of UO_2 shifts the curve to a lower value. The data have not been correlated with metallographic results in order to explain the shape of the curves, although all the cermets are known to have a continuous metallic skeleton. The difference in shape of the curves for specimens containing molybdenum and those containing stainless steel is probably related to the difference in the effect of temperature on the thermal conductivity of the two metals.

TABLE F-6. THERMAL CONDUCTIVITIES OF CERMET RODS AT TEMPERATURES FROM 100 TO 750 C

	Thermal Conductivity at Temperature Shown, w/(cm)(C)			
Specimen	100 C	400 C	600 C	750 C
70 w/o UO_2-stainless(a)	0.079	0.089	0.109	0.124
70 w/o UO_2-Mo(b)	0.198	0.197	0.197	0.197
80 w/o UO_2-stainless(c)	0.063	0.069	0.081	0.093
UO_2(d)	0.075	0.044	0.034	--

(a) 97 per cent of theoretical density 70 volume per cent UO_2-stainless steel cermet.

(b) 91.7 per cent of theoretical density 70 volume per cent UO_2-molybdenum cermet.

(c) 95.5 per cent of theoretical density 80 volume per cent UO_2-stainless steel cermet.

(d) UO_2 is 95.3 per cent of theoretical density. Unpublished BMI data.

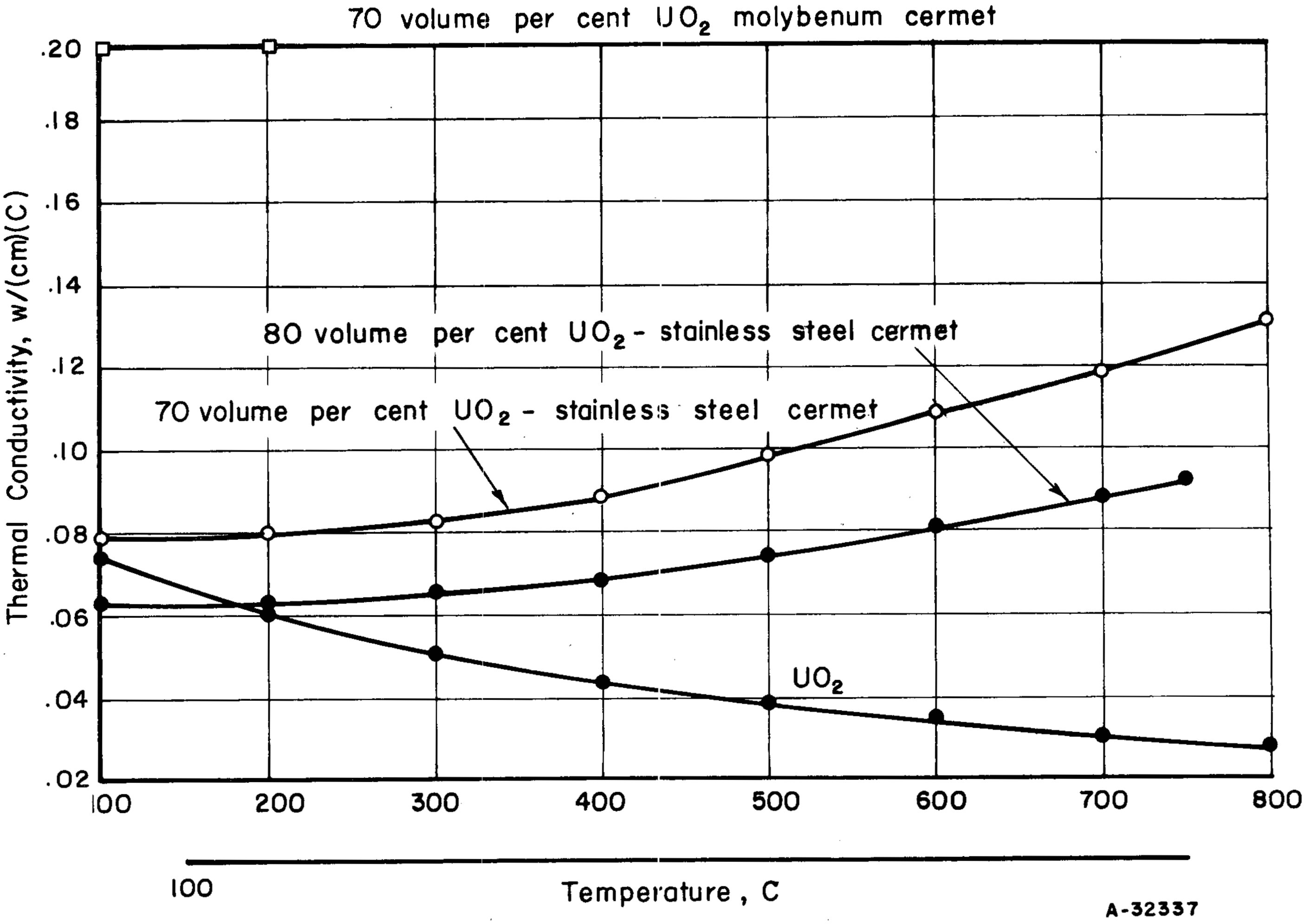

FIGURE F-1. THERMAL CONDUCTIVITY OF UO_2-BEARING CERMETS

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

S. J. Paprocki, E. S. Hodge, and P. J. Gripshover

The gas-pressure-bonding technique is being investigated as a possible method for fabricating molybdenum- and niobium-clad ceramic and cermet-type fuels. The high-temperature strength and favorable nuclear properties of molybdenum and niobium make them promising structural materials for use in high-temperature inert-gas-cooled and some liquid-metal-cooled reactors.

The optimum surface preparation for self-bonding of niobium at 2100 F for 3 hr at 10,000 psi was determined to be a pickling solution of 65 parts nitric acid and 35 parts water. Although this surface preparation produced excellent bonds with a slight amount of bond-line impurities, it was necessary to optimize the pickling operation to produce the least amount of bond-line contaminants. Investigations revealed that the best self-bonding of niobium was achieved by pickling the niobium surfaces for 1 min in the acid bath at a temperature of 120 F. Longer times and lower pickling temperatures tended to produce more bond-line contamination. With the optimum pickling operations it is possible to obtain excellent bonds with a negligible amount of bond-line contamination.

A compartmented niobium-clad UO_2 fuel plate was pressure bonded to determine if the optimum surface preparation and bonding parameters developed for simple flat-plate specimens are satisfactory for more complex configurations. This fuel plate contained three uranium dioxide cores measuring 0.040 by 0.870 by 0.870 in. The cores were placed in a pieced-component receptacle plate made up of six individual strips of niobium; 0.010-in.-thick cover plates were used on the top and bottom of the assembly. The specimen was pressure bonded at 2100 F and 10,000 psi for 3 hr. Metallographic examination of this fuel plate showed good bonding with only a small amount of contamination present in most areas.

The study of barrier layers to prevent reaction between niobium-clad fuel plates and protective bonding containers was continued. Early studies indicated that window glass was a satisfactory material for bonding temperatures up to 2100 F. Recent results, however, revealed that a slight reaction occurs with the niobium at this bonding temperature. In an attempt to determine if the impurities in the glass produced the reaction, specimens were bonded with pure quartz as a barrier layer. Examination of these specimens revealed that more alloying had occurred with the pure quartz than with the window glass. The oxygen content of the niobium was raised from 200 ppm before bonding to 1200 ppm after bonding, while the silicon was observed to have increased in a similar manner.

During previous bonding studies with niobium it was determined that hydrogen was introduced into the niobium during bonding with impure autoclave-gas atmospheres. These specimens were always brittle after bonding. It has been recently determined that the initial ductility of these specimens can be restored by heat treating for 2 hr in a vacuum at 2000 F to remove the hydrogen.

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The molybdenum-clad specimens are normally brittle after bonding at temperature. Since this lack of ductility may be due to reaction of the molybdenum with the bonding container, it appears necessary to develop a method of edge welding to form a bonding envelope with the molybdenum cladding similar to that employed with the niobium fuel plates. Additional edge-welded specimens are being prepared by arc-fusion edge welding in a controlled atmosphere. It is difficult to consistently weld without embrittling the molybdenum; consequently, a second method is being investigated for forming an edge-welded assembly. In this technique a 0.005-in.-thick shim of zirconium is inserted into a channel ground around the periphery of the two flat plates of molybdenum to act as a braze material. It is planned to bond this specimen at 1800 F and 10,000 psi for 3 hr in a bonding container. The container will then be removed and, if a gastight assembly has been formed, it will be rebonded at 2300 F and 10,000 psi for 3 hr to achieve a molybdenum-to-molybdenum bond. It is known that the initial bonding step will be achieved without introducing any contamination into the molybdenum cladding. The second bonding operation will be carried out without benefit of a protective container; consequently, contamination should not be introduced during this bonding cycle if the autoclave atmosphere is properly controlled.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

Sufficient qualitative information has been obtained to propose critical experiments to determine the kinetics of the process whereby similar metals are bonded in the solid state by the application of heat and pressure. Most of the work will be conducted over the temperature range 0.5 to 0.75 times the absolute melting temperature and at pressures up to 25,000 psi. A limited number of experiments will be run up to 0.9 times the absolute melting temperature at pressures to 5000 psi.

The elimination of macroporosity to form interfaces in which the metal surfaces are in intimate contact will be studied by bonding OFHC copper specimens at pressures up to 24,000 psi at temperatures of 800 to 1150 F for periods of 10 min. Although it has been shown that initial deformation is very rapid and that if sufficient pressure is applied intimate contact will be quickly achieved, it is not known to what extent the voids will be closed in periods of several hours when the applied pressure is not sufficient to eliminate the voids initially. Therefore, a second series will be run for a period of several hours to determine this effect.

In order to study the effect of pressure on the bond line, a series of standard specimens of OFHC copper annealed at 1400 F will be bonded by applying 24,000 psi for 10 min at 1150 F. This procedure should provide specimens in which the two metal interfaces are in intimate contact but in which there is no grain growth across the interface. Also, it should provide specimens in which the grain size is essentially stabilized so that no grain growth will occur at lower temperatures unless the temperature is sufficiently low for simultaneous cold work and recrystallization to occur. The bond line will be evaluated on the basis of metallographic examinations made before and after a high-temperature anneal. This anneal will be given the specimens after the low-temperature pressure anneal.

Factors such as recrystallization, radius of curvature, thickness of the bond line, and impurities which affect grain growth will be evaluated separately.

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, and J. B. Fox

This program is concerned with the development of a fabrication process which will maintain or improve fuel-element quality, while reducing manufacturing costs. The gas-pressure-bonding technique was selected as the most promising fabrication method for achieving these objectives. The ceramic, cermet, and dispersion fuel systems were selected because it is believed that these fuel systems offer the greatest potential for achieving a high burnup over a range of temperatures. Also, the gas-pressure-bonding process appears capable of making a substantial reduction in the fabrication cost of these fuels while improving their performance. The study is directed toward the refinement and further development of the gas-pressure-bonding process to accomplish simultaneous densification and cladding of these fuels with stainless steel.

The greatest emphasis will be placed on the development fuel elements and assemblies of various shapes made of ceramic uranium dioxide clad with Type 304 stainless steel. The techniques developed for the ceramic uranium dioxide fuel can be applied to the uranium dioxide cermet and dispersion systems with only a small amount of additional development work.

Initial studies are concerned with utilization of high-density green-compacted UO_2 ceramic cores, in place of sintered cores, to achieve specific densities while further reducing fabrication costs by eliminating the high-temperature sintering operation. A minimum UO_2 green-pressed density of 70 per cent of theoretical was selected in order to prevent excessive deformation of the stainless steel cladding during pressure bonding.

The compacting characteristics of six types of uranium dioxide have been reported. These powders included a Mallinckrodt ceramic grade, dense ceramic grade, high-fired grade, special dense grade, and spherical grade and a NUMEC high-fired grade. Compacting studies were continued on a seventh type of uranium dioxide powder, a Spencer fused grade. Both minus 20-mesh (as received) and selected minus 100-mesh powders were used for comparison. It was shown that the minus 20-mesh material reached a maximum density of 88.9 per cent of theoretical as compared with 80.7 per cent of theoretical for the smaller particle sizes. The results of the cold-compacting studies for the Spencer fused UO_2 are listed in Table FF-1.

TABLE FF-1. GREEN COMPACTIBILITY OF SPENCER FUSED UO_2 POWDERS USING CEREMUL "C" AS A BINDER AND A HEIGHT-TO-DIAMETER RATIO OF 0.9

Compact	Mesh Size	Green Density, g per cm^3	Density, per cent of theoretical	Pressure, tsi
U-1154	-100	8.64	78.9	40
U-1155	-100	8.68	79.3	45
U-1156	-100	8.82	80.6	50
U-1157	-100	8.85	80.7	55
U-1158	-20	9.75	88.9	55
U-1159	-20	9.70	88.6	50
U-1160	-20	9.61	87.9	45
U-1161	-20	9.62	87.9	40
U-1162	-20	9.50	87.2	35
U-1163	-20	9.38	85.6	25

The Spencer fused UO_2 (as received) gave a tap density of 64.4 per cent of theoretical (7.06 g per cm^3) as compared to a previous high of 63.3 per cent (6.94 g per cm^3) reported for MCW special dense UO_2.

Six rod-type specimens containing 22 MCW special dense UO_2 compacts green pressed at 50 tsi have been pressure bonded. The pressure-bonding containers for these specimens were fabricated from Type 304 stainless steel tubes having an inside diameter of 0.540 in. and a wall thickness of 0.020 in. The tubes were of various lengths, depending upon the number of compacts contained in each. The specifications for these green compacts, the gas-pressure-bonding conditions, and the final densification of the compacts are given in Table FF-2. Evaluations of these specimens reveal that in general the pressure-bonding variables used gave a fairly constant density range, except where the temperature was increased greatly. It appears from this evaluation that the achieving of specific densities will for the most part be through the selection of various types or combinations of oxides, not by changing bonding parameters.

Other rod-type specimens containing compacts made from Spencer fused UO_2 are being assembled for pressure bonding. These specimens contain compacts with initial green densities as high as 88.9 per cent of theoretical.

A single tube having the same dimensions as those used for the rod-type specimens was alternately partially filled with MCW special dense UO_2 and lightly tamped by hand until the tube was completely filled. The UO_2 density was approximately 77 per cent of theoretical. The specimen was pressure bonded at 2100 F for 2 hr at 10,000 psi. The resulting specimen was heavily deformed but very uniform throughout. Radiography demonstrated that the core was uniform throughout and there were no indications of cleavage or other defects. The resultant core density was 86 per cent of theoretical. Additional specimens of this type are planned in which selective sizing of the particles is used and in which pressure-bonding conditions are changed.

The establishment of a range of optimum gas-pressure-bonding conditions of time, temperature, and pressure for obtaining acceptable bonds with Type 304 material

TABLE FF-2. DENSIFICATION OF MCW SPECIAL DENSE UO_2 BY VARIOUS PRESSURE-BONDING CONDITIONS(a)

Compact	Height, in.	Green-Pressed Density		Pressure-Bonded Density(b)	
		G per Cm^3	Per Cent of Theoretical	G per Cm^3	Per Cent of Theoretical
Tube 9, Pressure Bonded at 2200 F for 1 Hr at 10,000 PSI					
U-1132	0.459	8.74	79.9	9.24	84.3
U-1133	0.459	8.74	79.9		
U-1134	0.460	8.71	79.6		
Tube 10, Pressure Bonded at 2100 F for 1.5 Hr at 10,000 PSI					
U-1135	0.455	8.80	80.4	9.15	83.5
U-1136	0.456	8.79	80.3		
U-1137	0.459	8.74	79.9		
Tube 11, Pressure Bonded at 2200 F for 3 Hr at 5,000 PSI					
U-1138	0.457	8.76	80.1	9.19	83.9
U-1139	0.458	8.75	80.0		
U-1140	0.458	8.75	80.0		
Tube 12, Pressure Bonded at 2100 F for 1.5 Hr at 10,000 PSI					
U-1141	0.456	8.79	80.3	9.16	83.6
U-1142	0.456	8.79	80.3		
U-1143	0.457	8.78	80.2		
Tube 13, Pressure Bonded at 2300 F for 1 Hr at 10,000 PSI					
U-1144	0.457	8.78	80.2	9.69	88.4
U-1145	0.458	8.75	80.0		
U-1146	0.453	8.80	80.4		
Tube 14, Pressure Bonded at 2100 F for 1.5 Hr at 10,000 PSI					
U-1147	0.456	8.79	80.3	9.18	83.76
U-1148	0.456	8.79	80.3		
U-1149	0.456	8.79	80.3		
U-1150	0.455	8.80	80.4		
U-1151	0.456	8.79	80.3		
U-1152	0.456	8.79	80.3		
U-1153	0.457	8.76	80.1		

(a) All compacts were green pressed using a Ceremul "C" binder to an initial density using 50 tsi of pressure. The compacts were 0.539 in. in diameter with a height-to-diameter ratio of 0.9.

(b) The over-all density of the compacts contained in a tube was calculated by measurement of the tube after bonding and knowing the weight of UO_2 in each tube.

has been developed. It was desired to have a selection of bonding parameters along with a selection of oxides to achieve almost any desired density.

Cylindrical 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 were pressure bonded at different bonding conditions. Two types of specimens were bonded at the same conditions. In one, the rods were butted directly together in the tube; in the other two 0.005-in. disks of as-rolled Type 304 stainless steel were placed between the two rods. This gave a comparison of machined surfaces versus as-rolled surfaces.

The components were put through a standard wash cycle after being pickled in 10 volume per cent nitric acid-2 volume per cent hydrofluoric acid aqueous solution and before assembly. The tubes were evacuated and sealed.

After the gas-pressure-bonding cycle, each bonded assembly was machined into a standard 1/4-in.-round tensile-test specimen. The load was applied along the specimen axis in a screw-type tensile machine. The applied load was gradually increased until the specimen broke. The ultimate strength of the pressure-bonded Type 304 stainless steel and the bond strength were obtained by this test. The tensile-test results as a function of bonding parameters and surface conditions are given in Table FF-3. The bonds were also evaluated metallographically after the test. The evaluation of these test results shows that the best conditions for strong, metallurgically bonded specimens were as follows:

Time, hr	Temperature, F	Pressure, psi
3	2000	10,000
1-1/2	2100	10,000
3	2100	10,000

Further refinement of the surface-preparation techniques may be required for the obtaining of strong, consistent stainless steel bonds. Parting at or near the bond line did occur more often with the specimens containing the rolled disks. However, the ultimate load required to break the machined-surface and as-rolled-surface specimens compared favorably. Except for the specimens bonded at 2200 F for 3 hr, the ultimate breaking load for pressure-bonded Type 304 stainless steel compared favorably with that of wrought material. It appears that the pressure-bonding conditions of temperature and time may be limited in order to maintain a strong structural cladding.

TABLE FF-3. TENSILE-TEST RESULTS FOR PRESSURE-BONDED TYPE 304 STAINLESS STEEL BOND SPECIMENS

		Pressure-Bonding Conditions					
Specimen	Type and Condition	Time, hr	Temperature, F	Pressure, psi	Approximate Yield Point, lb	Ultimate Load, lb	Comments
1	Machined from Type 304 stainless steel stock	--	--	--	2000	4460	Used as a comparison for the pressure-bonded specimens
2	Butted rods with rms surfaces of 115 to 135	3	2100	10,000	1700	4100	Specimen necked down and broke at a point away from the bond; complete grain growth
3	Butted rods with rms surfaces of 110 to 130	3	2100	10,000	1700	4100	Specimen necked down and broke at a point away from the bond; complete grain growth
4	Butted rods with rms surfaces of 120 to 150	1-1/2	2100	10,000	1700	4160	Specimen necked down and broke at a point away from the bond; complete grain growth
5	Machined from Type 304 stainless steel stock after the pressure-bonding cycle	1-1/2	2100	10,000	1700	4100	Used as a comparison for the bond-test specimens
6	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	3	2000	10,000	2000	4240	Specimen did not neck down all the way but parted between the disk and rod on one end; pieces of the disk were pulled away, leaving a jagged surface; complete grain growth across the disks and 70 per cent grain growth across the rod-to-disk interfaces
7	Butted rods with rms surfaces of 50 to 60 with two 0.005-in. disks in between	3	1900	10,000	1620	3500	Specimen did not neck down all the way but parted between the disk and rod on one end; no metal was torn from either surface; 10 per cent grain growth between the disks
8	Butted rods with rms surfaces of 5 to 60	3	1900	10,000	1600	3750	Specimen did not neck down all the way but parted at the bond interface; small pieces of metal were torn from both surfaces
9	Butted rods with rms surfaces of 5 to 60	3	2000	5,000	1520	3640	Specimen did not neck down all the way but parted at the bond interface only; small pieces of metal were torn from both surfaces
10	Butted rods with rms surfaces of 50 to 60 with two 0.005-in. disks in between	3	2000	5,000	1450	3700	Specimen did not neck down all the way and parted between the disks; there appeared to be no metal torn from either surface; approximately 30 per cent grain growth between the disks and rods

TABLE FF-3. (Continued)

		Pressure-Bonding Conditions			Approximate		
Specimen	Type and Condition	Time, hr	Temperature, F	Pressure, psi	Yield Point, lb	Ultimate Load, lb	Comments
11	Butted rods with rms surfaces of 50 to 60	1	2100	5,000	1420	3800	Specimen parted at the bond interface and did not neck down all the way; no metal appeared to be torn from either surface
12	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	1	2100	5,000	1444	3440	Specimen did not neck down all the way but parted at the bond interface between the disks; no metal appeared to be torn from either surface; approximately 10 per cent grain growth between the disks and rods
13	Butted rods with rms surfaces of 30 to 40	1	2100	10,000	1580	4060	Specimen necked down all the way and broke away from the bond; approximately 90 per cent grain growth
14	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	1	2100	10,000	1630	4100	Specimen did not neck down all the way but parted at the bond interface, between the disk and rod on one end; metal pulled away from both surfaces, leaving a jagged face; approximately 80 to 90 per cent grain growth between the disks and 50 per cent grain growth between the disks and rods
15	Butted rods with rms surfaces of 30 to 40	1	2200	5,000	1620	4120	Specimen necked down all the way but broke at the bond interface; large chunks of metal were torn from both surfaces
16	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	1	2200	5,000	1520	4170	Specimen necked down part way and parted at the bond interface between the disks; metal pulled away from both surfaces, leaving a jagged surface; approximately 80 per cent grain growth between the disks and rods
17	Butted rods with rms surfaces of 30 to 40	3	2100	5,000	1600	4100	Specimen necked down all the way and broke away from the bond; approximately 95 per cent grain growth

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TABLE FF-3. (Continued)

Specimen	Type and Condition	Pressure-Bonding Conditions			Approximate Yield Point, lb	Ultimate Load, lb	Comments
		Time, hr	Temperature, F	Pressure, psi			
18	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	3	2100	5,000	1680	4090	Specimen did not neck down all the way but broke at the bond interface between the disks; metal was pulled from both surfaces, leaving a jagged surface; approximately 95 per cent grain growth between the rods and disks
19	Butted rods with rms surfaces of 30 to 40	1/2	2200	10,000	1560	4090	Specimen did not neck down all the way but parted at the bond interface; small pieces of metal were pulled from both surfaces
20	Butted rods with rms surfaces of 30 to 40 with two 0.005-in. disks in between	1/2	2200	10,000	1580	4070	Specimen did not neck down all the way but parted at the bond interface between the disks; small pieces of metal were pulled from both surfaces; approximately 30 per cent grain growth between the disks and rods
21	Butted rods with rms surfaces of 20 to 30	3	2200	2,000	1430	3840	Specimen necked down all the way and broke at a point away from the bond; complete grain growth
22	Butted rods with rms surfaces of 40 to 50 and 30 to 40 with two 0.005-in. disks in between	3	2200	2,000	1480	3870	Specimen necked down all the way and broke at a point away from the bond; complete grain growth

FF-8

DEVELOPMENT OF URANIUM CARBIDE-TYPE FUEL MATERIALS

F. A. Rough and W. Chubb

Uranium monocarbide and its related alloys are being developed as a portion of the AEC Fuel-Cycle Development Program. These materials represent an alternate ceramic-type reactor fuel to uranium dioxide, and they may have distinct advantages with respect to cost, density, irradiation resistance, and ease of reprocessing. The discovery that uranium carbides can be cast from the molten state into geometric shapes of almost 100 per cent of theoretical density is believed to be an important factor with regard to the cost of fuel elements made from ceramic materials. The present program of research represents an outgrowth of this discovery. The program includes five related areas of research: (1) development of chemical and physical techniques for preparing carbides primarily by powder metallurgical reaction and pressing techniques, (2) development of melting techniques for preparing larger and higher quality carbide castings, (3) investigation of the chemical, physical, and mechanical properties of castings of uranium carbide, (4) determination of diffusion rates in uranium carbides, and (5) investigation of the nature and mechanism of irradiation-damage processes in uranium carbides.

During the first 4 months of this program, data have been reported on the effects of impurities upon the casting behavior, strength, and corrosion resistance of uranium monocarbide. The variation of the density and of the electrical resistivity of uranium-carbon alloys has been reported as a function of carbon content. Diffusion data for the interdiffusion of uranium and carbon in uranium carbides have been obtained at temperatures from 1600 to 1980 C, and a preliminary estimate of the activation energy for diffusion has been calculated.

During the past month, additional pressing and sintering studies have resulted in one pellet of 95 per cent of the theoretical density of uranium monocarbide. This pellet was produced by sintering crushed UC powder containing 4.85 w/o carbon for 30 min at 2120 C. Melting studies show that most alloy additions tend to reduce the castability and thermal-shock resistance of uranium monocarbide. Development of skull-melting techniques has resulted in the production of carbide castings 1 in. in diameter and up to 8 in. long. Metallographic examination of other castings shows that UO_2 reacts with molten UC to produce uranium metal and CO. Nitrogen reacts with molten UC_2 plus the solid solution, U(C,N). Annealing of cast uranium monocarbide for 1 hr at 1500 C increases the transverse rupture strength from less than 25,000 psi to more than 40,000 psi. Compatibility tests show that uranium monocarbide reacts with Inconel and stainless steel in less than 24 hr at 1200 C.

FF-9

Alternate Fabrication Methods for UC

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

An investigation of methods for producing dense UC cores is being made. Solid-state reactions of uranium or uranium oxides with carbon are being studied as methods of producing UC. Also, sintering studies are being conducted on carbide powders of varying carbon composition. The quality of the material produced as well as the economics of mass production are being considered as bases for evaluation.

The uranium-carbon reaction is being studied by sintering cold-pressed compacts of mixed uranium and graphite powders. For one series, AGOT-type graphite and 95 w/o uranium powder were weighed and blended for 2 hr in a V-type mixer prior to removal from the dry box. Methyl alcohol was immediately added to the mix to help prevent oxidation and to act as a binder during compaction. Cores pressed from the mix at 40 tsi in a 1/3 by 2/3-in. die had a density of 8.15 g per cm^3. After reacting in vacuum for 2 hr at 1100 C, the density was 7.02 g per cm^3. An additional heat treatment of 4 hr at 1100 C resulted in a density of 7.00 g per cm^3. Microscopic examination of the core showed agglomeration of the graphite which was probably a result of the addition of alcohol after mixing. Etching of the specimen indicated the formation of a layer of UC around the graphite particles. Additional cores are being prepared for reaction using various mixing techniques, different types of carbon, and hot consolidation prior to sintering.

In addition, a mix of the stoichiometric composition of UC was prepared from powders of UC_2 (8.74 w/o carbon) and minus 325-mesh uranium metal. A pellet pressed at 40 tsi from the powder mix had a density of 9.11 g per cm^3. After vacuum reaction for 2 hr at 1100 C, the density was 8.37 g per cm^3. A density of 8.34 g per cm^3 was measured after an additional 4-hr treatment at 1100 C. Microscopic examination indicated a dispersion of unreacted UC_2 in uranium. However, etching revealed the presence of UC along crystallographic planes in the UC_2 particles. Additional pellets are being prepared for reaction studies.

Sintering studies have been conducted on arc-cast and crushed UC powder of 4.70 and 4.85 w/o carbon. Pellets were prepared by removing the powder from the dry box and covering it with a solution of camphor in methyl alcohol. The alcohol was allowed to evaporate until the powder was only slightly moist when pressed at 40 tsi. The pellets were vacuum sintered for 90 min at temperatures ranging from 1730 to 2000 C. In addition, a pellet of each composition of UC powder was vacuum sintered for 30 min at 2120 C. Results of sintering studies are shown in Table FF-4. Additional pellets of UC made by various methods are being prepared for sintering. Also, investigations are being made on the preparation of UC powder by the uranium metal-methane reaction.

TABLE FF-4. DENSITY OF GREEN AND AS-SINTERED UC CORES

Specimen[b]	Density[a], per cent of theoretical		Sintering Conditions	
	Green	Sintered	Time, min	Temperature, C
A-1	68.2	69.8	90	1730
B-1	70.6	66.2	90	1730
A-2	68.7	76.0	90	1800
B-2	65.6	73.3	90	1800
A-3	68.5	80.6	90	1900
B-3	66.9	77.2	90	1900
A-4	64.8	80.5	90	2000
B-4	67.3	81.3	90	2000
A-5	68.5	94.9	30	2120
B-5	69.3	88.3	30	2120

(a) Per cent of theoretical density based on densities of 13.71 and 13.60 g per cm^3 for "A" and "B" specimens, respectively.

(b) Specimens prefixed by the letter "A" have UC of 4.70 w/o carbon; specimens prefixed by "B" have 4.85 w/o carbon.

Melting and Casting Techniques for Uranium-Carbon Alloys

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

Uranium carbide is considered to be a potentially desirable fuel material because of its high uranium density, melting point, and thermal conductivity. Previous work has demonstrated that high-density uranium carbide shapes can be obtained by melting and casting. An attempt is being made to develop reliable techniques for the production of high-quality shapes of uranium carbide. Methods of evaluation such as chemical analysis, radiography, resistivity measurements, ultrasonic testing, and metallographic examination will be used in an attempt to provide an accurate evaluation of the materials produced. To the extent possible, commercial sources will be used to provide UC of different qualities. Materials produced by solid-state reaction, gaseous reaction, and direct alloying will be evaluated to determine the effects of their variable compositional qualities on casting.

Hydrogen, iron, nitrogen, oxygen, silicon, and tungsten have been added to uranium monocarbide to evaluate the effect of these contaminants upon the castability of the material. It was observed that as little as 200 ppm silicon made casting extremely difficult. The other impurities had no noticeable effect on the castability of uranium monocarbide in amounts up to 800 ppm.

During the last month additions of 1, 10, and 50 a/o Al_4C, Ce_3C_2, Mn_3C, Mo_2C, NbC, TaC, TiC, VC, and ZrC were made to uranium monocarbide. Observations made during inert-electrode arc melting indicate that all additions significantly decrease the thermal-shock resistance of uranium monocarbide in that all the 100-g button castings broke during cooling. This phenomenon is rarely observed during the production of unalloyed UC using the same process. The zirconium carbide additions made the formation of a button difficult. In this case, as the alloy became molten, it flowed away from the point where the arc impinged on the charge and up the sides of the crucible, to give a dish-shaped form. Attempts will be made to cast the alloys into cylindrical shapes, and further observations as to the castability of the alloys will be made. Another experiment is planned in which a charge composed of UO_2 and carbon will be melted together using a carbon electrode in a dynamic inert atmosphere. The reduction of the UO_2 to UC is anticipated; the nature of the reaction and the final quality of the product are points of interest.

Work on the use of the inert-electrode skull arc-melting technique for melting of UC has been initiated. Four 1-in.-diameter castings 4 to 8 in. long have been produced. The first casting was poured into an unheated graphite mold, while the succeeding three were poured into molds at approximately 1400, 1700, and 2000 F. The surface quality of the castings improved with increasing mold temperature. Additional melts involving increased mold temperatures, varied carbon contents, and both vacuum and an inert atmosphere are planned.

Metallurgical and Engineering Properties of Uranium Monocarbide

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

The property studies in progress on uranium carbides are concerned with both determining and improving the properties of these materials. Variables such as impurity content, carbon content, and heat treatment and their effects on density, resistivity, thermal conductivity, and corrosion in various media are being investigated.

Hydrogen, iron, nitrogen, oxygen, silicon, and tungsten have been selected as the elements most likely to be inadvertently introduced as contaminants into UC during normal processing procedures. Cylindrical specimens 3/8 in. in diameter by 1-3/4 in. long were cast containing deliberate additions of these elements. When UO_2 was added to UC melts, metallographic examination of the specimens produced showed the presence of metallic uranium in the carbide grain boundaries and as spheroidal particles within the grains. Alloys containing 2 w/o or more carbon, when melted in a nitrogen atmosphere, exhibited two distinct phases believed to be U(C,N) and UC_2. Hydrogen present in the melting atmosphere had little effect on specimens containing greater than the stoichiometric amount of carbon (4.8 w/o carbon). Specimens containing less than 4.8 w/o carbon, when melted under hydrogen, decomposed to powder following contact with a metallographic etching solution. Analyses are being taken to determine the amount of hydrogen retained in these specimens. A silicon addition of 200 to 800 ppm to uranium monocarbide caused the formation of a grain-boundary phase. The composition of this phase is unknown. Similar amounts of tungsten or iron added to uranium monocarbide had no noticeable effect on the microstructure.

Typical specimens were selected from the above group and annealed for 1 hr at 1500 C. Density and rupture strength were determined before and after annealing, and the results are presented in Table FF-5. The rupture strengths of all specimens, except one containing silicon, increased significantly. The densities of the specimens did not change significantly. Further examinations of these alloys are in progress in an attempt to determine if the variations noted can be related to the impurity additions, the formation of U_2C_3, or to some other effect.

TABLE FF-5. EFFECT OF HEAT TREATMENT AT 1500 C FOR 1 HR ON DENSITY AND RUPTURE STRENGTH OF URANIUM MONOCARBIDE

Carbon Content of Base (Balance Uranium), w/o	Alloy Addition, ppm	As-Cast Properties		Annealed Properties	
		Density, g per cm^3	Rupture Strength, psi	Density, g per cm^3	Rupture Strength, psi
4.8	300 Fe	13.67	21,000	13.56	50,800
4.8	300 Si	13.49	13,700	13.46	9,700
5.0	300 Si	13.26	4,300	13.35	43,800
4.8	300 W	13.56	1,100	13.49	41,200
5.0	300 W	13.49	9,900	13.29	26,800
5.0	200 N	13.45	9,900	13.52	36,400
4.8	200 O	13.62	16,600	--	58,900
4.8	None	13.67	24,200	13.67	51,700

Compatibility tests involving uranium monocarbide enclosed in tubes of mild steel, stainless steel, Inconel, copper, molybdenum, and tantalum are in progress. All these metals appear to be compatible with carbide at 1000 C after 24 hr. Molybdenum, tantalum, and mild steel appear to be compatible with carbide at 1200 C after 24 hr, while Inconel and stainless steel tubes are destroyed by reaction with the contained carbide at 1200 C within 24 hr. These tentative results have been obtained from visual and radiographic examinations of the test capsules. Metallographic examination is in progress to determine if reactions have taken place which were undetected by X-ray examination. Additional compatibility tests with aluminum, beryllium, magnesium, niobium, and zirconium are in progress or planned.

One-inch-diameter carbide specimens are being prepared for thermal-conductivity and specific-heat measurements as a function of carbon content. Mechanical and physical testing of alloys containing additions of the refractory carbides are also in progress.

Uranium Monocarbide Diffusion Studies

W. Chubb and F. A. Rough

The diffusion rates of uranium and carbon in uranium monocarbide are being investigated in order to obtain a better understanding of the physical and mechanical behavior of this material. During the past few months experiments have been performed to obtain an estimate of the rates of interdiffusion of uranium and carbon in uranium carbides. The estimates have been made by measuring the thickness of the carbide layers formed between molten uranium and graphite. In addition, techniques are being developed for measuring the rate of self-diffusion of uranium in uranium monocarbide.

FF-13

During the last month two final interdiffusion melts at 1600 C were completed, and the resultant graphite-uranium products have been sectioned. Measurements on these melts will be reported next month. Three melts prepared previously have been resectioned and measured at a point about 1 in. below the original section, which was about 1/2 in. from the top of the melt. The carbide layers at 1-1/2 in. from the top of the melt are approximately twice as thick as the layers 1/2 in. from the top of the melt. Since erosion would produce an effect opposite to that observed, it must be assumed that the thickening effect is a temperature effect and that erosive effects are negligible. If it is assumed that the temperature was 100 C hotter at the 1-1/2-in. level than at the 1/2-in. level, then the results obtained at the two levels are in excellent agreement.

Data reported in the literature* for similar experiments performed at 1200 to 1400 C have been analyzed in terms of diffusion rates, and the results when extrapolated show good agreement with the results obtained at 1600 C.

Sectioning of the graphite crucibles containing uranium and uranium carbides has proven to be very difficult. Diamond cutoff wheels tend to seize in the uranium-uranium carbide mixture so that their use cannot be recommended. An Alundum cutoff wheel has been found to operate satisfactorily, but slowly, under these conditions. An Alundum cutoff wheel cuts pure uranium carbide, but in the absence of appreciable metallic uranium a diamond cutoff wheel cooled by kerosene will cut uranium carbide about three times faster than Alundum.

An attempt to bond UC to UC with a metallic uranium foil as a bonding agent resulted in oxidation of the foil and internal oxidation of the UC near the surface. This oxidation took place inside a graphite bottle in a helium atmosphere. The samples of UC were bonded, however, apparently by a layer of oxide. The results show that UO_2 and UC are compatible with one another and relatively insoluble in one another at 1400 C and 1 atm. This experiment will be repeated using greater precautions to protect the system from oxidation.

Future work in addition to completion of the above-mentioned studies will involve testing of chemical and radiochemical techniques for sampling and analyzing uranium carbide for uranium-235 and total uranium. It is expected that samples will be obtained by grinding on silicon carbide in the presence of nitric acid, and that the samples will be analyzed by radiochemical techniques for total uranium and uranium-235. Standards for these analyses are being prepared. Depleted uranium for preparation of diffusion couples has been on order for several months, but no delivery date has been quoted.

Irradiation Effects in UC

A. E. Austin and C. M. Schwartz

The effects of neutron irradiation and of fission upon the physical properties and structure of UC are being studied. For the determination of crystal structural properties of irradiated material, work is in progress on modification of apparatus for obtaining the necessary X-ray diffraction data.

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*Trans. AIME, 215, 553 (August, 1959).

FF-14

Low-angle X-ray scattering offers promise of detecting the formation of small voids or gas bubbles. To test this method, alloys of ZrC-UC are needed in order to reduce the X-ray absorption to acceptable levels. Arc melts were made of 90 w/o ZrC-10 w/o UC, and 60 w/o ZrC-40 w/o UC. These were found to be two-phase, consisting of a solid solution of the monocarbides and a body-centered metal solid solution. Additional arc melts are being prepared with increased carbon content in an attempt to provide a single-phase carbide solid solution.

GG-1 and GG-2

GG. VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

D. V. Grillot, D. A. Dingee, and J. W. Chastain

Subcooled-boiling heat-transfer research is being conducted for the Euratom-U. S. Joint Research and Development Board. During the past month effort continued on construction and instrumentation of the flow loop for the experimental studies. The detailed experimental program for the research was analyzed.

Based on preliminary studies, a praseodymium-144 β-ray source and Geiger tube detector will be used to evaluate the boiling-void content within the heated channels. A radiation source has been ordered. In addition, all major temperature, power, and pressure detectors have been ordered to complete the heat-transfer-loop instrumentation.

The experimental program will investigate subcooled boiling as a function of flow, pressure, heat flux, and fluid conditions at the inlet to the test section. The region of subcooled boiling has been defined, for the present research, as that portion of a heated channel between the points where the heat-transfer-surface temperature and the bulk fluid reach saturation temperature. In equation form this is given approximately by

$$L = \frac{Wc_p}{ph},$$

where

L = subcooled boiling length

W = mass flow rate

c_p = fluid heat capacity

p = heat-transfer-surface area per unit length

h = nonboiling heat-transfer coefficient.

Based on this definition, the inlet subcooling to the test section is being delineated for a range of loop pressure, flow, and heat-flux conditions to provide the maximum range of subcooled boiling within the test section.

It is anticipated that all long-lead-time materials and instrumentation required to complete the heat-transfer loop will be received during the coming month. Trial checkout tests will be performed.

H. PHYSICAL RESEARCH

F. A. Rough

A study of the reaction of niobium and nitrogen, supported by the AEC Division of Research, is reported this month. Present efforts are directed toward establishing the effects of nitrogen pressure upon the reaction kinetics and toward clarifying erratic kinetic results observed below 700 C.

Niobium-Gas Reactions

W. M. Albrecht and W. D. Goode

A fundamental study of the reactions of nitrogen and niobium is being made. The study consists of determinations of reaction kinetics and mechanisms and includes determination of diffusion coefficients and solubility of nitrogen in niobium.

The reaction-rate and diffusion studies made at atmospheric pressure in the range 675 to 1600 C have been completed. A topical report describing the results has been prepared.

Experiments are continuing to determine the effect of nitrogen pressure on the reaction rates. As previously reported, results indicate no change in the rate with pressure in the range 1/4 to 1 atm at 875 C. Experimental data are now being obtained at 1/8 atm at 875 C and at pressures from 1/8 to 1 atm at 1400 C.

Several reaction-rate experiments have been made in the range 300 to 600 C at atmospheric pressure. The initial results are erratic, with the rates varying between a parabolic and cubic rate law. Consideration is being given to possible causes of this behavior so that consistent rate data may be obtained.

I. SOLID HOMOGENEOUS FUELED REACTORS

W. S. Diethorn and W. H. Goldthwaite

Work in support of the Pebble-Bed Reactor (PBR) is continuing. Fission-gas retention and the structural integrity of fueled-graphite spheres, prepared by several vendors and varying in composition and method of manufacture, are being evaluated in laboratory tests and in-pile experiments.

LABORATORY EVALUATIONS OF FUELED-GRAPHITE SPHERES

J. F. Lynch, M. C. Brockway, S. Rubin,
W. C. Riley and W. H. Duckworth

The strength and coating integrity of fueled spheres, supplied by commercial vendors, are being studied in impact and compression tests, and hot-oil immersion tests, respectively. In the latter, as-received spheres are immersed in hot silicone oil. Pin-hole leaks in the coating are revealed by the emission of bubbles of gas, originally entrapped in the graphite matrix, which escape from the holes on heating. Fueled spheres coated with SiC, siliconized silicon carbide, or dense carbon, prepared by four different vendors, are being evaluated in these two tests.

A self-welding test with two SiC-coated spheres of the same type has been completed. The two spheres were held in contact, under a compressive load of 50 lb, in a graphite capsule heated in a porcelain tube at 200 F for 100 hr. Commercial, good-quality argon was passed through the tube during the test. The spheres did not adhere to each other. A colored spot, 1/32 in. in diameter, was visible on each sphere at the contact point. With the exception of a slight fracturing of the silicon carbide grains (60X), this was the only visible effect observed in the test. Self-welding tests on other spheres are planned for next month.

Plans are under way to study the permeability of sphere coating to gases up to 1500 F and pressures up to 300 psi.

EVALUATION OF METAL-COATED UO_2 PARTICLES

J. Koretzky, A. F. Gerds, W. C. Riley, and W. H. Duckworth

Metal-coated UO_2 particles dispersed in graphite is of potential interest as a PBR fuel. Carburization studies of nickel-, niobium-, and nickel-chromium-coated particles, dispersed in graphite, were initiated this month. Following this study, heat-treated and control specimens of this fueled graphite will be neutron activated to determine the effect of carburization on fission-product retention.

FABRICATION DEVELOPMENT OF Al_2O_3-CLAD UO_2 FUEL PARTICLES

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

Fuel pellets consisting of UO_2 particles clad with a shell of sintered Al_2O_3 powder are being developed.

In a continuation of work reported last month, near-spherical, clad particles containing 25 w/o UO_2 were prepared in the size range minus 12 plus 18 mesh. In a severe test of the cladding integrity, this first batch was heated in air at 1200 F for 5 hr. A small weight gain occurred during the first hour. Visual examination of the pellets revealed that a few had cracked during the test. The weight gain was equivalent to the oxidation of 0.14 w/o of the UO_2 in the pellets.

The exposed U_3O_8 was leached from these pellets by washing in nitric acid, the empty cladding shells separated by flotation. If the remaining pellets show little external uranium contamination in an alpha counter, these pellets will be neutron activated and examined for postirradiation fission-gas release.

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

W. S. Diethorn

Neutron activation and in-pile experiments are being conducted as part of the evaluation program on fueled graphite. The purpose of the neutron-activation studies is to supply data on fission-gas release during postirradiation heating. In the capsule experiments fission-gas release during irradiation and the structural integrity of the fueled graphite are studied.

Neutron-Activation Studies

R. Lieberman, H. S. Rosenberg, W. S. Diethorn, and D. N. Sunderman

Neutron activation and fission-gas-release studies during postirradiation heat treatment were completed on five fueled-graphite spheres in August. Experimental apparatus used in these studies was reported last month. Three uncoated and two coated spheres were studied. The coating of one sphere, FA-8, No. 4, was defected with a 30-mil-OD hole prior to activation. Before FA-8, No. 4 was defected, this sphere passed the hot-oil test.

Frequently, the heat treatments were terminated when the release activity leveled off or when the scintillation detector was saturated. For some spheres, gas release would have certainly continued if the heat treatment at a given temperature had not been terminated.

Results are reported in Table I-1. Successive runs at the same temperature on the same specimen, FA-1, No. 282, show good agreement. There is fair agreement of release data from duplicate FA-1 spheres at the same temperature. The release from the defected sphere, FA-8, is comparable to that for the uncoated FA-1 spheres at the same temperature (1500 F).

These studies will be continued with other types of fueled-graphite spheres, the Al_2O_3-UO_2 pellets, and metal-coated UO_2 shot.

In-Pile Capsule Experiments

D. B. Hamilton, D. Stahl, G. E. Raines, R. J. Burian,
J. E. Gates, and W. H. Goldthwaite

SP-3

Capsule SP-3, containing two pairs of coated fueled-graphite spheres, was removed from the BRR in August. This capsule was irradiated in a core position for 61 days of a total in-pile time of 83 days. Thermocouples positioned outside the spheres monitored the capsule temperatures. Specimen burnup, per cent of uranium-235 consumed, is in the range 2 to 3 per cent. This rough estimate will be verified by dosimeter results. Estimated average surface temperatures of the two pairs of spheres were 1300 and 1600 F, respectively, during irradiation.

All four spheres were a nominal 1.5-in. OD and consisted of a matrix, 150-μ UO_2 dispersed in graphite, covered with a thin coating. A shell of unfueled graphite separated the fueled matrix from the coating in the FA-6 specimens. Each pair of duplicate spheres was supplied by a different vendor. One pair, FA-6 specimens, was coated with 3 mils of silicon carbide, the other pair, FA-8 specimens, with 30 mils of siliconized silicon carbide.

Postirradiation Examination. Postirradiation examination is essentially complete. Metallographic work now under way will be reported when it becomes available.

Visual Examination. Visual examination of the two FA-6 spheres, 18E and 20E, revealed numerous cracks in the coatings. The most prominent crack on Specimen 18E was along an equatorial great circle, the plane of this circle being perpendicular to the axis of the irradiation capsule. The most prominent crack in Specimen 20E was along a circle at about 30-deg latitude from the equator. There is good visible evidence that these major cracks penetrated the graphite matrix of the spheres. In addition to the prominent cracks, many fine hairline cracks were observed intersecting the major cracks, usually perpendicularly. Severe chipping and spalling of the coatings was also observed along the cracks.

TABLE I-1. FISSION-GAS RELEASE FROM FUELED-GRAPHITE SPHERES(a)

Specimen	Coating	Postirradiation Heat-Treatment Temperature(b), F	Time at Temperature(c), min	Fission-Gas Release(d), per cent of xenon-133
FA-1, No. 278	None	1000	10	0.4
		1500 (5 d)	20	0.9
		1900 (1 d)	20	0.6
FA-1, No. 282	None	1000	20	0.1
		1500 (1 d)	60	0.6
		1500 (2 d)	60	0.7
		1500 (1 d)	90	0.7
FA-1, No. 283	None	800	10	0.4
		1100 (5 min)	15	0.1
		1100 (1 d)	11	0.3
		1500 (1 d)	65	0.9
FA-11, No. 298	Carbon, 5 mils	1500	220	0.07
		1500 (4 d)	110	0.08
FA-8, No. 4	SiC, 30 mils (defected)	1500	22	0.4

(a) Each sphere irradiated once.
(b) Time in parentheses is delay time between this heat treatment and prior heat treatment.
(c) Zero time begins when heat is applied to specimen. Heatup time is about 10 min for all specimens.
(d) Number of atoms released divided by number of atoms present at beginning of heat treatment, times 100.

The two FA-8 spheres, E-4 and E-5, were in good condition. There was no visible evidence of cracks in the coatings.

Hot-Oil Tests. Prior to irradiation, all four spheres passed the hot-oil immersion test. The cracked FA-6 spheres were tested and, as expected, a copious release of bubbles was observed from the cracks. No bubbles were observed when one of the FA-8 spheres was immersed in hot oil. The second FA-8 sphere showed a slow evolution of bubbles from one point on the surface, but in two subsequent immersions, bubbles were not detected. These hot-oil test results support the results of the visual examination and show that the coating integrity of the irradiated FA-8 spheres is good.

Weight and Dimensional Measurements. The uncracked specimens, FA-6, showed only small weight changes, -0.1 per cent and nil, respectively. The major diameter before and after irradiation was measured for all four spheres. Precise measurements were difficult on these near-spherical specimens. The two FA-6 spheres showed a diameter change of -0.3 per cent and nil, respectively. For the FA-8 spheres the changes were +0.01 and +0.08 per cent.

Impact Tests. The resistance of the specimens to impact failure was determined by two different methods, free fall to a solid surface and successive impacts on the specimen by a 1-lb weight dropped from greater and greater heights. After each drop the FA-8 spheres were tested in hot oil. One FA-6 sphere when dropped from a height of 1 ft fractured into four approximately equal segments, and a few chips. This specimen was, of course, cracked before this test. Unirradiated FA-6 spheres have withstood an impact from a 1-lb weight equivalent to 25 times the impact energy of a 12-in. free fall without fracturing the graphite matrix. Coating failure was observed, however, at this 25-fold greater impact energy.

As shown by a hot-oil test, coating integrity of a FA-8 sphere was not damaged by a 12-in. free fall. In successive drops of a 1-lb weight on this FA-8 sphere, in increments of 1 in-lb, the coating failed at an impact energy of 9 in-lb as shown by bubble evolution in a hot-oil test. A different spot on the sphere was impacted each time. This 9 in-lb value is only slightly higher than the value for unirradiated FA-8 specimens

SP-4

The static capsule experiment, SP-4, was inserted in a BRR core position in August. The purpose of this four-cycle (8 week) experiment is to determine the effect of irradiation on the UO_2-carbon reaction, the coating, and the graphite matrix of fueled graphite. Two of the six spheres are coated with SiC and carbon, respectively. The remaining four are not coated. Sphere-surface target temperatures are 1900 F. Fission heat is the only source of heat in this thermocoupled capsule.

Thermocouple data at a 2-megawatt BRR power level are not available because the BRR has been operated at a power level less than this value during the current cycle. Evaluation of the termal performance of this capsule should be complete at the end of the current cycle.

SP-5

Target data for the irradiation of the SP-5 sweep capsule has been postponed until October because specimen delivery has been delayed.

SPH-1

A conceptual design of the high-flux, high-burnup capsule, SPH-1, has been completed. Final design of this capsule is waiting selection of the irradiation site (MTR, WTR). The tentative irradiation date is November 1, 1959.

SPF Capsule Series

A number of identical furnace capsules, containing one coated sphere each, are under construction as part of the in-pile evaluation of fission-gas release from fueled graphite. The major source of heat in these capsules, over a wide range of neutron flux, is provided by an electrical furnace. Fission gas will be measured in the helium which flows through the capsule. A very simple capsule design has been adopted for this series of experiments.

The first two capsules in this series, SPF-1 and -2, are being fabricated and operation of at least one is planned for September.

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

CORROSION STUDIES OF THE FLUORIDE-VOLATILITY PROCESS

C. L. Peterson, P. D. Miller, W. N. Stiegelmeyer,
and F. W. Fink

The corrosion of various articles of process equipment used in studies of the Fluoride-Volatility process at ORNL is being evaluated by metallography and by other methods. This is a program of assistance to the ORNL Chemical Technology Division.

The Mark II nickel fluorinator (FV-100) was used for several pilot-plant fluorinations at ORNL. Sections cut from the shell of this vessel and from its internal components and external auxiliary equipment have been examined.

The vessel shell and its internal pieces were attacked most severely at areas in contact with the molten salt and at the liquid-vapor interface. Considerable loss of metal occurred during the operation of this fluorinator. At one submerged area this amounted to 83 mils on the basis of an original thickness of 250 mils. Intergranular attack was prevalent all over the inside surface of this vessel and a penetration of 33 mils was observable in the submerged area just described. The attack resembled that caused by sulfur. Analyses in one area showed that the sulfur content increased from 0.012 w/o, found on the outer surface and at the middle of the wall thickness, to 0.016 and 0.024 w/o on two 5-mil layers shaved from the inside surface.

The inner surface of CRP trap of the Mark II fluorinator was etched. The attack was more severe at the bottom end, where pitting was also noticed. Intergranular penetration of at least 11 mils could be measured in this area.

The waste salt line showed minor attack except at the exit end, where a metal loss of about 21 mils was measured.

Presently, examination of some piping from points in the pilot-plant circuit further removed from the fluorinator is under way. When this is completed a detailed report of the examinations will be prepared.

Evaluations of the corrosion of the Unit Operations INOR-8 hydrofluorinator are in progress and should be completed within the next month.

J-2

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

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

A second "cold" dissolution test has been run, using one of the 7-in. prototype (heat-cycled) Consolidated Edison pins. Unlike the first test, no passivation occurred; dissolution of the cladding began immediately upon insertion of the pin into the boiling 6 M H_2SO_4 decladding solution. With a larger vapor outlet and control of the rate of heat input, foaming was not serious in this test. Decladding appeared to be complete, except for the end caps, in about 2 hr. Core dissolution appeared to be quantitative, using the standard two-step procedure.

Examination of the Darex (dilute aqua regia) process has been included in the program on Consolidated Edison fuel pins. Accordingly, one or two cold dissolution tests by this process have been scheduled prior to hot-cell testing.

Present plans are to transfer the apparatus to the hot cells early in September for initial remotely operated cold tests. Following these experiments actual testing of irradiated pins can begin.

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

The evaluation of the effects of irradiation on cast uranium monocarbide is continuing. Routine results of irradiations at the MTR are reported. In addition, the results of preparation of cast UC pins for irradiation and of thermal-conductivity studies of cast UC slugs are reported.

Irradiation Of Uranium Monocarbide

D. Stahl, W. H. Goldthwaite, and J. H. Stang

The irradiation of Capsules BMI-23-3, BMI-23-4, and BMI-23-5, containing cylindrical specimens of uranium-carbon alloy (about 4.8 w/o carbon), is continuing in the MTR. During the latter part of Cycle 125, BMI-23-4 operated with a central-core temperature of about 1050 to 1100 F, and during Cycle 126 this temperature decreased to about 1000 F. Capsules BMI-23-3 and BMI-23-5 operated during these periods with core temperatures of about 1200 and 1300 F, respectively. The temperature levels listed for BMI-23-4 were obtained directly from the thermocouple placed within the hollowed-out upper full-length specimen; other levels listed were derived from data furnished by reference thermocouples located in the NaK annulus between the specimens and the water-contacting capsule shell.

Capsule BMI-23-6 was inserted into the A-13-NE position of the MTR on a 6-1/2-in. spacer. At startup, the core temperature was over 1800 F; the capsule was immediately raised 3 in. Following this, the core temperature was still somewhat higher than the design temperature of 1500 F, so the capsule was raised 1 in. At this point, the core thermocouple started to behave erratically and appears to have shorted. However, couples in the NaK space registered temperatures indicative of top-specimen surface temperatures ranging from 900 to 1000 F; the corrosponding top-specimen core-temperature range is estimated to be 1300 to 1400 F. Temperature levels in the bottom-specimen zone of Capsule BMI-23-6 are lower than those in the top-specimen zone by approximately 200 F.

Postirradiation Examination of Irradiated Uranium Monocarbide

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

Two capsules of cast and irradiated uranium carbide specimens are scheduled to arrive at the hot cell in October for study. Irradiation levels of these capsules are nominally 5000 and 10,000 MWD/T UC.

No new results are available to report.

Preparation of UC Pins for Irradiation in the SRE

C. K. Franklin, W. J. Hildebrand, S. L. Foster,
and R. F. Dickerson

As a part of its activities in the development of nuclear power reactors, Atomics International is interested in determining the in-pile behavior of an appreciable quantity of cast UC shapes. Battelle is participating in the program to the extent of preparing the as-cast shapes and evaluating their preirradiation qualities. Castings approximately 0.6 in. in diameter and varying in length are to be prepared. The surface quality of the castings will be varied from that representative of the as-cast condition to a condition representing complete removal of surface imperfections by machine grinding.

An evaluation of recently received enriched (10 per cent uranium-235) uranium metal has been completed, and it has been found to be satisfactory for alloying. This evaluation included both metallographic and analytical examinations. It was determined that the principal impurities in the uranium were nickel (200 ppm), nitrogen (30 to 50 ppm), oxygen (10 to 20 ppm), iron (20 ppm), and hydrogen (6.6 ppm).

A total of 18 in. of enriched UC castings 0.610 ± 0.002 in. in diameter has been prepared. Approximately 10 in. of this material has been selected for evaluation and conversion to final form by squaring the ends.

Preirradiation quality of the cast material will be established by the following evaluation procedures: (1) radiographic and metallographic examination of the castings, (2) analysis of selected samples, and (3) density and electrical-resistivity measurements. The next group of castings to be prepared will be approximately 0.64 in. in diameter. The as-cast surface will be removed by machine grinding to a final diameter of 0.610 ± 0.001 in.

Thermal Conductivity of Cast UC

A. Noe, E. A. Eldridge, and H. W. Deem

Thermal-conductivity measurements have been completed on two UC specimens, 79 and 100, prepared by casting. These are the first in a series of six specimens to be investigated for Atomics International and contain about 4.9 and 5.3 w/o carbon, respectively.

The apparatus and method which was used in making the thermal-conductivity measurements were essentially the same as the steady-heat-flow method described by Van Dusen and Shelton.* The method, in brief, consists in heating one end of a specimen, measuring the temperature gradients along the specimen, and determining the rate of heat flow through the specimen by means of a metal standard of known thermal conductivity attached to the cold end of the specimen. Radial heat flow into, or away from, the specimen is minimized by radiation shielding and an encircling guard tube in which temperatures are adjusted, as nearly as possible, to match those in the specimen and standard at corresponding levels. The radiation shielding used consists of small pellets rolled from fractional-mil-thick tantalum foil, poured into the annular space between the specimen assembly and the guard cylinder. The specimen is protected by a vacuum of approximately 2 x 10^{-5} mm of mercury during the measurements.

Four 36-gage Chromel-Alumel thermocouples are wedged in holes equally spaced along the specimen, and three similar thermocouples are placed in the Armco iron standard. This permits the calculation of three-thermal conductivity values, each at a different mean temperature for each thermal equilibrium. The specimens were plated with indium, by a loaded grinding wheel technique, and bonded to the Armco iron standard.

Table K-1 shows the interpolated thermal-conductivity values for the UC specimens taken from a smooth curve drawn through the experimental points. The absolute error of the thermal-conductivity values is estimated to not exceed ±5 per cent; the chief uncertainty is the thermal conductivity of the Armco iron used as a heat-flow meter. Comparisons can be made between different materials measured by this method, however, with an error estimated to not exceed ±2 per cent, as the same Armco iron was used as the heat-flow meter in both measurements.

TABLE K-1. INTERPOLATED THERMAL-CONDUCTIVITY VALUES FOR UC SPECIMENS 79 AND 100

Temperature, C	Thermal Conductivity, cal/(sec) (cm) (C) Specimen 79, 4.9 w/o C	Thermal Conductivity, cal/(sec) (cm) (C) Specimen 100, 5.3 w/o C
200	0.056	0.056
300	0.055	0.053
400	0.054	0.051
500	0.054	0.050
600	0.054	0.049
700	0.054	0.049
800	0.054	0.049
900	0.054	0.048
1000	0.055	0.048
1020	--	0.048
1050	0.055	--

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*Van Dusen, M. S., and Shelton, S. M. "Apparatus for Measuring Thermal Conductivity of Metals up to 600 C", Natl. Bur. Standards J. Research, 12, 429-40 (1934).

Work will be resumed on this program after the remaining specimens are received from Atomics International.

Comparison of these data with those reported earlier for cast UC* having a composition of 5.2 w/o carbon shows that both specimens had lower conductivity at elevated temperature than the earlier specimen. For example BMI-1309 reports a value of 0.060 cal/(sec)(cm)(C) at 700 C. These differences in data do not correlate with carbon content, suggesting that differences in other variables such as purity or quality may be responsible.

It is interesting that the 4.9 w/o carbon sample tended to show a conductivity minimum as did the earlier data for a 5.2 w/o carbon sample, while the 5.3 w/o carbon material tended to decrease in conductivity continuously with increasing temperature.

Additional data are needed in order to understand these differences.

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*Secrest, A. C., Foster, E., Dickerson, R. F., BMI-1309, "Preparation and Properties of Uranium Carbide Castings".

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

Activity in two investigations being conducted for the LAMPRE program at Los Alamos Scientific Laboratory is reported here. Preparation and metallurgical screening studies of the present group of arc-melted tantalum-base alloys are nearing completion. Several annealed specimens fabricated of materials in this group were recently submitted to LASL for plutonium-alloy-corrosion exposures. Cold-rolled specimens were submitted during July.

Capsule BMI-25-1, which contains tantalum tensile specimens, was discharged from the MTR during Cycle 126 shutdown. The irradiation of Capsule BMI-25-2 will continue until early November.

Development of Container Materials for LAMPRE Applications

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

Work on the series of binary tantalum-base alloys (a total of 23 alloys) containing additions of hafnium, thorium, titanium, tungsten (zone refined), yttrium, and zirconium is nearing completion. As reported in BMI-1366, cold-rolled-strip specimens of these materials were forwarded to LASL for plutonium-alloy-corrosion evaluations during July. During August, 11 specimens in the annealed condition (normally 2600 F for 1 hr in vacuo) were submitted. The remaining alloys have been annealed and, according to the present schedule, will be released to LASL early in September. Chemical-analysis, hardness, and microstructure data are being obtained on the annealed as well as on the as-rolled materials.

The release of the remaining 12 alloys in the annealed condition has been delayed because of difficulties encountered in obtaining a fine-grained recrystallized structure. This has been particularly troublesome in the case of the tantalum alloys containing additions of thorium and yttrium; these alloys required annealing treatments at temperatures up to 2900 F. This resistance to recrystallization in the tantalum-thorium alloys may be influenced by the scavenging action of thorium. The case parallels one encountered in prior Battelle metallurgical studies wherein thorium in the oxide form was found to raise the recrystallization temperature of molybdenum. Even though yttrium also acts as a scavenger, the behavior of the tantalum-yttrium alloys in resisting recrystallization is not understood, particularly since prior yttrium-containing alloys of tantalum were as readily recrystallized as unalloyed tantalum. Furthermore, preliminary analytical data show that the recovery of yttrium after arc melting is very low (less than 5 ppm); this amount would not be expected to affect recrystallization behavior.

During August, the room-temperature tensile properties listed in Table L-1 were determined on 0.030-in. strip specimens of tantalum-1.5 to 6.0 w/o hafnium and tantalum-1.5 and -3.0 w/o zirconium alloys in the annealed condition. Although it was expected that solid-solution strengthening would increase with increasing additions of hafnium to tantalum, the data indicate no appreciable difference in the tensile properties of the alloys containing additions of from 1.5 to 6.0 w/o hafnium. Increased additions of zirconium from 1.5 to 3.0 w/o, however, did result in a significant increase in tensile strength as well as a decrease in ductility.

TABLE L-1. ROOM-TEMPERATURE TENSILE PROPERTIES(a) OF ANNEALED TANTALUM-HAFNIUM AND TANTALUM-ZIRCONIUM ALLOYS

0.030-In. Strip Vacuum Annealed at 2600 F for 1 Hr

Specimen	Nominal Composition (Balance Tantalum), w/o	Hardness, VHN	0.2 Per Cent Offset Yield Strength(b), psi	Ultimate Tensile Strength(b), psi	Elongation in 1 In.(b), per cent
49	1.5 Hf	159-179	56,300	75,500	19
50	3 Hf	149-170	52,800	73,100	22
51	6 Hf	151-177	53,900	74,600	16
56	1.5 Zr	154-165	55,600	69,000	31
57	3 Zr	171-187	63,300	77,600	26

(a) Results are the average of triplicate specimens per composition.

(b) Corresponding values for tantalum are: (1) offset yield strength - 25,000 to 30,000 psi, (2) ultimate tensile strength - 34,000 to 38,000 psi, and (3) elongation - 30 to 44 per cent.

Arc melting of the fourth and final series of binary tantalum-base alloys, originally scheduled for August, has been delayed due to a recent difficulty of reducing the leak rate of the arc-melting furnace to a tolerable level. Melting is scheduled to resume early in September.

Effect of Irradiation on Tantalum

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

This program has the objective of determining the effects of irradiation on the mechanical properties of tantalum. Under LAMPRE flux conditions, it is expected that there will be two modifications of tantalum: fast-neutron damage and conversion of tantalum to tungsten. In the two-capsule irradiation under way at the MTR, tensile specimens of tantalum are being exposed at low temperatures to relatively high thermal fluxes (reported to be about 3 to 5 x 10^{14}nv) but relatively low fast fluxes (reported to be of the order of 1 x 10^{13}nv). Thus, the specimens should primarily experience conversion-damage effects.

The irradiation of the first capsule (designated as BMI-25-1) was completed on August 10 after three cycles of exposures in the MTR L-53 core position; the estimated conversion of tantalum to tungsten in the specimens is about 1.5 w/o. Capsule BMI-25-2, exposed in the L-57 core position, has been irradiated for three cycles; it is estimated that four more cycles will be required to achieve a target conversion of 3 w/o.

When the latter capsule is discharged, the capsules will be returned to the Battelle Hot-Cell Facility for postirradiation examinations.

The mechanical properties of unirradiated tantalum and arc-melted tantalum-1.5 and -3.0 w/o tungsten alloys are to be determined in laboratory experiments. Tantalum-alloy specimens for this purpose are nearly ready for testing.

N-1

N. DEVELOPMENTS FOR THE MGCR

W. C. Riley

Research on core materials in support of the MGCR program is in progress at Battelle. The major effect is on the development and evaluation of UO_2 dispersions in BeO and dispersions of UC and UC_2 in graphite, and the cladding of UO_2 particles with BeO. 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 is in progress.

FABRICATION AND CHARACTERIZATION OF FUEL MATERIALS

A. B. Tripler, Jr.

Microscopic cracks which appear to originate and terminate at exposed UO_2 grains in fueled-BeO pellets were eliminated by the use of high-shrinkage fuel particles. However, annular voids surrounding these fuel particles were noted.

Some promise is held for the use of Thermax carbon black as an addition to the filler material in increasing the density of the graphite matrix in fueled-graphite pellets. Linear shrinkages of 4 per cent have been observed in Thermax-containing bodies after baking at 2300 F. In other experiments green densities of 1.7 to 1.8 g per cm^3 were attained by using fillers consisting of mixtures of AGOT and "skeletal" graphites.

BeO-clad UO_2 pellets which had previously exhibited no weight gain after heating in air for 18 hr at 1200 F are now being tested by neutron activation.

A process for producing spheriodal UO_2 fuel particles was developed.

Work on the carbonization studies in the BeO-graphite system is under way, using BeO-clad UO_2 particles dispersed in graphite matrices.

Further neutron-activation testing of the BeO-UO_2 pellets was done in August. The fission-gas release for the current group of pellets is greater by between 1 and 2 orders of magnitude than the release reported earlier on similar pellets.

UO_2 Dispersions in BeO

A. K. Smalley and W. H. Duckworth

Fuel-element cores consisting of about 20 volume per cent of UO_2 particles dispersed uniformly in a densely sintered BeO matrix are being developed.

During August, further evaluations of BeO-UO_2 pellets containing high-shrinkage fuel particles, as described previously, were made. High-shrinkage fuel particles were used in an attempt to avoid the cracks in the matrix material adjacent to the fuel which occur when plain UO_2 is used. Crystal-size and permeability measurements and metallographic examinations were made on specimens in which the fuel was initially present as particles of either UO_2-10 w/o $Be(OH)_2$ or UO_3. The data are shown in Table N-1.

As reported previously, the bulk densities of specimens containing high-shrinkage fuel particles were lower than those of similar specimens in which the fuel was introduced as unadulterated UO_2 particles. The average crystal size of the BeO matrix was not appreciably influenced by the nature of the fuel particles. Pellets in which the fuel was introduced, either as UO_2 or as UO_2-10 w/o $Be(OH)_2$ had permeabilities of less than 0.03×10^{-5} darcy. Specimens in which UO_3 particles were employed had permeabilities ranging from 0.25 to 0.62×10^{-5} darcy.

Examinations of polished and etched sections at 250X indicated the following:

(1) No gross differences in the structures of the BeO matrices were apparent in specimens containing the three types of fuel particles.

(2) All specimens containing high-shrinkage fuel particles had prominent annular voids surrounding the fuel particles. The BeO and the UO_2 were in intimate contact in specimens in which the fuel was introduced as unadulterated UO_2.

(3) Fuel introduced either as UO_2-10 w/o $Be(OH)_2$ or UO_3 was quite porous in the sintered specimens. This also was in contrast to specimens in which the fuel was introduced as unadulterated UO_2.

Pellets containing the three types of fuel particles are being tested by neutron activation to assess the effects of the structural variables on fission-product retention. Results should be available during September.

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 UC or UC_2 particles in an amount equivalent to 20 volume per cent of UO_2.

TABLE N-1. BeO-UO_2 FUEL-ELEMENT CORES FOR NEUTRON-ACTIVATION TESTING

Starting Fuel Material(a)	Sintering Conditions(b): Maximum Temperature, F	Sintering Conditions(b): Time at Maximum Temperature, hr	Sintered Bulk Density(c): G per Cm^3	Sintered Bulk Density(c): Per Cent of Theoretical	Average Crystal Size in BeO Matrix, μ	Permeability, 10^{-5} darcy
UO_2	2600	1	4.27	92.3	5(c)	0.028
	2800	1	4.41	95.2	18(c)	0.030
	3000	1	4.54	98.0	37(c)	0.030
	3000	4	4.51	97.5	37(c)	0.030
UO_2-10 w/o $Be(OH)_2$	2600	1	4.15	89.5	--	0.026
	2800	1	4.42	95.2	19	0.025
	3000	1	4.35	93.7	28	0.026
	3000	4	4.43	95.5	36	0.025
UO_3	2600	1	4.05	87.4	6	0.62
	2800	1	4.17	90.0	21	0.25
	3000	1	4.20	90.6	31	0.34
	3000	4	4.22	90.9	39	0.27

(a) All specimens contained a nominal fuel loading of 20 volume per cent UO_2 in the sintered condition.

(b) In flowing hydrogen.

(c) These data reported previously.

During August the major effect was directed toward increasing the bulk density of the graphite matrix through: (1) use of methyl ethyl keton (MEK) as a solvent to disperse the coal-tar pitch in the preparation of graphite-pitch fuel mixtures, and (2) variations in type and particle size of filler carbon.

Results on the investigation of methyl ethyl ketone as a pitch solvent indicate that the bulk density of nonfueled compacts after baking was approximately the same as that obtained when benzene or carbon tetrachloride was used.

In the investigation of various filler carbons, results available to date indicate that considerable shrinkage occurs during baking when the filler carbon is composed of about 12 w/o of Thermax carbon black and 88 w/o of Texas 95 petroleum coke. Compacts containing about 60 w/o of UC or UC_2 particles and 30 w/o of the above filler carbon showed a linear shrinkage of about 4 per cent after baking at 2300 F. Further study of this type of filler is indicated.

Data on nonfueled compacts made from mixtures of minus 325-mesh AGOT graphite and a highly crystalline "skeletal" graphite, bonded with coal-tar pitch, indicate that green densities in the range of 1.7 to 1.8 g per cm^3 can be attained. Density data on these compacts after baking are not available yet.

In future work, the study of methods to increase the density and decrease the gas permeability of the matrix carbon in fueled compacts will be continued.

Cladding of UO_2 Particles With BeO

A. K. Smalley and W. H. Duckworth

A process for fabricating fuel pellets consisting of particles of UO_2 contained within impermeable BeO shells is being developed.

BeO-clad UO_2 pellets, prepared by the procedure described previously, exhibited no weight gain on heating for 18 hr at 1200 F in air, indicating that the BeO claddings effectively prevented oxidation of the UO_2 fuel particles to U_3O_8. A sample of these pellets was submitted for neutron-activation testing to assess the ability of the claddings to retain fission products. Another portion of these pellets was used in the carbonization studies, described in another section of this report.

During August a process for preparing spheroidal fuel particles was developed involving pelletizing in a planetary mixer. The use of spheroidal particles will allow control of the BeO cladding thickness. The effect of the cladding thickness on cladding integrity will be evaluated initially by the high-temperature oxidation-resistance test.

N-5

Carburization Studies in the BeO-Graphite System

J. Koretzky, A. J. Roese, S. A. Rubin, W. C. Riley, and W. H. Duckworth

The effect of temperature and time on the reaction of graphite with BeO will be studied.

BeO-clad UO_2 particles prepared by the method described in BMI-1366 were dispersed in various types of graphite matrices. Specimens will be heat treated at temperatures above 1700 F for from 1 to 100 hr. Specimens will be examined metallographically for evidence of carbide formation.

The effect of carbide formation on fission-product retention will be determined by neutron-activation tests.

Preliminary Characterization by Neutron Activation

P. Gluck, R. H. Barnes, and D. N. Sunderman

The program to investigate the release of fission products from potential fuel materials for the MGCR is continuing.

The preliminary phase of this investigation, which is now in progress, consists of a study of the release of fission products during postirradiation heat treatment from UO_2-BeO, UC-graphite, and UC_2-graphite fuels. The results of these preliminary studies are to be used to select a promising material for more extensive evaluation.

Data reported last month indicated that of the three materials the UO_2-BeO specimens had the best fission-gas retention. Current studies are directed toward further evaluation of fission-gas retention in UO_2-BeO with specimen-fabrication techniques as the variable. The data reported this month cover the results of fission-gas-release studies that were conducted on twelve 48.25 w/o UO_2-BeO pellets.

The procedure employed for these fission-gas-release studies was the same as that reported last month. Table N-2 summarizes the results of the fission-gas analyses. Xenon-133m, tellurium-132, and its daughter, iodine-132, were found in addition to xenon-133 and iodine-131 when the period between irradiation and heat treatment did not exceed 12 days. The tellurium-132, iodine-132, and xenon-133m present could not be determined accurately because of the mutual interference of gamma-ray photopeaks of nearly the same energy. Estimates indicate that the amounts of tellurium-132 released from Specimens D-2, A-4, and C-10 were of the same order of magnitude as the xenon-133. The quantity of fission products released is higher (by factors of up to 100) than the values reported last month for a series of four UO_2-BeO pellets. The only apparent difference between the two series of tests was the type of activated charcoal used. In the first series a low-purity charcoal was used, while a high-purity charcoal was used in the work reported here. Tests are being conducted to determine if the differences in the two series of results can be attributed to the charcoal.

TABLE N-2. FISSION-GAS RELEASE FROM 48.25 w/o BeO-UO_2 PELLETS DURING POSTIRRADIATION HEAT TREATMENT IN VACUUM AT 1800 AND 2000 F FOR 24 HR

Pellet[a]	Firing Temperature, F	Annealing Temperature, F	Decay Time[b], days	Thermal-Neutron Flux, nv	Total Uranium Burnup, a/o	Fission-Gas Release[c], per cent	
						Xenon-133	Iodine-131
A-4	2800	2000	11.5	1.68×10^{12}	3.07×10^{-6}	0.69	1.37
A-5	2800	2000	11.5	1.49×10^{12}	2.72×10^{-6}	0.63	1.14
C-6	3000	2000	11.5	1.25×10^{12}	2.29×10^{-6}	1.74	4.07
C-8	3000	2000	11.5	1.18×10^{12}	2.15×10^{-6}	1.79	3.49
C-10	3000	2000	11.5	1.13×10^{12}	2.06×10^{-6}	1.86	4.60
D-2	3000	1800	9.67	4.45×10^{11}	6.15×10^{-7}	0.21	0.28
D-3	3000	1800	9.67	4.31×10^{11}	6.30×10^{-7}	0.12	0.23
D-4	3000	1800	9.67	3.87×10^{11}	5.66×10^{-7}	0.20	0.30
A-3	2800	1800	66	1.95×10^{12}	3.30×10^{-6}	0.46	0.38
B-3	2600	1800	66	2.21×10^{12}	3.76×10^{-6}	1.17	1.24
C-9	3000	1800	33.2	1.16×10^{12}	2.12×10^{-6}	0.55	1.07
D-5	3000	1800	16.8	3.65×10^{11}	5.35×10^{-7}	0.17	0.33

(a) Specimen designation is 15171-52.

(b) Radioactive decay time between irradiation and radioassay.

(c) Fission-gas release is the ratio of atoms released divided by the atoms present at the beginning of heat treatment multiplied by 100.

During next month, studies will be continued on the UO_2-BeO, UC-graphite, and UC_2-graphite systems.

STUDIES OF FISSION-GAS RELEASE FROM FUEL MATERIALS

R. H. Barnes

Design, construction, and assembly of equipment for the neutron activation and in-pile fission-gas-release studies is continuing. Experimental work in these two phases of the study will be initiated in October and November, respectively.

Detailed Neutron-Activation Studies

P. Gluck, R. H. Barnes, and D. N. Sunderman

During August, work continued on the assembly of the apparatus for the post-irradiation fission-gas-release study. Completion of the apparatus is expected in October.

N-7

In-Pile Studies

N. E. Miller and G. E. Raines

Capsule design is well under way. A thermal mock-up of the in-pile capsule will be built to provide information on the thermal and mechanical performance of the capsule. Electrical heaters for the mock-up are currently being fabricated.

HIGH-BURNUP IRRADIATION EFFECTS IN FUEL MATERIALS

W. E. Murr, J. E. Gates, and R. F. Dickerson

A study of the radiation stability of ceramic-type fuels under conditions simulating those of MGCR design is in progress. Fuels under investigation include 20 volume per cent uranium dioxide in beryllium oxide, 20 volume per cent uranium monocarbide in graphite, and 20 volume per cent uranium dicarbide in graphite. Specimens were prepared for irradiation by sealing four pellets of a given type of fuel, each approximately 0.222 in. in diameter by 0.250 in. long, into stainless steel tubes with 0.012-in.-thick walls under a helium atmosphere. Six specimens, two containing each type of fuel material, were placed in an irradiation capsule containing thermocouples and electrical heaters. Four such capsules, designed to maintain specimen-surface temperatures of 1500 F in a specified neutron flux, were included in the program. The irradiation of one capsule containing 93 per cent enriched fuel has been completed. The irradiation of the three remaining capsules containing 30 per cent enriched fuel is in progress in the MTR.

The specimens irradiated in the first capsule achieved burnups of about 1.5 a/o of the uranium at surface temperatures ranging from 1300 to 1550 F. The postirradiation examination of these specimens included determination of fission-gas content of the gas contained by the capsule, measurement of dimensions of the specimens, determination of fission-gas content of the gas contained inside three selected specimens, measurement of the density of the BeO fuel pellets, and metallographic examinations of each fuel material. The results of these examinations have been reported previously. In general, the specimens did not exhibit any gross deformation or microstructural changes.

In last month's report, the percentages of fission gas released from the fuel pellets of UO_2 in BeO and UC_2 and UC in graphite irradiated in the first capsule were reported as 0.13, 0.30 and 0.40 per cent, respectively. Additional calculations have revealed that these figures are low and should be 0.40, 0.93, and 1.24 per cent, respectively. The nominal composition of these fuels has been reported as 48.3 w/o UO_2 in BeO, 66.6 w/o UC_2 in graphite, and 61.3 w/o UC in graphite. The actual compositions are 59.2 w/o UO_2, 74.5 w/o UC_2, and 73.0 w/o UC, respectively.

Additional metallographic examinations have been performed on pellets of both UC and UC_2 in graphite. Two unirradiated control pellets and two irradiated pellets, one of each fuel composition, were mounted, ground, and polished simultaneously. A more fluid resin was used in mounting the irradiated pellets in an attempt to retain the particles of fuel and graphite. Examination indicated that the two unirradiated pellets retained their matrix material. Large areas, predominantly near the center of the irradiated pellets, were pitted and eroded, although not as extensively as encountered in the earlier examinations. Thus it appears that irradiation caused the graphite matrix surrounding UC and UC_2 particles to become brittle. The matrix material then is easily loosened and removed during metallographic preparation.

Sections of the Type 316 stainless steel cladding from Specimen 9, which contained UC-graphite samples, Specimen 2, which contained UO_2-BeO samples, and Specimen 4, which contained UC_2-graphite samples, were examined metallographically in both the polished and etched condition. Microhardness traverses were obtained from the inner to the outer edge of the cladding. Comparison with unirradiated Type 316 stainless steel tubing indicated that the cladding from all three specimens increased in grain size by a factor of approximately two. A trend toward greater hardness near the inside of the cladding was not observed. The cladding from Specimens 2 and 9 appeared similar to unirradiated material, except for larger grain size. The cladding from Specimen 4 contained a slightly different type of microstructure and a thin, dark zone, approximately 1/4 to 1/2 mil wide, on the side of the cladding adjacent to the fuel. This is believed to be an etching effect rather than carburization. The average Knoop hardness of the cladding of Specimen 2 was 189, of Specimen 9 was 208, and of Specimen 4 was 236, while the unirradiated control specimen had a hardness of 165. In general, there did not appear to be any deleterious effects in the cladding.

The remaining three capsules of the series have been in operation in the MTR since the beginning of MTR Cycle 125. During initial insertion, the heaters were connected incorrectly so that they did not provide the proper amount of auxiliary heat to maintain the 1500 F specimen-surface temperature. During the shutdown for MTR Cycle 126, the heaters of Capsules BMI-31-1 and BMI-31-2 were rewired, and current temperature readings obtained from the four operating thermocouples indicate that the specimen temperatures are now near 1500 F, the desired temperature. Capsule BMI-31-3 is operating between 1000 and 1140 F, but will be rewired at the end of MTR Cycle 126. It is believed that the specimen can then be maintained at the desired temperature. All heaters are in operation in all the capsules at the present time, but two of the six thermocouples in each capsule are inoperative. It is planned to operate Capsule BMI-31-1 for a total of 8 MTR cycles and Capsules BMI-31-2 and BMI-31-3 for a total of 14 MTR cycles.

DIFFUSION OF FISSION PRODUCTS IN CLADDING MATERIALS

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

The diffusion of fission products in "A" Nickel cladding material is being investigated. Specimens of "A" Nickel are being impregnated with fission products by irradiation recoil.

N-9 and N-10

A capsule in which 20 specimens of uranium sandwiched between two "A" Nickel foils were irradiated has been removed from the pool of the Battelle Research Reactor and opened. The sandwiches were extracted and the welds holding each sandwich together were sheared. After removal of the uranium foils, the fission-product activities within the "A" Nickel foils were as high as 500 mr per hr at the exposed surface. This indicates considerable recoil of fission products into the foils. Two 5-mil foils were pressed together at 700 C under an applied load of 7 tons. Microscopic examination revealed that bonding of the impregnated faces of the foils was not quite complete due to ripples originally present in the foils. Bonding of another pan of 5-mil foils is being attempted using smoother foils and higher temperature and pressure during the pressing operation.

The use of cathodic bombardment to study introduction and diffusion of fission gas in metals is being evaluated. "A" Nickel specimens were subjected to argon bombardment in a cathodic etching unit. The specimen surface was cleaned to remove surface-adsorbed gases, and then sealed in Vycor tubes and heated to 900 C. The tubes were punctured and a sample of the released gas was obtained. Spectrographic analysis indicated the presence of small amounts of argon. No oxygen was detected, indicating that air did not leak into the tubes. Therefore, it appears that the argon present diffused from the "A" Nickel. This work indicates that the argon was successfully introduced into the metal lattice by the cathodic etching unit.

CARBON-TRANSPORT CORROSION STUDIES

N. E. Miller, D. J. Hamman, J. E. Gates, and W. S. Diethorn

Selected metal and graphite specimens have been exposed to radiation in helium-filled quartz tori designed to promote convective flow of the helium and gaseous impurities past the specimens. The initial results were reported last month in BMI-1366. Additional metallography is under consideration.

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor-Flow Studies

G. R. Whitacre and H. R. Hazard

Air-flow studies in a quarter-scale model of the Experimental Gas-Cooled Reactor are being conducted at Battelle to provide design data. Work previously reported includes model construction and installation and initiation of a program to determine the effect of hole location in the core-support cylinder on core-flow distribution, mixing, and pressure drop.

In August, six different core-support-cylinder configurations were studied, and one was selected as best on the combined basis of flow patterns, pressure drop, mixing, and core-flow distribution. A cylinder with this hole configuration scaled to prototype thickness was then placed in the model and core-flow-distribution and pressure-drop tests were run to check the previous results with the sheet-metal cylinder.

In September, complete thermal-shield and upper-plenum flow studies will be run with the final core-support cylinder in place. Also, the effects of Reynolds number and shield flow rate on pressure drops and core-flow distribution will be studied. This should conclude the experimental program.

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

The investigations covered in this section of the report are being conducted in assistance to Alco Products and are concerned with the development of fuel, absorber, and suppressor materials for the SM-2.

The reference fuel system consists of a Type 347 stainless-UO_2 (MCW spherical) dispersion clad with Type 347 stainless steel. Boron in the form of a dispersion of ZrB_2 is incorporated into the fuel core as a burnable poison.

Studies are being conducted to determine the effects of variables such as impurity content, chemical composition, and sintering conditions on the reaction of ZrB_2 with the stainless matrix and the subsequent loss of boron through vaporization. Results indicate that by proper control of these variables this reaction can be eliminated or sufficiently decreased to maintain boron content within the desired specification.

A noninstrumented capsule containing eight specimens is being irradiated in the MTR in a reported flux position of about 4×10^{14} nv. Two additional capsules are scheduled for insertion within the next month. An instrumented capsule embodying thermocouples and auxiliary heating coils has been fabricated and is scheduled for insertion in the ETR.

Materials Development

S. J. Paprocki, D. L. Keller, G. W. Cunningham, D. E. Lozier,
A. K. Foulds, W. M. Pardue, and J. M. Fackelmann

The selection of suitable materials and the development of techniques for their fabrication into fuel elements, suppressor components, or control rods for the SM-2 reactor is the objective of this program. Irradiation specimens are also prepared as required.

Fuel Materials

The reference fuel element contains a core of approximately 26 w/o UO_2 and 1 w/o ZrB_2 dispersed in a Type 347 prealloyed stainless powder matrix (0.030 in. thick) and clad with 0.005-in.-thick Type 347 stainless steel.

The incorporation of a boron-bearing burnable poison in such a manner that boron loss is low or negligible has been a major problem area in the development of SM-2 fuel plates. As previously reported, when ZrB_2 is used under carefully controlled procedures, massive unreacted particles of ZrB_2 are retained in the matrix and little or no loss of boron occurs. However, a reaction and subsequent loss of boron can be produced if poor sintering conditions, ZrB_2 with low boron content, impure ZrB_2, or impure metal matrix powders are used. Therefore, in order to determine specifications for the core materials and to establish sintering procedures, it is necessary to obtain information concerning the effect of these variables on ZrB_2 compatibility and boron loss.

In reference to sintering atmospheres, previously reported results have shown that erratic results are obtained for specimens sintered in hydrogen whereas losses are negligible in vacuo. The boron loss is believed to be related to the oxygen partial pressure, which varies from run to run in the hydrogen furnace but should remain relatively constant and at a low value in a good vacuum system. It should be noted that the heaviest losses occur in hydrogen, regardless of the boron content, impurity levels, etc., of the ZrB_2 powder.

It has also been shown that, in general, higher boron losses occur in ZrB_2 with lower boron contents. These results may be partially influenced by the presence of greater amounts of impurities in the powders of lower boron content. Additional work will be required to determine the effect of nonstoichiometry on the boron loss.

A series of tests is in progress to determine the effect of such impurities as carbon, niobium, nitrogen, oxygen, and silicon in producing secondary reactions of ZrB_2 with the stainless matrix. It is expected that these impurities will behave differently, depending upon whether they are initially present in the matrix or the ZrB_2. If present in the ZrB_2 they may be combined in the form of undesirable borides which will react with the stainless, whereas the pure ZrB_2 may react at a lower rate with the impurities in the metal matrix. For example, no definite trend could be established with carbon additions to the matrix of 0.1 to 1 w/o, but higher boron losses appear to be related to higher carbon contents of ZrB_2. In this case, the carbon probably is combined to form B_4C, which will react with the matrix.

The role of silicon has not been clearly defined, but it is possible that a complex silicoboride may form, resulting in the loss of boron. When silicon is added to the matrix either as an elemental powder or as an alloyed constituent, the oxidation of ZrB_2 powders due to oxygen in the matrix is reduced. Nevertheless chemical analyses indicate that, when silicon is alloyed with the matrix, more boron is lost as silicon content is increased. After sintering for 2 hr at 2300 F in vacuo, a compact containing low-silicon (<1 w/o) metal powder contained 0.136 w/o boron, while a compact containing high-silicon (~2.5 w/o) powder contained 0.123 w/o boron. On the other hand, when silicon was added as an elemental powder to an iron matrix a 2 w/o silicon compact sintered at 2200 F contained 0.142 w/o boron compared with 0.154 w/o boron for a specimen containing 3 w/o silicon. The behavior in iron is probably not typical of the stainless steels since the iron-silicon reaction may be important in determining the boron loss.

The effect of niobium additions was studied in both elemental iron-chromium-nickel matrices and iron matrices. In both cases, the boron loss increased with increasing niobium contents up to 2 w/o. Also, at the 2 w/o niobium loading a slight amount of a secondary phase could be detected. However, in the one specimen which contained 5 w/o niobium, the boron loss was decreased. The boron analyses of these plates are listed in Table P-1.

TABLE P-1. BORON ANALYSES OF NIOBIUM-CONTAINING SPECIMENS

Matrix	Nominal Niobium Addition, w/o	Boron Analysis, w/o
Elemental iron-chromium-nickel	1.0	0.236
	2.0	0.211
	5.0	0.253
Iron	0.5	0.139
	1.0	0.134
	2.0	0.115

Both oxygen and nitrogen are known to be detrimental, but no quantitative data have been obtained. It is believed that oxygen reacts to form ZrO_2 and B_2O_3 with a resulting vaporization of B_2O_3 and thus a boron loss.

It has been fairly well established that stoichiometric ZrB_2 is more stable than the compound with lower boron contents. High-purity ZrB_2 containing 19.1 w/o boron is being used to aid in this evaluation. The powder is available only in minus 325-mesh size and has to be consolidated into larger particles before it can be used as a dispersion. A series of compacts has been prepared using ZrB_2 powder crushed from an arc-melted button and powder from a hot-press-forged button. A metallographic examination of the sintered compacts has been made but no chemical analyses have been received. The metallographic examination indicated that excellent results can be obtained with this powder. There was no indication of secondary phases in the matrix, ZrO_2 around the particles, or any extraneous phases in the ZrB_2 particles.

In the fabrication studies, a second die is being completed which should produce compacts of the desired size for full-size fuel elements, and small-scale specimens are being roll clad in order to study fabrication variables. A second series of specimens has been roll clad using total reductions ranging from 5 to 1 to 15 to 1 in thickness. This series contains spherical UO_2 while the first series contained high-fired UO_2. In addition, a series of specimens is being prepared to determine the effect of sintering and rolling temperatures, particle size, and boron content on the ZrB_2 which is contained in the core.

Metallographic examination and comparison of the series containing spherical UO_2 with the series containing high-fired UO_2 indicated that a better quality UO_2 dispersion was obtained at all reductions when spherical powder was used. Even when the spherical UO_2 fractures, the sections of the particle are not, in general, stringered out by the rolling process. As might be expected, the particles which have gone through the higher reductions are somewhat more elongated. There appeared to be a small percentage of particles at all reductions which fractured and stringered. These particles were probably in elongated egg shapes initially.

Fabrication of Irradiation Specimens

Fabrication is continuing on the specimens for the remaining six ETR irradiation capsules. The specimens containing the high fuel loading of UO_2 and UN (Types 16, 17, and 18) have been completed as have the reference (Type 6) specimens containing spherical UO_2-ZrB_2. Analyses of these specimens for boron content have been completed and are satisfactory.

An arc melt of NiB was made and used as the source of boron-10 to induction melt and fabricate the type of irradiation specimen which contains a Type 347 stainless steel-boron frame. The frame is presently being machined from this material. Frames for all remaining specimens have been machined, and the cores are being prepared.

Fabrication-study specimens listed in Table P-2 were selected for three MTR mock-up capsules. These specimens were sintered in hydrogen and not taken from those prepared for the irradiation capsules.

TABLE P-2. SPECIMENS FOR MTR MOCK-UP CAPSULES

Specimen Identification			UO2		Poison			
Number	Type	Notches	Amount, w/o	Weight, g	Type	Boron, w/o	Core Weight, g	Remarks
1-7H	7H	0	26	0.514	ZrB_2	0.073	1.977	All specimens were hydrogen sintered
2-7H	7H	1	26	0.514	ZrB_2	0.073	1.978	
3-7H	7H	2	26	0.510	ZrB_2	0.073	1.960	
1-6H	6H	1	26	0.514	ZrB2	0.212	1.975	
2-6H	6H	2	26	0.513	ZrB_2	0.212	1.973	
3-6H	6H	3	26	0.514	ZrB_2	0.212	1.978	

Development of Suppressor Materials

Work has been initiated to select materials and determine a method for incorporating a high-cross-section material at the ends of the fuel-element cores as a flux suppressor. Materials to be considered include boron-stainless alloys, europium oxide-stainless dispersions, and core sections containing less than nominal UO_2 or more than nominal ZrB_2 or both. Because of physics design calculations and relative ease of fabrication, major emphasis is being placed on the boron-stainless alloys. It will still be necessary to consider boron losses, but, since the sections will be used only for suppressing the flux peaks at the ends of the plates and will not be used to control reactivity, the boron loading does not have to be as closely controlled as the boron loading for the burnable-poison-containing fuel cores.

Development of Control Materials

Subscale control elements containing cores of 33 w/o EU_2O_3 dispersed in an elemental stainless matrix have been fabricated for use in corrosion tests at Alco Products. These elements are 4 in. long, 1 in. wide, and have cores 0.090 in. thick. Both clad defected plates and bare cores were prepared. The clad plates contain a silicon-free stainless barrier foil approximately 0.005 in. thick between the core and cladding.

Europium oxide of the desired particle size (minus 100 plus 200 mesh) was prepared by pressing EU_2O_3 minus 325-mesh particles into a compact containing Ceremul "C" as a binder, drying the compact, crushing the compact into minus 80 plus 100-mesh particles, and sintering the particles in a platinum boat at approximately 1600 C.

Core compacts were prepared by blending dry powders of Eu_2O_3 and minus 325-mesh chromium, iron, and nickel for 1 hr, adding a camphor-alcohol binder, blending 2 hr. The compacts were roll clad at a temperature of 2200 F.

Additional work on the preparation of spherical Eu_2O_3 to be used in fabrication studies is in progress.

Encapsulation Studies

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

In order to determine radiation stability of fuel materials developed for the SM-2 program, a ten-capsule program has been initiated. The program includes the encapsulation, irradiation, and examination of specimens in three noninstrumented capsules in the MTR and in seven highly instrumented capsules in the ETR.

The three nonlead capsules, BMI-32-1, BMI-32-2, and BMI-32-3 arrived at the MTR during July. Capsule BMI-32-1 was inserted during Cycle 126 shutdown in the MTR L-53 core position and Capsule BMI-32-2 is scheduled to be inserted on August 31 into the L-56 MTR core position. Capsule BMI-32-3 will be inserted into an undesignated MTR core position during Cycle 128 shutdown on September 20. Reported thermal fluxes for the L-53 and L-56 positions are 3.2 to 4.2 and 3.0 to 5.0 x 10^{14} nv, respectively.

The first of the seven instrumented capsules, BMI-32-4, was shipped to the ETR early in August. It is scheduled to be inserted in the 0-6 position (beryllium reflector) at the beginning of the next full-power cycle, which starts on October 4. Information pertaining to the specimens encapsulated in BMI-32-4 is reported in Table P-3.

In addition to the capsules containing irradiation specimens, three nuclear-mock-up capsules have been shipped to NRTS for flux-perturbation experiments. The mock-ups are similar in design to the irradiation capsules and each contains reject specimens fueled with 26 w/o UO_2 plus boron for the specimen simulation. Each capsule contains aluminum-0.6 w/o cobalt dosimeter wires. One of the mock-up capsules willbe exposed in the MTR core (L-50 row), a second will be exposed in an ETR reflector position (O-6 position), and the third will be exposed in an MTR reflector position (probably an A-38 or A-39 position). The exposures, scheduled for mid-September, will be at 1/100 full-reactor power for about 15 min. The capsules will then be returned to the Battelle Hot-Cell Facility, where dosimeter analyses will be made by direct-wire counting.

TABLE P-3. FUELED CORE COMPOSITIONS FOR SPECIMENS[(a)] CONTAINED IN CAPSULE BMI-32-4, TO BE IRRADIATED IN ETR O-6 POSITION

Specimen Location in Capsule, Starting from Top Position	UO_2 in Type 347 Stainless Steel Dispersion (Nominal Uranium-235 Content is 0.423 G), w/o	Poison Type	Nominal Boron Content, w/o
1	26	ZrB_2	0.209
2	26	None	--
3	26	ZrB_2	0.093
4	26	None	--
5	26	ZrB_2	0.213
6	26	CbB_2	0.207

(a) Core dimensions are 1 by 1/2 by 0.030 in., while standard specimen dimensions are 1-1/2 by 9/16 by 0.042 in.; the cladding is Type 347 stainless steel.

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".

MATERIALS DEVELOPMENT PROGRAM

D. L. Keller

Results of spectrochemical analyses and oxygen/uranium determinations on UO_2 pellets are reported. These pellets were selected at random from the group fabricated for the four fuel-element subassembiles, in the IB-IT series.

Progress of capsule-irradiation programs is reported for stainless-UO_2 and -UN dispersion fuel elements, solid UO_2 and annularly loaded UO_2 fuel pins, and graphite-UO_2 fuel bodies. These materials are being irradiated at both the MTR and the BRR.

The IB-IγT fuel assembly irradiated in the BRR in-pile loop is presently undergoing hot-cell examination. Preliminary results are reported.

Results from the GCRE critical experiments on reactivity worths of mock-up control blades, void-coefficient measurements, and flux and power distributions in the IB-IϕT are reported.

Fabrication of UO_2 Pellets

H. D. Sheets and C. Hyde

Previously, about 6500 cylindrical UO_2 ceramic pellets, 0.160 in. in OD, were prepared for in-pile testing. About one-third of these were solid. The remainder were hollow, and had an ID of 0.048 in. Present objectives include (1) completion of the examination of pellets selected randomly from this lot and (2) demonstration of the feasibility of dry pressing and sintering UO_2 pellets 0.160 in. OD with hole diameters varying from 0.040 to 0.130 in. This program will include both laboratory effort and discussions with manufacturers of ceramics and of ceramic-processing equipment.

Microscopic examination of polished sections of random pellets chosen from the large group made for in-pile testing showed that the specimens were free of cracks and laminations. Most of the porosity appeared to be intergranular. The open porosity of the specimens, as measured by water saturation, was found to be less than 0.5 per cent. A group of pellets chosen at random was subjected to oxygen/uranium-ratio determinations and spectrochemical analyses. The results are presented in Table Q-1.

TABLE Q-1. CHEMICAL COMPOSITION OF UO_2 PELLETS

Pellet Geometry	Enrichment, per cent	Oxygen/Uranium Ratio	Carbon Content, w/o	Spectrochemical Analysis of Impurity Content, ppm										
				Ni	Cr	Ca	Al	Be	B	Si	Fe	Mg	Mo	Cu
Solid	25	2.01	<0.001	10	5	15	40	0.2	1	40	40	10	15	10
	50	2.00	0.001	30	5	15	10	0.4	7	40	40	7	15	40
Hollow	25	2.00	0.001	30	15	30	40	0.1	1	80	80	15	15	40
	50	2.00	<0.001	30	5	15	<10	0.5	5	60	60	5	15	10

Efforts to make precise surface-area measurements were unsuccessful. The surface area was found to be less than 0.3 m^2 per g, which is the lower limit of the sensitivity of the apparatus available.

Necessary parts for dies for making pellets having inside diameters in the range of 0.040 to 0.130 in. are being made. It is anticipated that they will be available during the next report period.

Encapsulation Studies

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Irradiation of Clad Pin-Type Specimens Containing Dense UO_2

Capsule BMI-27-1 was discharged from the MTR at the end of Cycle 125 (August 10) and is now awaiting shipment to the Battelle Hot-Cell Facility. This capsule, along with Capsule BMI-27-2, which was discharged during July, contained six Inconel-clad UO_2-fueled pins. It is estimated that fission burnups of specimens in BMI-27-1 range from 6 per cent for the peak flux specimens (which were maintained in the 1400 to 1600 F range) to about 3 per cent for the remaining specimens.

During August, a careful review was made of the temperature-level history of Capsule BMI-27-1 to pinpoint periods during which specimen overheating may have occurred. Evidence of such overheating, possibly to a level as high as the melting point of Inconel, was found during hot-cell examinations. However, the review provided no evidence of such excursions; in fact, evidence of excursion past 1700 F was not found. At present, some consideration is being given to the possibility that specimen hot spots occurred as a result of high local fuel-burnup rates. This analysis, however, is faced with a variety of uncertainties in flux-distribution characteristics.

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Irradiation of Specimens Containing UO_2 in Graphite

Capsule BMI-29-1 was inserted in the MTR A-39-SE position for Cycle 126, which started on August 15; the reported peak unperturbed flux in this position is 1.2 x 10^{14} nv. This capsule contains six clad pins fueled with 8 w/o UO_2 (highly enriched) in graphite. Cladding materials are Hastelloy X, Inconel 702, and Carpenter 20 Cb.

During the first irradiation cycle, specimen-surface temperatures as indicated by thermocouple data ranged from 1600 to 1775 F; the design temperature is 1700 F. Approximately 1-1/2 kw of heat was supplied by the externally controlled electrical heaters incorporated into the capsule. According to present estimates, the target specimen burnup level of 15 total a/o will be achieved in three irradiation cycles; the corresponding discharge date is mid-October.

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

This program has the objective of comparing the irradiation stability at 1650 F of stainless steel-clad dispersions of stainless steel-30 w/o MCW spherical UO_2 (highly enriched) with that of similarly clad dispersions of stainless steel-30 w/o hydrothermal UO_2. Two capsules, each containing two plate specimens of each type of UO_2, were fabricated and shipped to the MTR during July for irradiation. These capsules have been designated BMI-33-1 and BMI-33-2, and will require three and two normal reactor cycles, respectively, to achieve the desired fission burnups of 9 and 6 a/o. Because of limited clamping space for lead-tube experiments in the MTR, the capsules will not be inserted before Cycle 128 shutdown (September 21).

Irradiation of UO_2-Graphite Specimen Assemblies With an Integral Corrosion-Gas Flow System

Preliminary studies to obtain basic capsule-design information for the elevated-temperature irradiation of SiC-coated graphite bodies (containing 8 w/o highly enriched UO_2) continued during August. The three capsules involved in this program are designated as the BRR II-2C group and will be irradiated in a core-face position at the Battelle Research Reactor. Their salient features, which are described in some detail in BMI-1366 and BMI-1357, are essentially as follows:

(1) Each capsule will contain two small cylinders (1-3/8 in. in OD by 1 in. long) of the fueled material enclosed in a tight-fitting Inconel X or Hastelloy X can with welded headers.

(2) Heat will be transferred radially from the individual cans to the water-contacting capsule shell through a thin helium-argon annulus; the nominal specimen-temperature level of 1700 F will be maintained by regulating the conductance across the annulus by gas-composition control.

(3) To study the degree of corrosion attack by GCRE reference gas, pressurized to 400 psi, on the cladding materials, this gas will continuously flow through a circuit formed within each specimen assembly by small-bore (5/16 in. in OD by 22-in. wall) tubes joined to the can headers by welding.

Data from nuclear-mock-up capsules exposed at BRR during July are summarized in Table Q-2. As can be seen, capsule-perturbation factors appear to depend somewhat on location along the length of the irradiation space. Also, some effect in unperturbed flux and, consequently, fission-heat-generation rate can be expected as a result of the GCRE loop. For example, the peak specimen flux with the loop in place is about 0.48×10^{13} nv; the calculated heat-generation rate corresponding to this flux is about 1900 Btu per in. of specimen length. With the loop removed, values are 0.34×10^{13} nv and about 1400 Btu/(hr)(in.). Heat-transfer calculations based on a 1700 F specimen temperature indicate that a spread in fission heat of this magnitude is well within that which can be compensated for by regulation of the gas composition in the annulus region.

Laboratory thermal-mock-up experiments have been hampered by failure of auxiliary electrical heating elements, and, consequently, only limited data were obtained during August. Those obtained, however, continued to be encouraging in that control of reference temperatures through regulation of the helium-argon composition in the mock-up annulus was again demonstrated. The fluctuation was ±10 F at a level of about 1200 F. In this situation, the power input was constant and the gas mixture was held at atmospheric pressure. At present, mock-up repairs are being made to permit proof tests of regulation at 1700 F.

During September, the final out-of-pile stages of the program will be initiated with emphasis placed on the following:

(1) Capsule and control-system final design

(2) Fabrication of capsule parts

(3) Reparation of hazards report

(4) Initial setup activity at reactor.

Effects of Irradiation

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

A study of the radiation stability of fuel-element materials for compact gas-cooled reactors includes (1) the evaluation of encapsulated solid and annularly loaded UO_2 clad with Inconel, and (2) the evaluation of in-pile-loop subassemblies containing fuel pins of solid UO_2 clad with Inconel.

TABLE Q-2. SUMMARY OF NUCLEAR-MOCK-UP DATA(a) FOR BRR II-2C CAPSULES

				Average Effective Specimen Flux Data			
		Nominal Unperturbed Flux at Corresponding Location, 10^{13}nv		With GCRE Loop in Place(c)		Without GCRE Loop in Place	
Capsule	Location of Geometric Center of Fueled Specimens with Respect to Grid Plate, in.	With GCRE Loop in Place(b)	Without GCRE Loop in Place	Flux, 10^{13}nv	Perturbation Factor	Flux, 10^{13}nv	Perturbation Factor
Top	21-15/16	0.72	0.42	0.37	0.51	0.20	0.48
	19-15/16	0.80	0.49	0.39	0.49	0.24	0.49
Center	15-1/16	1.10	0.78	0.48	0.44	0.34	0.43
	13-1/16	1.12	0.80	0.47	0.42	0.33	0.41
Lower	7-3/15	0.95	0.69	0.42	0.44	0.30	0.43
	6-3/16	0.87	0.63	0.34	0.39	0.25	0.40

(a) In the center capsule, actual fuel-bearing graphite specimens were used; in the others, the fuel was simulated by an aluminum-bronze alloy. Dosimeters consisted of gold foils (bare and cadmium covered).

(b) The GCRE loop occupies two positions, 52 and 53, created by the removal of two outer-row fuel-element subassemblies; the II-2C capsules are to occupy an adjacent face location, Position 64.

(c) Values for this case are extrapolated from various dosimeter data.

Capsule Program

Metallographic examination of the fuel pins from Capsule BMI-27-2 indicates that the fuel pins reached excessively high local temperatures, on the order of 2500 F or above. Two sections of the cladding have been examined metallographically from each of the four specimens which failed in this experiment. There is no evidence of any eutectic having been formed which might have caused the cladding to melt at lower temperatures. A chemical analysis will be performed on a sample of cladding in the melt area, however, in order to confirm this. Metallographic examination of the UO_2 from the failed specimens and sections from the two good specimens from this experiment has not been completed.

Loop Program

One in-pile-loop subassembly designated as Experiment IB-IαT was received at the Battelle Hot-Cell Facility during August, and postirradiation evaluation has been initiated. This assembly consisted of 19 fuel pins containing UO_2 pellets and clad with Inconel. Sixteen of the 19 fuel pins had concentric tubular spacers brazed to them in order to insure proper channel spacing. The postirradiation evaluation of this assembly will consist of channel-spacing measurements, visual examination of each pin, dimensional and bowing measurements of each fuel pin, and a leak check of each pin. A leak check on each fuel pin is required because of the high fission-product activity obtained on gas samples taken from the in-pile loop during irradiation.

The disassembly of the unit has been completed, and all measurements except bowing measurements and leak checking have been completed.

Pin 16, which was located in the outer ring of the element and at 12 o'clock with respect to the reactor, appears to have experienced a corrosive attack at a point 13 in. from the inlet end of the subassembly. This attack occurred at a fuel-pin spacer and in the area adjacent to the spacer. Pins 15, 17, and 6, which were adjacent to Pin 16, also experienced an attack on the face of the fuel pin that was in contact with the spacer from Pin 16. The cause of this attack has not been determined, but metallographic examination of Pin 16 and one of the fuel pins adjacent to it is planned. It is hoped that metallographic examination will reveal the cause of attack.

GCRE Critical-Assembly Experiments

R. A. Egen, J. W. Ray, W. S. Hogan, D. A. Dingee, and J. W. Chastain

During the past month experiments were completed which (1) evaluated the reactivity worth of mock-up scissor-acting safety and shutdown blades, (2) compared production GCRE-I elements and critical-assembly elements on the basis of fuel and steel equivalence, (3) measured flux and power distributions in the IB-IφT (oxide pin) fuel element, and (4) determined the coefficient of reactivity for boiling voids at the core-reflector interface.

Q-7

The safety and shutdown blades were mocked-up using two stainless steel blades approximately 3 in. wide by 12 in. long and tapering from 5/8 in. to 1/8 in. at the tip (core) end. The maximum worth of these blades occurs at a separation angle of about 15 deg. The worth at this separation is 0.40 per cent $\Delta k/k$ with bare blades and 1.14 per cent $\Delta k/k$ with 20-mil-thick cadmium foil covering the blades.

An experiment was conducted to determine differences in fuel and structural composition between critical-assembly fuel elements and production GCRE-I elements. The critical assembly was calibrated by measuring the reactivity effect of variations in fuel and stainless steel composition in critical-assembly elements. Corresponding reactivity measurements with production elements were made, and preliminary estimates of the differences in fuel and steel content from those in the critical assembly in the production elements are tabulated below.

Element Designation	Deviation in Fuel Content, g	Deviation in Steel Content, g
CE-1	-3.8	748
CE-2	2.3	796

A IB-IϕT mock-up element was inserted in the critical assembly in place of a standard critical-assembly element. This element contained 19 fuel pins, each approximately 0.22 in. in OD and having a 0.030-in. wall, arranged in a hexagonal array. The active fuel (UO_2) length was 22.75 in. The fuel enrichment in the outer 12 pins was about 24 per cent and in the inner 7 pins was about 48 per cent. Flux and power distributions were measured. These data are being analyzed. However, a preliminary analysis shows the ratios of power between the central pin and pins in the middle and outer rings to be 0.89 and 0.94, respectively. This ratio appears to vary azimuthally.

Studies of the void coefficient of reactivity were made at the core-reflector interface in order to estimate the reactivity effect of local boiling. In one series of tests various thicknesses of polyethylene were attached to the entire surface of the radial lead reflector which increases the local hydrogen density. By extrapolating these data, the effect of a reduced hydrogen density (boiling) can be estimated. In a second experiment the effect of voids on the upper axial reflector was estimated by lowering the water. Data from both sets of experiments are being analyzed.

During the coming month final data analysis for the above experiments will continue. Some additional experiments dealing with IB-IϕT mock-up elements may be undertaken. Preparation of an interim report summarizing studies with the ML-1 reactor mock-up and preparations for conducting critical-assembly experiments with the ML-1B mock-up will commence.

Q-8

IN-PILE-LOOP PROGRAM

G. A. Francis

Program activity on the in-pile recirculating gas loops located at the Battelle Research Reactor and Engineering Test Reactor continued during the present report period. At the BRR, preparations were completed for beginning the irradiation of the IB-IβT element. Data analyses were completed for a flux study of the nuclear mock-up (IB-IφT) of the IB-IT series of elements and for the gas samples taken during the operation of the IB-IαT element. At the ETR, the in-reactor loop piping was installed. During the reactor cycle following the installation, the loop was operated with a dummy element to check out the various subsystems and controls.

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

Preparations have been completed for beginning the irradiation of the IB-IβT element upon arrival of the unit at the BRR. Hazards approval has been received for operation of this fuel-element subassembly. A gas-monitoring system has been designed and equipment ordered to permit continuous monitoring of the loop gas for activity.

Gas samples were taken periodically from the loop coolant for checking the gas-radioactivity content during the operation of the IB-IαT element which ended on July 18, 1959. Both 100-channel and single-channel gamma-ray spectrometers were used to check these samples. Results of these analyses showed that the fission gases xenon-133, xenon-135, and krypton-85m were the major constituents with small amounts of krypton-87, krypton-88, and argon-41 also present. Prior to and during early periods of thermal cycling of the specimen, slight increases in the gas activity were detected. During the final period of operation, the total gamma-activity level reached an estimated 50 to 60 millicuries in the loop, indicating the possibility of a crack-type failure. Preliminary results from the hot-cell examination of the specimen indicate that a leak exists in the cladding of one of the fuel pins.

The thermal-neutron flux distribution was measured in the IB-IφT fuel element, the nuclear mock-up of the IB-1T series of elements. The axial peak-to-average flux ratio over the fuel-bearing length of the pin was nearly constant for all 19 pins, averaging 1.24. The flux variation was measured around the circumference of five of the pins and found to be about a factor of two, except for the center pin which showed a smaller variation. The ratio of the flux at the center of the pins to the average surface flux was determined to be 0.36 for the inner pins (48.5 per cent enriched) and 0.50 for the outer pins (24.5 per cent enriched).

The power distribution in the element was estimated from the flux measurements and from the activity of the fuel pellets. The results of the two measurements were in good agreement and indicated a fairly uniform power distribution in the element.

The flux measurements predict a total power of 29.9 kw for the ϕ and β elements (same fuel loading) and 33.5 kw for the α element (higher fuel loading). This latter value is in good agreement with the estimated 32 to 33 kw power determined by a heat balance on the loop during operation with the α element.

ETR Loop Program

J. V. Baum and E. O. Fromm

During the current report period, the in-reactor tube and the associated loop piping were installed. The hot check (operation for a full reactor cycle with a dummy specimen) of the loop has begun.

The hot check has included preheater operation at several power-input levels and variations of system flow from 0.5 to 0.9 lb per sec with the diverter valve in the maximum and minimum cooling positions. Temperature data from thermocouples located at various points in the loop and on the dummy element are being analyzed to define the heat-transfer situation in the system. Preliminary results show that the gamma heating of the in-reactor tube will present no problem during operation although, as was expected, coolant flow must be maintained during reactor operation.

The various instruments, controls, and mechanical components of the system performed satisfactorily with the exception of the blowers. During the initial period of operation, it was necessary to switch blowers frequently because of overheating of the bearings. The hot check has continued with one of the three blowers operating satisfactorily.

Plans call for replacement of the bearings during the next scheduled reactor shutdown. The loop operation will continue during the next reactor cycle to obtain satisfactory bearing performance of the three blowers, which will complete the hot check of the system.

RWD:CRT/all