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Report No. BMI-1366
C-25 Metallurgy and Ceramics
(M-3679, 22nd Ed. Rev. 1)

Contract No. W-7405-eng-92

PROGRESS RELATING TO CIVILIAN APPLICATIONS DURING JULY, 1959

by

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Classification cancelled (or changed to UNCLASSIFIED)
by authority of Letter from Declassification Branch dated 2-4-60
by J. C. Ridenour TISOR, date 4-1-60

August 1, 1959

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A. ASSISTANCE TO HAPO

F. R. Shober

The study of the creep properties of Zircaloy-2 at elevated temperatures is being made on annealed and on 15 per cent cold-worked material. The study also includes the effect of cyclic test temperatures as well as constant test temperatures. Long-term, 10,000 to 15,000-hr, tests have been initiated on annealed Zircaloy-2. In the study of the corrosive action of high-temperature water on defected Zircaloy-2-clad uranium specimens, four additional specimens have been tested.

The construction of the loop system for studying the rate of dissolution of silver chloride has been completed. This is a part of a program to develop a fuel-element leak-detection system. The development and evaluation of various forms of exchange media are being carried out. These exchange media will be further evaluated in future loop studies. In work on the development of a thermal-neutron-flux monitoring system, the major effort has been in the evaluation of a series of promixing ceramic resistor compositions. The ceramic elements are to be used for the flux-probe instrument.

Mechanical Properties of Zirconium Alloys

L. P. Rice and J. A. VanEcho

A study of the creep properties of cold-worked (15 per cent) and of annealed Zircaloy-2 sheet at 290, 345, and 400 C is continuing. Long-term (10,000 to 15,000 hr) creep-property determinations of the cold-worked material are nearing conclusion. A series of thermal-cycling creep tests on both annealed and cold-worked material is in progress. A few long-term, approximately 15,000 hr, creep tests on annealed Zircaloy-2 sheet have been started and others are scheduled to duplicate the program on cold-worked sheet.

Cycling Zircaloy-2 between room temperature and 290 or 345 C results in a larger amount of creep deformation than is produced in constant-temperature tests when data for creep tests of 3000 hr duration are compared. At 290 C, the cycling produces about 6-1/2 per cent more total creep than under static-temperature conditions. At 345 C, creep acceleration is stress dependent, with the highest stress (30,000 psi) producing about 40 per cent more creep, and the lowest stress (15,000 psi) about 3-1/2 per cent more creep than in the constant-temperature tests. The increase in total creep deformation is based only on the time at the greatest temperature during test. If the results are compared for total time at test, then the increase in the total creep resulting from cycling is somewhat less.

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

E. F. Stephan and F. W. Fink

The lapse-time motion-picture study of the corrosive action of high-temperature water on defected fuel elements was continued.

Four elements were studied this month. The same type of failure observed at the defect previously and described in BMI-1357 was also seen in the specimens examined this month. The equipment had to be modified to accomodate the large diameter specimens, D-8 and a tubular specimen. Table A-1 summarizes the data for all the specimens studied so far.

The tubular specimen had a defective end weld. It is planned to study another tubular specimen as well as a duplicate to Specimen C-8. This will complete the study and a final report will be submitted.

Development of a Fuel-Element Leak Detector

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

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

The construction of the loop system for studying the rate of dissolution of silver chloride has been completed. This study will be initiated in early August. After concluding the study of AgCl dissolution, the factors which affect the isotopic exchange reaction, i.e., column size, particle size, flow rate, etc., will be investigated.

The development and evaluation of various forms of exchange media are being carried out simultaneously with the above work. Silver chloride-coated silver shot has been prepared by anodic formation of the silver chloride in place. Sturdy silver chloride films approximately 0.5 mil thick have been obtained by this technique.

The physical properties of the silver chloride-coated silver shot and other exchange material being prepared will be evaluated in future loop studies.

Thermal-Neutron-Flux Monitoring System

D. R. Grieser, J. W. Lennon, M. J. Snyder and C. V. Weaver

The objective of this program is to develop a thermal-neutron-flux monitoring system for the HAPO reactors. The basic unit for the proposed system is a flux sensor

TABLE A-1. CORROSION IN HIGH-TEMPERATURE WATER OF DEFECTED[(a)] NATURAL-URANIUM FUEL CORES EXTRUSION CLAD WITH ZIRCALOY-2

Specimen	Water Temperature, C	Time to First Noticeable Swelling at Defect, hr	Time to First Gas Evolution, hr	Time to Complete Reaction, hr	Uranium Dissolved at Complete Reaction, g
G-11	200	8	15	24	136.95
G-12	225	6	7.5	13	135.20
G-13	250	1.7	2.5	10	132.28
G-14	300	1.0	1.5	5	119.63
G-22	300	0.7	1.3	6.6	136.63
G-16[(b)]	300	0.7	1.25	6.25	134.45
G-17[(c)]	200	0.75	1.1	6.1	135.91
G-15	345	0.4	0.8	3.17	134.55
C-8[(d)]	300	0.3	0.75	2.5	156.81
D-8[(e)]	300	24.0	49.0	54.0	335.30
Tubular	300	0.5	0.9	5.5	366.03

(a) The defect consisted of a 25-mil hole drilled through the nominal 30-mil cladding.
(b) Cladding was nominally 20 mils thick.
(c) Cladding was nominally 10 mils thick.
(d) Specimen was beta treated and water quenched.
(e) Specimen had uranium-2 w/o zirconium core, diffusion treated.

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based on the principle of balancing the temperatures of a fission heated-uranium fueled sensing element and a nearly identical electrically heated balancing element.

In work on the development of ceramic elements for the flux-probe instrument, the major effort is on the evaluation of a series of promising resistor compositions. A series of compositions has been prepared in which electrical resistivity at room temperature and UO_2 content met the specifications for a prototype element (0.032 to 3.2 ohm-in.). The composition range was:

Material	Range, w/o
$MoSi_2$	37.0 - 44.0
UO_2	19.7 - 19.1
Al_2O_3	43.3 - 36.9

Specimens were fired in a purified hydrogen atmosphere for 2 hr at 2900 F.

After 128 hr at 1200 F in an argon atmosphere, the resistance of all specimens had decreased slightly.

In other studies made on compositions within the range just described, the effects of using organic binders to aid in fabrication and of varying the particle size of the raw materials are being investigated.

In future work, the major effort will be concerned with determining the stability of the resistors when exposed under various conditions of interest. Also, techniques for fabricating resistors of the required size and shape will be investigated further, as will the effects on resistivity of such variables as raw-material particle size and type, impurities, binder addition, and mixing and fabricating procedure.

Efforts are being made to improve the brazing technique developed for making integral low-resistance junctions between the ceramic elements and the electrical leads.

B. DEVELOPMENTS FOR ALUMINUM-CLAD FUEL ELEMENTS

R. J. Carlson and N. E. Daniel

Data obtained by differential thermal analysis show that the binary aluminum-uranium and aluminum-nickel eutectic temperatures are lowered approximately 16 C in ternary combination. The program concerned with the preparation of aluminum-35 w/o uranium alloys containing ternary additions is being continued. Emphasis will be placed upon determining the casting characteristics of the alloys containing up to 3 w/o of either tin or zirconium and upon measuring the physical properties of alloys in the as-extruded condition.

Aluminum-Uranium-Nickel Eutectic Alloys

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

The composition and temperature of the aluminum-uranium-nickel ternary eutectic in high-aluminum alloys are being determined. A number of alloys having compositions that generally bracket the area where a ternary eutectic would be expected have been prepared by induction-melting techniques. These are being studied by metallographic techniques and thermal analysis.

Solidus temperatures of these alloys as determined by differential thermal analysis are as follows:

Alloy Composition (Balance Aluminum), w/o	Solidus Temperature, C
13.3 uranium	640
5.9 nickel	640
21 uranium-3.1 nickel	625
18.9 uranium-1.4 nickel	626
15 uranium-3 nickel	624
8 uranium-2.4 nickel	624
8 uranium-4 nickel	622
7.9 uranium-5.5 nickel	626

The data show that the binary eutectic temperatures are lowered approximately 16 C in ternary combination to about 624 ± 2 C at the ternary eutectic.

Metallography indicates that none of the above alloys has exactly the ternary eutectic composition. Consequently, three additional aluminum-base alloys have been prepared containing 13.1 w/o uranium and 5.6 w/o nickel, 12 w/o uranium and 8 w/o nickel, and 14.2 w/o uranium and 6.8 w/o nickel in order to aid in its location.

Preparation of Aluminum-Uranium Alloys

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

Previous experience with the aluminum-35 w/o uranium alloys has shown that improvements in the casting and fabricating characteristics of the alloy can be obtained through the use of suitable ternary additions. Among the additions which have improved the qualities of the aluminum-35 w/o uranium alloy are silicon, tin, and zirconium. It is believed that the improvements noted can be attributed to an increase in the matrix-to-compound ratio that is brought about by the inhibition of the UAl_3-to-UAl_4 reaction and to decreases in primary particle size. Although silicon is beneficial from an alloying viewpoint, it is known to introduce complications in the recycling processes. Since tin and zirconium should be equally as effective as silicon in inhibiting the UAl_3-UAl_4 reaction, a program of research concerned with the optimization of the contents of these two additions is being continued at Battelle. Emphasis will be placed upon determining the casting characteristics of alloys containing up to 3 w/o of either tin or zirconium and upon measuring the physical properties of alloys in the as-extruded condition.

In the first part of the program the effects of additions from 0.5 to 3.0 w/o of tin and zirconium on the castability and fabricability of the aluminum-35 w/o uranium alloy will be investigated. Castability will be evaluated on the basis of alloy fluidity, and on the basis of casting homogeneity and soundness. Fabricability will be evaluated directly on the basis of the extrusion pressures necessary to effect a 16:1 reduction in a cast alloy billet, the surface condition of the extruded material, and the integrity of the as-extruded alloys.

Work in the immediate future will be concerned with the production and evaluation of twelve, 3-in.-diameter by 10-in.-long ingots needed for the portion of the program outlined above. The evaluation of these ingots will include radiographic examination of sections from each ingot, metallographic examination of at least two specimens from each ingot, and chemical analyses and X-ray diffraction studies of selected specimens.

C. RADIOISOTOPE AND RADIATION APPLICATIONS

M. Pobereskin

During the past month, the study of the radiation chemistry of inclusion compounds was suspended. For the systems investigated, there was little or no difference between the hydrocarbon degradation products produced by irradiation of the inclusion-compound systems and those produced by irradiation of bulk hydrocarbons. A low-yield host-guest reaction in the urea complexes was observed, but the complex nature of the reaction products is of little practical interest. Studies are continuing on the use of tracers in cement quality control, the use of intrinsic tracers for process control, radiation-induced graft polymerization, and radiation-induced nitration reactions.

Activation analysis for magnesium and silicon in cement and cement raw materials does not appear feasible. Manganese-56 masks the magnesium-27, and the degree of activation of silicon-31 is too low. Better dosimetry and standards will be necessary if activation analysis is to be practical. Radiometric analysis for calcium is being studied, using a phosphate precipitate and phosphorus-32 as a tracer.

Laboratory experiments, using simulated process solutions and copper-64 as a tracer, are being carried out to study the copper-removal step of the Sherritt-Gordon nickel process. A short-lived radioisotope, which will follow the iron chemistry, is being sought as a substitute for iron-59. This will be used to develop a system for control of iron removal from the cobalt-leach circuit of the process.

In the study of factors affecting graft polymerization, the source and nature of free radicals in the polymethacrylates is being studied using measurements of electron paramagnetic resonance. These measurements will be continued and applied to further grafting studies.

Preliminary runs, in the absence of radiation, have been completed for the study of nitration of hydrocarbons. Analysis of data from these experiments and additional experiments of this type will be used to guide the experimental program for the system under irradiation.

Development of Radioactive-Tracer Quality-Control Systems

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

The study of neutron-activation analysis of cement raw materials was continued during July. Nine samples of limestone, clay, slag, and sand have been irradiated. Attempts were made to analyze for aluminum, calcium, magnesium, manganese, silicon, and sodium by measuring the areas under the various gamma photopeaks and comparing them with suitable standards.

Attempts to analyze for magnesium and silicon in these materials will be abandoned. All the samples were found to have considerable manganese-56 activity, which completely masks out the activity from the magnesium-27. Both radioisotopes have gamma photopeaks at 0.85 Mev. It was not possible to obtain sufficient 2.6-hr silicon-31 activity for analysis in the samples irradiated due to the relatively short irradiation times (3 to 5 min).

The results obtained for the nine samples analyzed are summarized in Table C-1. The data are semiquantitative at best. In only one instance was it possible to compare the manganese and sodium analyses. In this case the manganese results were 17 per cent low, and the sodium results were 50 per cent high. Manganese assays taken on the hundred-channel analyzer and on a single-channel gamma spectrometer agreed within 5 per cent. The calcium results were 50 per cent high in both cases, which indicates the calcium standard may not be accurate. A new calcium standard will be used in future runs. The aluminum results were in error by 9 to 50 per cent. The best aluminum results were obtained in the last run in which more complete dosimetry was used to correct for thermal-flux differences between samples. The large error in the aluminum results for the sand sample has not been explained. In future runs more complete dosimetry and improved standards will be used in an attempt to improve accuracy.

In the course of the above analyses it was determined that the calcium content of a sample must be greater than 20 to 30 w/o in order to obtain enough calcium-49 activity for analysis. It was also determined that about 90 per cent of the sodium-24 activity induced in the Al_2O_3 powder previously used as an aluminum standard resulted from a sodium impurity and only 10 per cent was due to the $Al^{27}(n, \alpha)Na^{24}$ fast-neutron reaction. In the future powdered aluminum metal of 99.999 per cent purity will be used as an aluminum standard.

Work on the radiometric analysis of calcium in cement has continued during this report period. Recent investigations have shown that the precision of the EDTA titration using silver-110 iodate as an end-point indicator is limited by the proximity of the silver-EDTA and calcium-EDTA complex stabilities. Consequently, a new method involving the precipitation of calcium as $Ca_3(PO_4)_2$, using phosphorus-32 as an indicator, is being investigated.

Future work will include the development of the calcium analysis by $Ca_3(PO_4)_2$ precipitations, and improvement of the activation-analysis techniques using more accurate dosimetry and improved standards.

Use of Intrinsic Radioactive Tracers for Process Control

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

In the study of intrinsic tracers during July, effort was concentrated on studying radiotracer control of the copper-removal step in the Sherritt-Gordon nickel-refining process. In addition, a search was initiated to find a radiotracer other than iron-59 which may be used to control the iron-removal step in the Sherritt-Gordon process.

TABLE C-1. RESULTS OF ACTIVATION ANALYSIS OF CEMENT RAW MATERIALS

	Experimental and Calculated Analyses, w/o									
	Al_2O_3		CaO		Mn_2O_3		Na_2O		SiO_2	
Sample	Experimental	Calculated	Experimental	Calculated	Experimental	Calculated	Experimental	Calculated	Experimental	Calculated
Limestone 8768	(a)	3.8	(a)	34.3	0.049	--	0.18	--	(b)	16.6
Clay 7440	(a)	8.7	(a)	4.0	0.073	--	1.40	--	(b)	65.7
Slag 19459	(a)	12.8	(a)	44.9	0.19	0.23	0.18	0.12	(b)	35.27
Limestone 4983	0.33	0.6	80.0	53.4	0.002	--	0.043	--	(b)	0.7
Limestone 7604	2.13	1.7	76.2	49.6	0.016	--	0.14	--	(b)	5.6
Clay 6956	9.54	7.7	(c)	18.3	0.046	--	1.13	--	(b)	44.0
Shale 8772	13.5	16.2	(c)	3.9	0.040	--	0.48	--	(b)	52.2
Shale 8773	9.89	10.8	(c)	Trace	0.022	--	0.63	--	(b)	64.0
Sand 4299	4.35	0.8	(c)	0.6	1.06	--	0.29	--	(b)	89.2

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

(b) Silicon-31 activity was too low for analysis.

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

Laboratory experiments were carried out to determine the sensitivity of measuring copper-64 activity by using a 2 by 2-in. NaI (Tl) crystal scintillation probe immersed in the simulated process solution. The results indicate that the maximum counting efficiency attainable by this method is about 12 per cent of the copper-64 activity per liter of solution.

Experimental analysis of exchange between copper in solution and copper sulfide precipitate is being continued. The results of these experiments should be available in August.

Project activity during next month will consist of concluding the copper-exchange study, developing more sensitive methods of measuring copper activity in solution, and searching for a suitable substitute for the iron-59 tracer. Sherritt-Gordon will be requested to supply actual process materials for the latter phase of the laboratory analysis.

Graft-Polymerization Studies

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

Work continued during July on the study of radiation-induced changes in polymers leading to graft copolymerization. During this month, it was found that grafting occurs to some extent during the fractionation treatment of polymers. Several possible reasons for this effect exist. Among these are the existence of residual peroxide or stable free radicals on the polymer chain and grafting due to chain transfer during the thermally initiated polymerization of vinyl pyrrolidone. In order to remove these possibilities, a number of steps were taken. The polymethacrylates were prepared by radiation polymerization and heated at 100 C for 24 hr under vacuum. The polymer samples were then sealed in ampoules and subjected to grafting conditions with and without preirradiation. The fractionation procedure was modified to eliminate any heating of the grafted specimen until the excess monomer was removed.

In order to more fully explore the formation of free radicals, work was begun, using electron paramagnetic resonance (EPR), to obtain an independent measurement of free-radical concentration. Samples of polymethyl- and polybutylmethacrylates were sealed in ampoules at about a pressure of 0.1 mm of mercury and irradiated. The spectra of the irradiated polymers indicated at least two species of sites were present, one stable and one unstable. After the transitory site decayed, the tubes were opened to the air. The apparent active-site concentration increased by a factor of two and rapidly decayed with a half-life of approximately 15 min. Another sample of polymethylmethacrylate was irradiated in a tube under a vacuum of about 0.1 μ. EPR measurements disclosed only one type of active site which decayed relatively slowly under this condition. After 24 hr, about one-half the sites remained. Upon exposing the contents of the tube to air, the apparent site concentration increased by almost two orders of magnitude and then decayed with a half-life of 15 min. Repetition of the above procedure with polybutylmethacrylate instead of polymethylmethacrylate disclosed similar results.

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During the coming month, EPR measurements of site concentration will be continued on these and other polymethacrylates and comparisons made with the chemical method of measurement to gain further insight into the reported effects. Grafting studies will also be made, and graft chain length will be calculated.

Nitration of Hydrocarbons

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

During the past month construction of the experimental equipment for the study of nitration of hydrocarbons was completed. The equipment was tested and found to operate successfully over the desired range of variables.

Thermal runs (in the absence of radiation) with the nitric acid-cyclohexane system are under way. These will be used to provide reference data on the effect of temperature on product yields. Two thermal runs have been made, but analyses have not been completed. Qualitative tests showed the presence of the nitro group and possibly the nitrato group. It has been observed that the yields of oxidation products (primarily adipic acid) increase with increasing temperature. The formation of adipic acid may complicate the study of nitration reactions at high temperatures. When the results of additional thermal runs become available, it may be necessary to reappraise the approach to the study of nitration.

During August additional thermal runs will be made and the analyses of current runs will be completed. Dosimetry measurements in the irradiation vessel and the first irradiation runs are also scheduled during this period.

F. RESEARCH FOR AEC REACTOR DEVELOPMENT DIVISION PROGRAM

S. J. Paprocki and R. F. Dickerson

REACTOR MATERIALS AND COMPONENTS

R. F. Dickerson

The investigation directed toward obtaining a more fundamental insight into the role of valence compensation in the stabilization of uranium dioxide is being continued. Work with substitution of CaO for some of the La_2O_3 or the Y_2O_3 addition indicates that, while it appears feasible, in most cases the materials produced have appeared slightly less stable than some of the binary compositions. The effects of high temperature and high pressure on U_3O_8 continue to be studied. The oxygen limits of the gamma phase resulting from application of high temperature and pressure on U_3O_8 are being investigated, and further work is being done on U_3O_8 with a Bridgman-anvil high-pressure die.

The experimental work connected with the study of irradiated zirconium-2 w/o uranium alloy hydride was completed, and the results were reported in BMI-1357. No further reports will be included in this section.

The irradiation of capsules containing specimens of Type 347 stainless steel has been continued in the ETR. Unfortunately, because of erratic operation of the reactor, only about 1 day of exposure at full power has been achieved during the past month. Four nickel dosimeter wires have been returned to Battelle, and these are being analyzed for determination of the actual neutron exposure accumulated by the specimens during ETR Cycles 15, 16, and 17. Because of the shortage of suitable test space, the insertion of the gamma-heat capsules has been indefinitely delayed.

Some niobium-base binary alloys being studied as possible alternate cladding material for the EBR were arc cast into approximately 5-lb ingots. Certain of these alloys could not be cold rolled even after breakdown of the cast structure. At the same time, a control ingot of unalloyed niobium could not be cold rolled. This material should have been readily cold fabricated. Analytical data obtained to date indicate oxygen pickup during the fabrication. The source of this contamination is being sought. Corrosion testing of niobium-base alloys in 600 and 680 F water has been continued, and results of 196-day tests are available. These results indicate that alloys prepared from commercially pure niobium are as corrosion resistant as those prepared from high-purity stock. To date the most corrosion-resistant alloys are ternaries containing 28.2 a/o titanium-6.1 a/o chromium and 10 a/o zirconium-5 a/o iron, and all alloys containing more than 40 a/o zirconium.

Because of the importance of Zircaloy-2 as reactor structural and fuel-element cladding material, it is important to understand the effect of irradiation on creep strength. With this in mind, a program concerned with the performance of relatively simple in-pile creep studies has been initiated. Initial planning for this program is well under way.

F-2

Valence Effects of Oxide Additions to Uranium Dioxide

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

An investigation is being conducted which is directed toward obtaining a more fundamental insight into the role of valence compensation in uranium dioxide containing certain oxide additions. Uranium dioxide upon oxidation undergoes a phase transformation to U_3O_8, which is a volatile oxide. In order to stabilize the more refractory uranium dioxide fluorite structure, materials of lower valence are added in solid solution such that, as the uranium goes to higher valence state upon oxidation, valence compensation can occur. The general reaction is of the type,

$$RO_2 + R_2O_3 \rightarrow 3RO_2 \text{ (oxygen defect fluorite structure)},$$

which upon oxidation yields,

$$RO_3 + R_2O_3 \rightarrow 3RO_2 \text{ (stoichiometric)}.$$

Details of the valence-compensation criteria have been given in BMI-1318.

A detailed discussion of valence compensation with respect to stabilization of ions in certain valence states has been given by Kroger and Vink.* Proceeding from a consideration of the defect solid state, methods are discussed by which ions are stabilized in certain valence states to form a condition of "balanced valence" in crystals. The operative mechanism of valence compensation is entirely analogous to the valence-compensation concept employed in the present investigation.

Important differences are apparent, however, between the system presently being investigated and those systems discussed by Kroger and Vink. The present work involves not only the stabilization of an ion in a certain valence state (U^{+6}) but results in stabilization of the structure itself. In addition, in some systems such as the NiO system containing Li_2O, a stoichiometric relationship exists between the amount of Li_2O added and the amount of Ni^{+3} formed.** In the uranium-lanthanum oxygen system, however, stoichiometry apparently occurs for only one specific composition, namely,

$$UO_2 + La_2O_3 + 1/2\ O_2 \rightarrow (U^{+6}La_2{}^{+3})O_6.$$

Current work has been directed toward reducing the amount of additive required for stabilization of uranium dioxide. In a binary system it is found that the most stable materials from the standpoint of volatility require 55 mole per cent of a trivalent additive such as La_2O_3 or Y_2O_3. Since a divalent material should be more effective in valence compensation than a trivalent additive, the partial substitution of CaO for La_2O_3 or Y_2O_3 was investigated. Use of CaO alone is not possible since the solubility limit is inadequate for complete compensation to occur.

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*Solid State Physics, Vol 3, Academic Press, Inc., New York (1951), "Relations Between the Concentrations of Imperfections on Crystalline Solids" (F. A. Kroger and H. J. Vink), pp 307-405.

**Semiconducting Materials, Academic Press, Inc., New York (1951), "Oxide Semiconductors" (E.J.W. Verwey), pp 151-61.

F-3

A new series of solid solutions has been prepared and evaluated. The analyzed compositions of these materials in mole per cent are:

UO_2-22 La_2O_3-18 CaO

UO_2-26 La_2O_3-23 CaO

UO_2-20 Y_2O_3-19 CaO

UO_2-20 Y_2O_3-20 CaO

UO_2-22 La_2O_3-21 CaO

These compositions were oxidized for periods of 1, 5, and 20 hr at 1760 C in dry flowing air. The relative stabilities of the materials were evaluated by weight-change data. Weight changes observed ranged from a gain for UO_2-22 mole per cent La_2O_3-21 mole per cent CaO for 20 hr to a weight loss of 15 per cent in 5 hr for one sample of UO_2-20 mole per cent Y_2O_3-20 mole per cent CaO. The latter sample represents an anomalously large weight loss. The average of these materials appears to be of the order of a few per cent weight loss, which is equivalent to the vaporization experienced with the binary systems containing the same total amount of additive. Thus, while it appears feasible to substitute CaO for trivalent additives, with one exception the materials produced to date have exhibited higher losses than previously experienced in the most stable compositions in the binary systems. Experimental work will continue to complete the investigation of the ternary oxides.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

An investigation is being made of the effects of combined high pressure and high temperature on reactions of uranium oxide with various mixed oxides. Current work has been devoted to a detailed study of the effects of high pressure and temperature on U_3O_8. Results indicate that the direct insertion of thermocouples into the sample region greatly facilitates the loss of oxygen from the sample, allowing reduction of U_3O_8 to occur. The reduction was observed to occur at temperatures as low as 500 C at pressures to 60,000 atm.

Work will continue to better define the effect of high pressure and high temperature on U_3O_8. Analysis of the oxygen content of the samples subjected to high pressure will be performed by microbalance techniques. This analysis is being made to confirm that the cubic U_4O_9 structure, frequently observed during the high-pressure work on U_3O_8, contains an excess of oxygen. In addition, the oxygen limits of the new gamma phase of U_3O_8, formed under pressure, will be investigated. These results, together with those obtained on U_3O_8 by use of the Bridgman-anvil high-pressure die, should complete the investigation of the behavior of U_3O_8 under pressure. The study of reactions with mixed oxides will be continued at higher temperatures than used previously.

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Irradiation-Surveillance Program on Type 347 Stainless Steel

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

A program is in progress to determine the effect of irradiation on the mechanical properties of AISI Type 347 stainless steel. The program is being conducted in support of the KAPL-C-33 loop and other loops using Type 347 stainless steel in the ETR. Specifically, the program is for the purpose of determining changes in mechanical properties resulting from fast-neutron irradiation (neutrons having greater than a 1-Mev energy) upon Type 347 stainless steel at 120 and 600 F. Fast-neutron damage data are available for this material at exposures up to 3.76×10^{21} n per cm^2. However, it is intended to measure tensile, cyclic-strain fatigue, and impact properties of Type 347 stainless steel at exposures well beyond this level, with the intent being to stay 6 months ahead of exposure levels accumulated by the KAPL-C-33 loop. The KAPL loop will ultimately obtain a total fast-neutron exposure of approximately 1.6×10^{22} n per cm^2.

Three capsules containing tensile and cyclic fatigue specimens are planned for an early ETR loading. The specimens will be irradiated at process-water temperature and will be subjected to postirradiation annealing studies in order to provide a comparison with data to be obtained from specimens irradiated near 600 F.

Total irradiation exposures for the "cold" capsules (irradiated at process-water temperatures) as of the end of ETR Cycle 17 (June 14, 1959) are listed in Table F-1. The ETR has been largely inoperative since June 14, having accumulated only about 1 day of exposure at full power. Table F-1 also includes a list of current and proposed irradiation parameters of the various capsules prepared for the surveillance program.

Four nickel dosimeter wires, irradiated in the K-8 position of the ETR during Cycles 15, 16, and 17, have been returned to Battelle for determination of flux encountered during these cycles. The wires will be analyzed at Battelle, and one wire will be returned to the ETR for analysis in order that a comparison of ETR and BMI flux values can be made.

One thermocouple-lead capsule and seven capsules without leads are awaiting insertion in the ETR. The capsules are designed to operate at about 600 F in a position having a fast flux greater than 1.0×10^{14} nv and a gamma flux of about 18 w per g. These irradiation conditions are considered the minimum requirements necessary to initiate this portion of the surveillance program. At the present time, ETR reactor space of this quality is not available.

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

Capsule	Type Specimens in Capsules	Proposed Irradiation Temperature, F	Approximate Removal Date(a)	Approximate Exposure at Time of Removal(b), nvt	Total Exposure as of June 14, 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}	2.260×10^{21}	3.120×10^{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}	1.327×10^{21}	2.468×10^{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}	3.198×10^{21}	2.434×10^{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}	1.499×10^{21}	2.477×10^{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	0.84×10^{22}	2.49×10^{21}	2.840×10^{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	0.61×10^{22}	3.389×10^{21}	2.722×10^{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}	2.994×10^{21}	3.079×10^{21}	ETR, K-8-NW	Being irradiated

TABLE F-1. (Continued)

Capsule	Type Specimens in Capsules	Proposed Irradiation Temperature, F	Approximate Removal Date(a)	Approximate Exposure at Time of Removal(b), nvt	Total Exposure as of June 14, 1959, nvt		Location	Remarks
					Top	Bottom		
BMI-24-15	Impact	600	--	--	--	--	ETR	To be irradiated
BMI-24-16	Impact	120	June, 1960	0.61×10^{22}	3.052×10^{21}	2.886×10^{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

(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 capsules.

F-7

Development of Niobium-Base Alloys

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

Utilization of a niobium-base alloy as an alternate cladding material in a future core design for the EBR requires evaluation of the fabricability and of selected mechanical and physical properties of these alloys. The alloys being studied are niobium-1 w/o chromium, niobium-2 w/o chromium, niobium-4.5 w/o zirconium, niobium-10 w/o tantalum-2 w/o chromium, niobium-20 w/o titanium-1.5 w/o chromium, niobium-40 w/o titanium-10 w/o aluminum, unalloyed niobium, and an alloy of vanadium-10 w/o titanium-1 w/o niobium for comparison with the niobium alloys.

Melting, hot and cold fabricability, metallographic examinations, chemical analyses, and a short heat-treatment study have been finished. Edge cracking of the forged ingots occurred during cold rolling. The ingots were sealed in stainless steel packs and annealed at 1050 C for 1 hr and then furnace cooled. After annealing, cold rolling of the ingots was continued. 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, but the material in the center of the sheet was sound. The rolled sheet was sheared into blanks from which tensile and corrosion specimens are being prepared.

The unalloyed niobium and niobium-40 w/o titanium-10 w/o aluminum failed to cold roll. The niobium-40 w/o titanium-10 w/o aluminum alloy fractured into two pieces and showed a large-grained macrostructure. This alloy can probably be warm fabricated and attempts to do this will be made. The unalloyed niobium should have been readily cold fabricable, but analysis of the forged and rolled slab indicates that the oxygen content had increased from 200 to 800 ppm, apparently reducing ductility enough to cause failure. The source of this contamination is being sought.

Attempts to hot roll the niobium-2 w/o chromium alloy and the niobium-10 w/o tantalum-2 w/o chromium alloy encapsulated in evacuated stainless steel will be made at 2000 F. If these alloys fail to fabricate at 2000 F, higher rolling temperatures may be employed.

The mechanical properties of alloys which were fabricable will be determined at 650 and 800 C. The thermal conductivity will be determined on the three alloys having the better tensile properties at 800 C. In the meantime, sheet-to-sheet weldability of the cold-fabricable alloys will be studied.

Development of Corrosion-Resistant Niobium Alloys

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and F. W. Fink

The development and evaluation of niobium-base alloys for possible service in pressurized-water reactors are being continued. Corrosion data and elevated-temperature properties are being obtained on high-purity-base alloys prepared on the

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	112	-1.47	42	Disintegrated	28	Disintegrated
10.5 Zr	--	--	196	0.67	56	0.10
26.1 Zr	--	--	196	0.07	--	--
35.7 Zr	--	--	196	0.66	--	--
45.7 Zr	--	--	196	0.55	--	--
1.08 W	--	--	196	-2.60	56	0.82
4.67 W	--	--	196	-29.3	--	--
9.56 W	--	--	7	Cracked	--	--
2.45 Mo	--	--	196	-7.10	56	0.77
5.20 Mo	--	--	196	-1.30	56	0.81
7.40 Mo	--	--	196	0.62	--	--
4.42 V	--	--	196	0.42	56	0.82
6.59 V	--	--	196	0.73	56	0.66
8.93 V	--	--	196	0.59	56	0.57
10.7 V	--	--	196	0.78	--	--
13.7 V	--	--	196	0.50	--	--
24.2 V	--	--	196	0	--	--
4.90 Fe	--	--	196	0.10	56	1.58
9.41 Ti	--	--	196	0.65	56	0.63
18.8 Ti	--	--	196	0.48	--	--
24.3 Ti	--	--	196	0.52	--	--
30.5 Ti	--	--	196	0.40	--	--
33.8 Ti	--	--	196	0.33	--	--
12.0 Ti-0.5 Cr	--	--	196	0.66	--	--
20.2 Ti-2.1 Cr	--	--	196	0.39	--	--
28.2 Ti-6.1 Cr	--	--	196	0.20	--	--
12.0 Ti-4.2 Mo	--	--	196	0.64	--	--
17.4 Ti-6.2 Mo	--	--	196	0.54	--	--
23.1 Ti-7.8 Mo	--	--	196	0.45	--	--
10.4 Ti-5.0 V	--	--	196	0.56	--	--
16.1 Ti-8.4 V	--	--	196	0.40	--	--
22.6 Ti-11.0 V	--	--	196	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	140	0.67	140	-1.04	140	-17.4[(a)]
7.18 Mo	140	0.61	140	0.55	70	Cracked
12.5 V	140	0.39	140	0.46	140	0.70
46.8 Zr-5.06 Ti	112	0.30	112	0.78	112	1.58[(b)]
11.2 Ti-3.2 Mo	112	0.20	112	0.39	112	0.69
18.8 Ti-8.7 Mo	112	0.47	112	0.49	112	0.46
9.9 Zr-9.4 V	28	0.16	28	0.30	28	0.21
5.7 Zr-11.4 V	28	0.12	28	0.24	28	0.31
9.1 Ti-6.3 Cr	28	0.11	28	0.24	28	-0.85
High-Purity Niobium, Rocking-Hearth Melts						
Unalloyed Nb	--	--	56	1.28	--	--
Unalloyed Nb	--	--	28	-22.5	--	--
			28	0.48	--	--
1 Zr	--	--	56	-52.9	--	--
5 Zr	--	--	28	0.72	--	--
10 Zr	--	--	28	-0.13	--	--
40 Zr	--	--	28	0.33	--	--
65 Zr	--	--	28	0.50	--	--
75 Zr	--	--	28	0.74	--	--
90 Zr	--	--	28	0.41	--	--
2.5 Ti	--	--	56	0.36	--	--
10.0 Ti	--	--	56	0.46	--	--
25.0 Ti	--	--	56	0.34	--	--
1 Cr	--	--	56	1.03	--	--
5 Cr	--	--	56	0.64	--	--
10 Cr	--	--	28	0.26	--	--
1 Fe	--	--	28	0.78	--	--
10 Zr-5 Ti	--	--	28	0.33	--	--
25 Zr-5 Ti	--	--	28	0.43	--	--
25 Zr-15 Ti	--	--	28	0.51	--	--
25 Zr-25 Ti	--	--	28	0.21	--	--
35 Zr-5 Ti	--	--	28	0.32	--	--
35 Zr-15 Ti	--	--	28	0.23	--	--
45 Zr-5 Ti	--	--	28	0.24	--	--

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
10 Zr-5 Mo	--	--	28	0.16	--	--
35 Zr-5 Mo	--	--	28	0.34	--	--
45 Zr-5 Mo	--	--	28	0.24	--	--
35 Zr-5 Al	--	--	28	0.51	--	--
45 Zr-5 Al	--	--	28	0.25	--	--
10 Zr-5 Cr	--	--	28	0.24	--	--
45 Zr-5 Cr	--	--	28	0.24	--	--
10 Zr-5 Fe	--	--	28	0.20	--	--
2.5 V	--	--	56	0.53	--	--
2.5 V-2.5 Ti	--	--	28	0.40	--	--
2.5 V-2.5 Mo	--	--	28	0.39	--	--
2.5 V-2.5 Fe	--	--	28	0.46	--	--
2.5 V-2.5 Cr	--	--	28	0.37	--	--
2.5 V-2.5 Al	--	--	28	0.49	--	--
5 V-2.5 Zr	--	--	28	0.20	--	--
5 V-25 Zr	--	--	28	0.63	--	--
5 V-35 Zr	--	--	28	0.64	--	--
5 V-45 Zr	--	--	28	0.27	--	--

(a) Erroneously reported in BMI-1357 as having disintegrated in 28 days.

(b) Erroneously reported in BMI-1357 as a weight loss.

basis of screening tests conducted on a series of commercial-purity niobium-base alloys. Additional screening alloys are being prepared to improve fabricability without losing high-temperature strength and corrosion resistance.

Corrosion testing of the niobium alloys in 600 and 680 F water and 750 F 1500-psi steam has been continued. Results after exposure times ranging up to 196 days are presented in Table F-2. The results to date indicate 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 exhibits a much longer corrosion life. Alloys which possess adherent tarnish films and appear most corrosion resistant are ternary alloys containing 28.2 a/o titanium-6.1 a/o chromium and 10 a/o zirconium-5 a/o iron, and all alloys containing more than 40 a/o zirconium.

Corrosion tests have been started on unalloyed niobium as a part of a cooperative testing program with Bettis and Knolls. All specimens at the three participating laboratories are from the same high-purity-base niobium strip and were prepared for corrosion testing at Bettis. Corrosion tests are being conducted in 680 F degassed water and 750 F 1500-psi steam following prescribed procedures for these particular experiments. Results after a 7-day exposure for six specimens in each condition revealed a dark-gray film on 680 F water specimens and a similar film with some gold tarnish on 750 F steam specimens. Weight gains in 680 F water ranged from 58.7 to 60.8 mg per dm^2, with an average of 59.5 mg per dm^2; in 750 F steam weight gains ranged from 55.0 to 61.7 mg per dm^2 and averaged 57.4 mg per dm^2.

All corrosion tests are being continued except those on commercial-base alloys exposed 196 days in 680 F water. These specimens have been removed from testing and are being examined for evidence of oxygen contamination and hydrogen absorption, and the nature of the oxide film.

Four 50-g alloy ingots of the compositions listed below were hot rolled to 60-mil-thick strip in stainless steel packs at 1800 F.

Alloy	Nominal Alloy Content (Balance Niobium), a/o	Quality of Rolled Strip
N46	5 iron	Excellent
N47	10 iron	Light edge cracking
N52	2.5 vanadium-2.5 nickel	Excellent
N55	2.5 vanadium-2.5 zirconium	Some surface roughness

As indicated, strip of good to excellent quality was obtained from each of the alloys. Samples of each strip are being prepared for hot-hardness measurements and for corrosion testing in 680 F water.

Room-temperature tensile tests were conducted on the three alloys listed on page 12, after vacuum annealing for 1 hr at 1500 C (2730 F), with the results shown. As is apparent from these data, each of the three alloys is quite brittle.

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Alloy	Alloy Content (Balance Niobium), a/o	Tensile Properties	
		Ultimate Strength, psi	Elongation, per cent
NL8	9.9 zirconium-9.4 vanadium	91,000	0
NL9	5.7 zirconium-11.4 vanadium	46,000	0
NL10	9.1 titanium-6.3 chromium	73,000	0

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

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The wide use of Zircaloy-2, a zirconium-base alloy, at elevated temperatures in reactors as cladding for fuel elements and as a reactor structural material has indicated the need for data on the effect of irradiation on the creep properties during irradiation. A considerable amount of data has been obtained on the irradiation-induced changes in the mechanical properties of Zircaloy-2 as determined after irradiation, but none during. Although it has been shown by postirradiation tests that irradiation increases the 0.2 per cent offset yield strength and decreases the ductility, arguments can be advanced to show that irradiation might be expected to soften Zircaloy-2. This is especially true in the temperature range where Zircaloy-2 has been shown to exhibit strain aging. If irradiation eliminates or reduces strain aging, the creep strength will be altered.

Preparatory to the detailed study of the creep behavior of Zircaloy under irradiation, it is planned to perform relatively simple in-pile creep studies directed toward determining whether the creep in pile and creep out of pile differ markedly from each other. This comparison between creep behavior of a specimen in a reactor and one out of a reactor will be based on the differences of measurements taken before and after irradiation. All test conditions are to be the same for specimens in and out of the reactor except for the exposure to neutrons and gamma flux in the reactor.

It is planned to use a tubular test specimen fabricated either from tubing or thick plate. It is further planned to use a specimen which will be composed of several gage sections, each having a different wall thickness. These test lengths will each be separated by a section of tube having a thicker wall. Each test section will be approximately 3-1/3 in. long and have a 2-1/2-in.-long reduced gage section. The maximum outside diameter will be approximately 0.5 in. and the inside diameter will be approximately 0.45 in. The inside diameter will be the same for all sections, while the outside diameter will be different for each section to provide the different wall thicknesses desired. When pressurized internally, a specimen of this design will produce with a single pressure as many different circumferential stresses as there are different wall thicknesses represented.

It is proposed to use internal pressures up to 2000 psi. Since it is desired to produce creep without rupture at the test temperatures, it is estimated that for a temperature of 650 F, wall thicknesses will have to be such that stresses between 15,000 and 21,000 psi will be developed. For the test at 750 F the specimen design will be such that stresses between 6,000 and 12,000 psi will be produced. It appears that the minimum test time should be 500 hr, while the maximum may be 1000 hr or over.

It is thought that the tubular specimens can be fabricated from thick Zircaloy-2 plate with the varied wall thickness produced by machining. The use of specimens prepared from plate should reduce the anomalies usually associated with preferred orientation in Zircaloy-2. A high degree of preferred orientation is quite often apparent in rolled rod, swaged rod, thin sheet, and extruded tubing. It is essential that the specimens be designed so that creep deformation will occur in the reduced gage section without being affected by the thick-wall section at the ends of each gage section. Prior to closure, a solid rod will be inserted inside the tubular specimen. This will serve two purposes: (1) to occupy volume and thereby reduce the total volume of compressed gas, and (2) to provide an anchoring pin for thermocouples.

The test capsule and auxiliary equipment will consist of a stainless steel or aluminum capsule containing the test specimen, auxiliary heaters, the necessary out-of-reactor controls, and a supply of inert gas under pressure. Although a substantial portion of the heat in the specimen will be provided by gamma heating of the specimen itself, resistance heaters for each test section will be required in order to balance the heat and provide constant temperatures.

Test procedure after encapsulation will be to arrange for insertion in a core position in the ETR. It is necessary that the capsule be irradiated in a high fast flux, approximately 1 x 10^{14} nvt fast at a minimum. Prior to reactor startup, the specimens will be brought to test temperatures and stabilized with no internal pressure in the tube. Upon startup of the reactor, test temperatures will be re-established and internal pressure will be applied to the tube. Pressure and temperature will be regulated throughout the test. The specimens will remain at temperature and under pressure during reactor shutdown, and this time will be compensated for in the out-of-pile tests. It is thought better to maintain a constant test temperature since some slight accelerated creep has been noted in cyclic-temperature tests (room temperature to 650 F). A possibility does exist that the damage occurring when the reactor is at power will anneal out at test temperature when the reactor is at zero power. This will be considered carefully, and the exact test conditions will be chosen in the future. As mentioned earlier, it is planned to have the specimen in test for 500 to 1000 hr. Upon removal from the reactor, the capsule will be returned to the Battelle Hot-Cell Facility for measurement of the specimens. Metallographic samples will be taken.

It is realized that the flux in the reactor test hole will vary over the test-specimen length, and it is proposed to determine the flux in the various test sections. Therefore, one of the key parts of this test is the provision of adequate dosimeters. It is proposed to use at least one dosimeter for each test section. Based on experience to date, it is probable that nickel will be used for this purpose. In the case of the 500-hr tests, the dosimeters will be located inside the test capsule and will be removed when the test specimens are removed. For the longer tests, the dosimeters will be located outside the capsule. These dosimeters could be removed and replaced during the test run, if necessary.

It is planned also to develop techniques to study strain aging in Zircaloy-2. It is thought that light and electron microscopy, together with X-ray diffraction and internal-friction studies, may be helpful in studying the strain-aging mechanism. Deformation mechanisms and dislocation movements will be studied by transmission electron microscopy in a thin section of the metal itself. Before this can be done, techniques to make thin films of Zircaloy-2 will have to be developed. Thin films also can be used in the X-ray diffraction studies.

Capsule- and equipment-design studies have been initiated.

STUDIES OF ALLOY FUELS

R. F. Dickerson

Property data for binary alloys of niobium and uranium containing from 10 to 50 w/o uranium continue to be accumulated. All alloys maintained an adherent oxide after 500 hr in 572 F air and in 600 F CO_2. A brown spalling oxide is formed at 662 and 752 F. After 84 days in 600 F water, it is interesting to note that there is very little change in corrosion rate due to differences in uranium content. The alloys exhibit a weight gain of about 1 mg per cm^2, which compares to about 0.2 mg per cm^2 for Zircaloy-2. In 680 F water tests, after 84 days there is a trend in certain alloys (30 w/o uranium and 50 w/o uranium) that indicates the low-oxygen-content material is less corrosion resistant.

Attempts to improve the corrosion resistance of thorium-uranium alloys with zirconium, zirconium and niobium, yttrium and cerium additions have been disappointing. An increase in the corrosion resistance by a factor of two was the best obtained. In addition to continued metallurgical studies on thorium alloys, work with thorium and thorium-uranium carbides has been initiated.

Development of Niobium-Uranium Alloys

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

Niobium-rich niobium-uranium alloys are potential high-temperature metallic fuels for nuclear reactors. As a result of a lack of information concerning their properties they have received very little application as fuels. Previous work with the uranium-rich uranium-niobium alloys has indicated that impurities in the niobium can affect the properties of the alloys. Consequently, the effect of impurities on the physical and mechanical properties of niobium-rich niobium-uranium alloys and upon the compatibility of the alloys with various environments is 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.

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Niobium-10 and -20 w/o uranium alloys have been successfully fabricated by forging and rolling into slab material. Fabrication of alloys containing more than 20 w/o uranium has been unsuccessful. Ingots of the niobium-30 and -40 w/o uranium alloys are being prepared for possible extrusion. Similar alloys have been successfully extruded. If sound material is produced by extrusion of these alloys, attempts will be made to extrude niobium-50 and -60 w/o uranium alloys.

Corrosion testing in air at 572, 662, and 752 F has been completed, and the results show that the niobium-uranium alloys containing up to 60 w/o uranium are resistant to oxidation for 500 hr at 572 F and form a protective adherent oxide. A spalling oxide is formed at 662 and 752 F. In CO_2 the results of tests at 600 F were similar to those in air at 572 F (all alloys formed an adherent black oxide).

The results of corrosion studies in water are shown in Table F-3 for 70 and 84 days on test at 600 and 680 F. All alloys have been removed from tests in 750 F steam after 42 days due to excessive weight losses.

Compatibility studies of niobium-uranium alloys with NaK are being continued. No additional results have been obtained.

High-temperature tensile tests indicate that the niobium-20 w/o uranium alloy should perform well in certain high-temperature applications. The alloys exhibit extremely little decrease in strength with increasing temperatures to 1600 F; tensile data at room temperature and 1600 F have been reported earlier in the program. No apparent effect on the properties due to impurity content was noted during these tests.

It is planned to determine the creep properties of fabricable niobium-uranium alloys at 1600, 1900, and 2400 F in vacuum. These tests will be 1000 hr in duration. The stress necessary to produce rupture in 100 hr will also be determined. It is also planned to investigate the 1000-hr creep life in argon and NaK at 1600 F.

Specimens for thermal-conductivity, thermal-expansion, and electrical-resistivity evaluations have been machined and will be electroplated prior to testing.

Future work will also include a study of the effect of impurities on the gamma-immiscibility gap in the niobium-uranium system.

Development of Thorium-Uranium Alloys

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

A program to develop thorium-uranium-base alloys of improved radiation stability by ternary and quaternary alloying is in progress. In addition, the effect of process variables, including starting materials and casting and fabrication techniques, on distribution of the uranium phase is being investigated. The ternary additions being investigated are niobium, molybdenum, and zirconium, and niobium and zirconium are being investigated as quaternary additions. Carbide and nitride combinations of thorium are also being studied with principal effort at present being devoted toward methods of preparation.

TABLE F-3. CORROSION DATA FOR NIOBIUM-URANIUM ALLOYS IN 600 AND 680 F WATER AFTER 70 AND 84 DAYS[a]

Alloy Content (Balance Niobium), w/o	Impurity Content		Specimen Condition	Total Weight Change, mg per cm^2			
				In 600 F Water		In 680 F Water	
	Oxygen, ppm	Zirconium, w/o		After 70 Days	After 84 Days	After 70 Days	After 84 Days
10 uranium	600	0.74	Fabricated	0.55	0.62	0.65	0.72
	700	0.17	Ditto	0.95	1.05	-9.2	-10.1
	300	0.02	Ditto	1.08	1.10	-35.7	-37.7
20 uranium	600	0.74	Ditto	0.89	1.06	1.06	1.18
	700	0.17	Ditto	0.97	1.08	1.67	1.44
	300	0.02	Ditto	0.66	0.76	1.03	1.09
30 uranium	600	0.74	As cast	0.84	0.94	-0.71	-0.76
	700	0.17	Fabricated	0.79	0.76	-0.91	-0.89
	300	0.02	As cast	1.36	1.43	-2.34	-2.46
40 uranium	600	0.74	Ditto	0.63	0.78	1.53	1.72
	700	0.17	Ditto	0.83	1.01	0.77	0.78
	300	0.02	Ditto	0.84	1.04	1.47	1.70
50 uranium	600	0.74	Ditto	0.85	0.96	1.44	0.0
	700	0.17	Ditto	0.86	0.80	0.42	0.42
	300	0.02	Ditto	0.63	0.95	-2.90	-3.96
60 uranium	600	0.74	Ditto	-0.31	-0.48	-69.5	(b)
	700	0.17	Ditto	0.71	0.50	-8.1	-9.55
	300	0.02	Ditto	0.56	0.58	-5.1	-6.20
100 niobium	131	0.05	Fabricated	--	0.53	--	0.97
	500	0.03	Ditto	--	0.66	--	(c)
Zircaloy-2	--	--	Ditto	0.18	0.20	0.24	0.30

(a) Average of two specimens.
(b) Both specimens taken off test.
(c) Completely oxidized after 56 days of exposure.

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Attempts to improve the corrosion resistance of thorium with cerium, niobium and zirconium, yttrium, and zirconium additions have been disappointing. An increase in corrosion resistance by a factor of two was the best obtained. A few alloys in which the zirconium-uranium ratio is further increased will be prepared and tested. However, results to date do not hold much promise for alloying as a means of obtaining improved corrosion resistance.

Studies of the remaining ternary and quaternary alloys will involve investigations of the effect of casting variables and the effect of hot and cold work and heat treatment on the microstructure of these alloys. The results will be compared with those already obtained on binary thorium-uranium alloys. The effect of alloying on recrystallization will also be investigated. Alloy material, in addition to that already on hand, is being prepared for these studies.

Buttons of thorium and thorium-uranium carbides have been arc melted and cast. The buttons are found to decompose rapidly on exposure to the atmosphere, with the decomposition rate decreasing with increasing uranium content. Metallographic examination of a thorium carbide-30 w/o uranium carbide revealed a single-phase structure consisting primarily of equiaxed grains, although some areas contained large elongated grains.

FISSION-GAS RELEASE FROM REFRACTORY FUELS

J. B. Melehan, W. G. Rieder, R. H. Barnes, and F. A. Rough

The materials and equipment preparations for study of fission-gas-release behavior of UO_2 are of continuing primary interest in this program. These include the fabrication and preirradiation inspection of sintered and fused UO_2, and the design of equipment for carrying out irradiations at the Battelle Research Reactor and also for the collection and analysis of released fission gases. Construction of both the heating furnace for the in-pile beam-tube specimens and the gas-trapping and -monitoring components is now in the advanced stages.

Preparation of Fused and Sintered UO_2 Irradiation Specimens

Only limited experimental efforts were made in the preparation and study of fused UO_2 for use in postirradiation heat treatment for determination of fission-gas release rates. Physical measurements of plate and spherical specimens, determinations of weight, dimensions, surface area, and crystallographic orientation, and electron- and optical-microscopy studies are continuing.

Decontamination heat treatments for the removal of UN_2 second phase are still in progress. Only partial success has been experienced with techniques employed thus far.

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Preparation of sintered UO_2 irradiation specimens for use in postirradiation and in-pile gas-release studies is under way, although no preirradiation characterization studies have begun.

Design and Construction of Beam-Tube Furnace

Shop drawings of the in-pile equipment were completed and fabrication was started. Fabrication is approximately 70 per cent complete. Purchase orders have been placed for all major components of the in-pile equipment. A request for approval to purchase a 5-kw induction heater from the low bidder was submitted for AEC approval. Review of problems associated with load-coil cooling-water activation led to the decision to purchase a recirculator-exchanger. A review of quotations on temperature-recording and control equipment and of the experiment operating criteria led to the decision to purchase certain components. A perspective layout was made of the complete fission-gas-release experiment installed in the BRR.

Preparation for In-Pile Study

Construction of the gas-sampling system for the in-pile studies is continuing. Assembly of the apparatus to study the release of fission gas from fuel specimens in the radioisotope laboratory during postirradiation heat treatment is continuing. Assembly of both systems is expected to be completed in September.

GENERAL FUEL-ELEMENT DEVELOPMENT

S. J. Paprocki

Investigations are being conducted to develop cermet fuel materials containing from 60 to 90 volume per cent of fuel dispersed in metal matrices of chromium, molybdenum, niobium, and stainless steel. These include development of fabrication techniques and determination of physical and mechanical properties. Results of these studies indicate that the cermets are extremely promising fuel materials. The uranium content is relatively high, and they possess a thermal stability and thermal conductivity that are far superior to ceramic fuels. They also offer structural support and the metal phase can be metallurgically bonded to compatible cladding materials.

Techniques have been developed for the solid-phase bonding of niobium by use of the gas-pressure-bonding process. One method utilizes zirconium at the interface to promote diffusion. Bonding is achieved at 1650 F with a helium gas pressure of 10,000 psi. Direct self-bonding of niobium is also achieved by treatment at 2100 F with a 10,000-psi gas pressure. The bonds possess complete grain growth across the interface and an excellent ductility.

The mechanism involved in the solid-state bonding of similar metals is being investigated. The studies are fundamental and designed to relate theory to actual results obtained through the use of the gas-pressure-bonding process.

Fabrication of Cermet Fuel Elements

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

Cermet fuel materials consisting of from 60 to 90 volume per cent of ceramic fuel dispersed in chromium, molybdenum, niobium, or stainless steel matrices are being investigated. Methods investigated for fabricating these materials to 90 per cent of theoretical density or better are hot press forging, hot swaging, and gas-pressure bonding. Major emphasis is now being placed on the gas-pressure-bonding technique.

Rods of 80 volume per cent UO_2-molybdenum and -chromium have been fabricated by pressure bonding green-pressed cermets for 3 hr at 2300 F under a helium gas pressure of 10,000 psi. The rods were prepared by cold pressing 80 volume per cent UO_2-metal powder mixtures in a 1-in.-diameter die at 40 tsi to densities of 70 to 73 and 72.3 to 72.6 per cent of theoretical, respectively, for the chromium- and molybdenum-matrix cermets. The green-pressed cermets were then fitted in a stainless tube to which stainless end plugs were welded subsequent to evacuation and pressure bonding. Densities of 93.6 and 86.7 per cent of theoretical were measured, respectively, on the chromium- and molybdenum-matrix cermets after pressure bonding. The UO_2-molybdenum cermet has had its stainless steel sheath machined off and has been fitted into another stainless steel tube for additional pressure bonding at 2500 F. Segregation was visibly evident in the UO_2-chromium cermets; therefore, additional specimens are being prepared for pressure-bonding studies.

Thermal-conductivity measurements on cermets prepared by pressure bonding are being continued. In addition, 80 volume per cent UO_2-molybdenum and -niobium powder cermets in the form of flat plates are being prepared for pressure-bonding studies.

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

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

Molybdenum and niobium retain their strength at elevated temperatures and possess favorable nuclear properties. These qualities make them potential cladding and structural materials for high-temperature reactor applications. The gas-pressure-bonding technique is being studied as a possible method for cladding ceramic and cermet-type fuel with these metals.

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Evaluation of a niobium specimen prepared by edge brazing with zirconium and pressure bonding at 2100 F and 10,000 psi for 3 hr has revealed excellent bonding. The surfaces of this specimen were prepared for bonding by pickling in a solution of 65 parts HNO_3 and 35 parts HF. Bend tests made on this specimen showed the ductility of the niobium to be equal to that of as-received material; tests made on sections taken from the brazed areas of the specimen also revealed excellent ductility. This technique provides a method for edge welding niobium plates without the use of fusion welding which often severely embrittles niobium.

Various heat treatments have been investigated to improve the ductility of some as-bonded specimens which were embrittled during the bonding cycle due to hydrogen pickup. It has been found that embrittled specimens treated in vacuum at 2000 F for 2 hr possess ductility equal to that of as-received material.

The most satisfactory surface preparation for molybdenum plates has been found to be a pickle in concentrated nitric acid under a layer of kerosene. The role of kerosene in this operation is not clearly understood; however, it has been postulated that it acts only as a film between the nitric acid and the air and takes no active part in the reaction. In an effort to determine if this postulate was true a specimen was prepared by pickling in nitric acid under cyclohexane. The cyclohexane should be completely inert with nitric acid but would not present a similar protective film. This specimen was pressure bonded at 2300 F and 10,000 psi for 3 hr. Examination of this specimen revealed that poor bonding had occurred, indicating that apparently kerosene does take some active part in this reaction. Additional specimens will be prepared to confirm these observations.

In an effort to improve the as-bonded ductility of molybdenum several specimens have been subjected to bonding cycles of lower temperatures and shorter times. The bonds obtained at the lower conditions were not as good as those obtained at the higher bonding parameters, and no improvement in the ductility of the as-bonded molybdenum was observed. Solution heat treatments at 2000 F for 2 hr followed by a water quench have also failed to improve ductility of molybdenum-clad specimens. Efforts are being made to develop an edge-welding technique that will permit the pressure bonding of molybdenum without a protective envelope which introduces contamination.

Factors Affecting Pressure Bonding

G. W. Cunningham and J. W. Spretnak

An investigation is being made of the phenomena related to pressure bonding of similar metals. Work has continued with self-bonded specimens of copper or zirconium in an effort to determine the nature of the interfacial bond produced when the metals are placed in intimate contact. The term "intimate contact" as referred to in this study means that a bond line is detectable but no voids or impurities are visible at magnifications up to 1000X. Also, at this stage there is little or no grain growth across the interface. Tests to date suggest that, although the interface resembles an ordinary grain boundary in appearance, it probably contains a much higher concentration of vacancies or areas of misfit. High-temperature diffusion annealing, low-temperature annealing under pressure, and diffusion annealing of silver into the interfacial boundary are being used to study the bond.

For the high-temperature annealing studies, a copper specimen was bonded by subjecting it to a pressure of 10,000 psi at 1100 F for 10 min. Specimens were examined in the as-bonded condition, after a 2-hr anneal at 1900 F, a 26-hr anneal at 1900 F, and a 2-hr anneal at 1900 F subsequent to a 50 per cent reduction in thickness by cold pressing. As bonded the interfacial boundary appeared to be similar to a grain boundary and no void areas were detected. After 2 hr at 1900 F small voids could be seen in the boundary, but no grain growth across the interface had occurred. After 26 hr at 1900 F the voids appeared to be somewhat larger and in some cases spaced farther apart; grain growth across the interface had occurred over less than 20 per cent of the section examined. Grain growth over more than 40 per cent of the interfacial boundary was visible in the case of the specimen cold reduced before annealing. These results indicate that restriction of grain growth across the interface is not due to the presence of impurities in these specimens, and they also suggest that vacancies must either diffuse away from the boundary, collect into widely spaced voids in the boundary, or be swept away by grain boundaries. In the case of grain boundaries, it appears that wide-angle boundaries such as those produced during recrystallization and grain growth are necessary to serve as vacancy sinks. Specimens are being prepared for additional annealing studies.

A series of copper specimens is being prepared for annealing under pressure at temperatures of 1200 F and pressures to 24,000 psi. When modification of the hot-press unit is completed, pressures up to 3000 psi at 1900 F will be used. It has been shown in previously reported work that intimate contact and adhesion of the bonding surfaces are obtained within a very short period of time if the pressure is sufficiently high to cause plastic flow. However, very little diffusion occurs and there is little or no grain growth across the interface. Under these conditions, it would not be expected that a large increase in pressure over that required for intimate contact would be especially beneficial except that it might reduce the total number of vacancies at the interface, but a very large pressure might be required for complete obliteration of the interface. As would be predicted, comparison of specimens hot pressed 10 min at 1100 F at pressures from 10,000 to 24,000 psi does not show any significant difference. Comparison of annealed specimens will be necessary to determine whether differences exist in the number of vacancies at the interface.

On the other hand, an increase in pressure would be expected to be beneficial during a diffusion anneal. According to a relationship worked out by Barnes at Harwell on the basis of the Gibbs-Thomson equation, increasing the pressure effectively increases the vacancy saturation limit of the metal. However, since diffusion is time dependent, this relationship can be tested only during an anneal of several hours. A series of specimens now being prepared will be annealed at times to 4 hr at a pressure of 10,000 psi and a temperature of 1150 F.

Diffusion of silver into the interfacial boundary has also provided indirect evidence that a higher concentration of vacancies exists at the interface. A specimen pressure bonded at 1100 F and 10,000 psi for 10 min has been used to form a diffusion couple with a copper-4 w/o silver alloy. After annealing 200 hr at 1245 F and cooling 1 F per min in order to precipitate the silver, the couple was sectioned and the diffusion of silver along the interfacial boundary was found to be greater than along any grain boundary. Detailed metallography is in progress, and it may be possible to give a quantitative discussion of the results.

An interesting experiment with zirconium also suggests the presence of vacancies at the interface. Previous work at Battelle has shown that improved interfacial bonds on Zircaloy-2 are obtained when the component is heated into the beta region for 5 min, cooled to the alpha region, annealed, and cooled to room temperature. In connection with a different problem, work at The Ohio State University has indicated that when iron is cycled above and below the alpha-gamma transformation large internal stresses are developed. This work suggests that vacancies can be removed from the interface and grain growth across the interface promoted by both internal pressure and the use of grain boundaries as vacancy sinks when a metal is cycled around the phase-transformation temperature. The value of such a treatment and the importance of studying this phenomenon has been demonstrated by an experiment in which crystal-bar zirconium was pressure bonded at 1250 F for 10 min at a pressure of 10,000 psi and subsequently cycled five times from 600 to 900 C and annealed 30 min at 550 C. As pressure bonded, the interface showed intimate contact but no grain growth across the interface. After cycling and annealing, some small voids or possibly hydrides or oxides (which are visible in other areas of the specimen) could be seen at the bond line, but there was 100 per cent grain growth across the interface. Additional work is planned with iron and zirconium.

FF. FUEL-CYCLE PROGRAM STUDIES

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

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

The prime objectives of this program are to utilize the gas-pressure-bonding technique to reduce fuel-element manufacturing costs, and, at the same time, to improve the quality of the fabricated elements. The study is directed toward the refinement and further development of the gas-pressure-bonding process for the simultaneous densification and cladding with stainless steel of ceramic, cermet, and dispersion fuels. The initial emphasis is being placed on pure UO_2 ceramic cores.

In order to reduce fabrication costs of UO_2 ceramic fuel elements, a high-temperature sintering operation is not being considered in this program. Instead, initial studies are concerned with the cold compacting of high-density UO_2 bodies which are subsequently pressure bonded to near theoretical density, if desired. In order to prevent excessive deformation of the stainless steel cladding during bonding, a minimum UO_2 cold-pressed density of 70 per cent of theoretical has been selected.

During the past month, cold-compacting studies were made with several grades of commercial UO_2 powder. These include Mallinckrodt's ceramic grade, dense ceramic grade, high-fired grade, and a special dense grade as well as a high-fired grade prepared by Nuclear Equipment and Materials Company. Other types of UO_2 powder on order are Mallinckrodt's spherical grade and a fused grade from Spencer Chemical Company.

The compacting characteristics of these powders will be evaluated on the basis of their particle shape, size, and distribution, their porosity, and their method of preparation.

In Table FF-1, the results of the cold-compacting studies are listed. The compacting pressure was varied between 20 and 55 tsi. Below 20 tsi, the green strength of the pellets was not adequate to permit either rapid ejection from the die or subsequent handling during assembly. Above 55 tsi, it is thought that the process would become uneconomical because of excessive die wear and breakage.

In Figure FF-1, the data obtained with the Ceremul "C" binder is plotted as a function of density and compacting pressure. The superiority of several of the UO_2 grades from the standpoint of densification during compacting is quite apparent.

Further studies along this line will include an investigation of other binders and grades of UO_2.

FF-2

TABLE FF-1. GREEN DENSITIES OF UO_2 COMPACTS

Specimen	Binder	Compacting Pressure, tsi	Height-to-Diameter Ratio	Density, g per cm^3	Density, per cent of Theoretical
NUMEC Minus 325-Mesh High-Fired UO_2					
U-1044	Camphor-alcohol slurry	20	0.5	7.40	67.5
U-1045	Camphor-alcohol slurry	25	0.5	7.86	71.8
U-1046	Camphor-alcohol slurry	30	0.5	7.84	71.5
U-1047	Camphor-alcohol slurry	35	0.5	7.96	72.9
U-1048	Camphor-alcohol slurry	40	0.5	8.14	74.3
U-1049	Camphor-alcohol slurry	50	0.5	8.06	73.6
U-1052	Ceremul "C"	20	0.5	7.71	70.4
U-1053	Ceremul "C"	25	0.5	7.80	71.3
U-1054	Ceremul "C"	30	0.5	7.90	72.1
U-1055	Ceremul "C"	35	0.5	8.07	73.7
U-1056	Ceremul "C"	40	0.5	8.05	73.5
U-1057	Ceremul "C"	50	0.5	8.12	74.1
U-1058	Ceremul "C"	20	1	7.72	70.5
U-1059	Ceremul "C"	25	1	7.85	71.7
U-1060	Ceremul "C"	30	1	8.00	73.1
U-1061	Ceremul "C"	35	1	8.07	73.7
U-1062	Ceremul "C"	40	1	8.15	74.4
U-1063	Ceremul "C"	50	1	8.22	75.0
MCW Depleted As-Received Special Dense Grade UO_2					
U-1094	Ceremul "C"	20	1	8.33	76.0
U-1093	Ceremul "C"	25	1	8.52	77.8
U-1092	Ceremul "C"	30	1	8.60	78.5
U-1089	Ceremul "C"	35	1	8.64	78.8
U-1091	Ceremul "C"	40	1	8.75	80.0
U-1090	Ceremul "C"	50	1	9.34	85.3
U-1095	Camphor-alcohol slurry	20	1	7.78	71.0
U-1096	Camphor-alcohol slurry	25	1	8.29	75.6
U-1097	Camphor-alcohol slurry	30	1	8.35	76.3
U-1098	Camphor-alcohol slurry	35	1	8.48	77.4
U-1099	Camphor-alcohol slurry	40	1	8.53	77.9
U-1100	Camphor-alcohol slurry	50	1	8.67	79.1
U-1101	Camphor-alcohol slurry	55	1	8.75	80.0
MCW Depleted As-Received Dense Ceramic Grade UO_2					
U-1102	Camphor-alcohol slurry	20	1	6.17	56.3
U-1103	Camphor-alcohol slurry	25	1	6.31	57.6
U-1104	Camphor-alcohol slurry	30	1	6.38	58.2
U-1105	Camphor-alcohol slurry	35	1	6.49	59.2
U-1106	Camphor-alcohol slurry	40	1	6.59	60.2
MCW Depleted Minus 400-Mesh High-Fired UO_2					
U-1107	Camphor-alcohol slurry	20	1	7.54	68.8
U-1108	Camphor-alcohol slurry	25	1	8.04	73.3
U-1109	Camphor-alcohol slurry	30	1	7.89	72.0
U-1110	Camphor-alcohol slurry	35	1	8.05	73.5
U-1111	Camphor-alcohol slurry	40	1	8.11	74.0
U-1112	Camphor-alcohol slurry	50	1	8.29	75.6
U-1113	Camphor-alcohol slurry	55	1	8.34	76.1

TABLE FF-1. (Continued)

Specimen	Binder	Compacting Pressure, tsi	Height-to-Diameter Ratio	Density, g per cm^3	Density, per cent of Theoretical
		MCW Depleted As-Received Special Dense Grade UO_2			
U-1114	Ceremul "C"	20	1	8.33	76.0
U-1115	Ceremul "C"	25	1	8.44	77.0
U-1116	Ceremul "C"	30	1	8.54	78.0
U-1117	Ceremul "C"	35	1	8.70	79.4
U-1118	Ceremul "C"	40	1	8.75	79.9
U-1119	Ceremul "C"	50	1	9.36	85.5
U-1120	Ceremul "C"	55	1	9.47	86.4
		MCW Depleted As-Received Dense Ceramic Grade UO_2			
U-1121	Ceremul "C"	20	1	6.08	55.5
U-1122	Ceremul "C"	25	1	6.24	56.9
U-1123	Ceremul "C"	30	1	6.33	57.8
U-1124	Ceremul "C"	35	1	6.38	59.3
		MCW Minus 400-Mesh High-Fired UO_2			
U-1127	Ceremul "C"	25	1	7.93	72.4
U-1128	Ceremul "C"	30	1	8.04	73.4
U-1129	Ceremul "C"	35	1	8.14	74.2
U-1130	Ceremul "C"	40	1	8.15	74.4
U-1131	Ceremul "C"	50	1	8.34	76.1

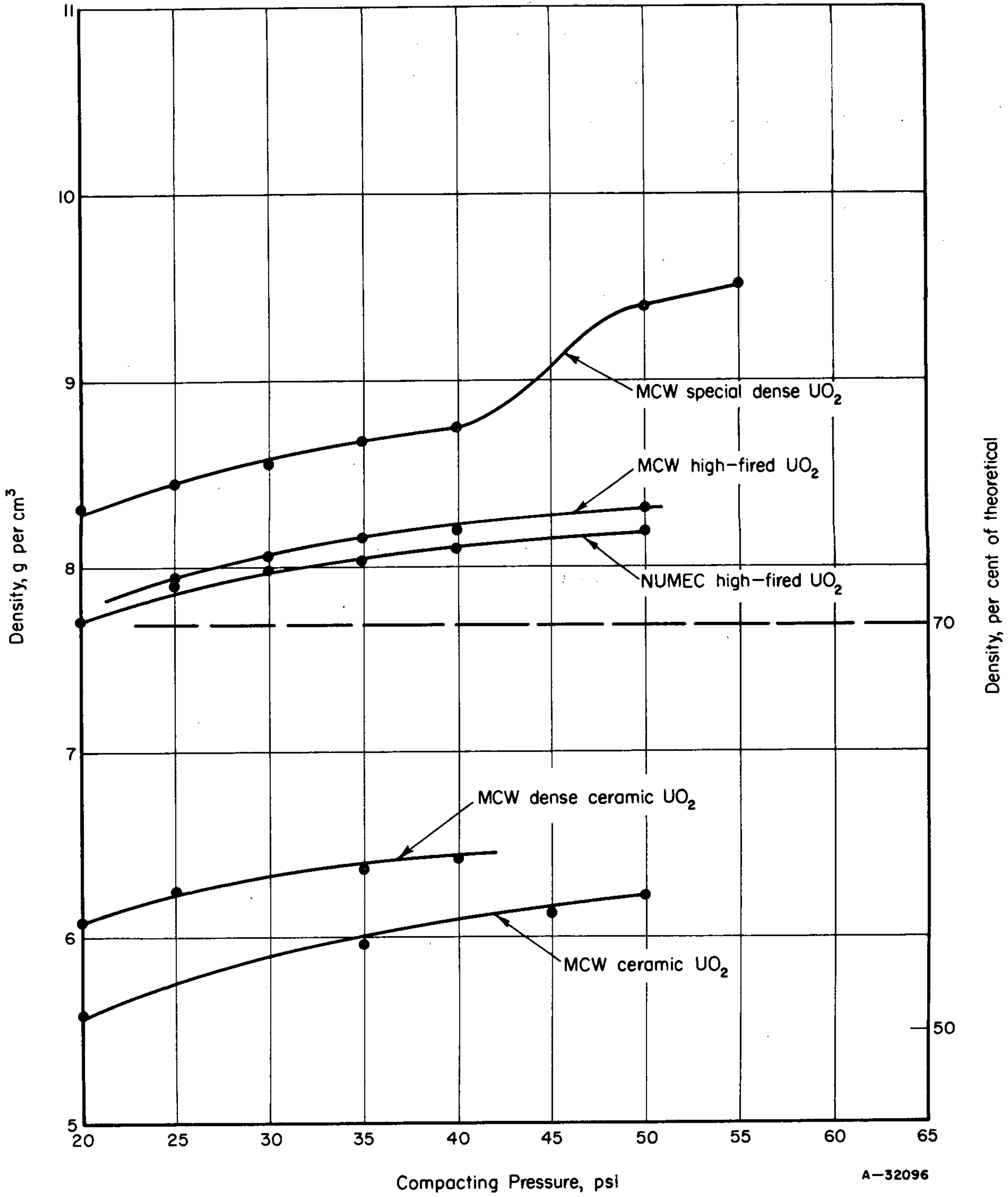

FIGURE FF-1. EFFECT OF COMPACTING PRESSURE ON THE GREEN DENSITY OF VARIOUS TYPES OF UO_2 POWDERS

Ceremel "C" binder was employed in all specimens.

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The tap density of the various UO_2 powders was measured and is reported in Table FF-2.

TABLE FF-2. TAP DENSITY OF SEVERAL COMMERCIAL UO_2 POWDERS

UO_2 Type	Density G per Cm^3	Density Per Cent of Theoretical
MCW ceramic grade	1.75	16.0
MCW dense ceramic grade	3.74	34.1
MCW high-fired grade	5.93	54.1
MCW special dense grade	6.94	63.3

From these data it is apparent that the objective of 70 per cent of theoretical density prior to pressure bonding might be accomplished by charging the loose UO_2 powder directly, thereby eliminating the need for cold compacting with binders and subsequent degassing. A single tube was filled with special dense UO_2 by successive hand pressing and tapping. The UO_2 density was approximately 77 per cent of theoretical. This specimen is being pressure bonded to determine the extent of further densification. Further studies in this direction will include an investigation of various methods of vibrating the loosely packed UO_2 to promote densification.

A cold-pressed UO_2 flat-plate compact approximately 0.250 by 1.0 by 1.0 in., which was placed into a flat-plate swastika-type container and pressure bonded at 2100 F for 2 hr at 10,000 psi, has been evaluated. The compact was utilized to check the design of the pressure-bonding container to be used in the initial study of fabrication of stainless steel-clad UO_2 flat-plate compacts, as well as methods of loading and degassing the compacts. The pressure-bonded specimen was evenly and well deformed, indicating densification and cladding had been achieved. The compact had an original green density of 61.13 per cent of theoretical (6.70 g per cm^3) and was densified during pressure bonding to a density of 98.9 per cent of theoretical (10.83 g per cm^3).

Eight rod-type specimens containing a total of 40 compacts with initial green densities as high as 86 per cent of theoretical have been pressure bonded. The specifications for these compacts are presented in Table FF-3. The cores measure 0.540 in. in OD, have a cladding thickness of 0.020 in., and are of various lengths. These specimens are being evaluated to determine the final densification of each of these compacts.

The investigation into the effect of the bonding parameters of time, temperature, and pressure on the bonds obtained with Type 304 stainless steel is being continued.

TABLE FF-3. PRESSURE-BONDING STUDIES

Green Compacts for Pressure Bonding								Pressure-Bonding Conditions			
Specimen	Binder	Compacting, Pressure, tsi	Height, in.	Diameter, in.	Approximate Height-to-Diameter Ratio	Density, g per cm^3	Density, per cent of theoretical	Tube	Time, hr	Temperature, F	Pressure, psi
					Usable Scrap High-Fired UO_2						
U-1005	Camphor/alcohol	35	0.753	0.540	1.5	7.44	67.8	1	2	2100	10,000
U-1006	Camphor/alcohol	40	0.750	0.540	1.5	7.40	67.5	1	2	2100	10,000
					MCW Ceramic Grade UO_2						
U-1035	Camphor/alcohol	20	0.532	0.540	1	5.86	53.3	1	2	2100	Ditto
U-1028	Ceremul "C"	35	0.516	0.539	1	5.97	54.5	1	2	2100	"
U-1029	Ceremul "C"	45	0.517	0.539	1	6.12	55.9	1	2	2100	"
					NUMEC High-Fired UO_2						
U-1058	Ceremul "C"	20	0.432	0.538	1	7.72	70.5	2	2	2100	"
U-1060	Ceremul "C"	30	0.467	0.538	1	8.00	73.1	2	2	2100	"
U-1062	Ceremul "C"	40	0.526	0.538	1	8.15	74.4	2	2	2100	"
U-1063	Ceremul "C"	50	0.489	0.539	1	8.22	75.0	2	2	2100	"
U-1064	Camphor-alcohol slurry	35	0.445	0.539	1	7.83	71.5	3	1	2100	"
U-1065	Camphor-alcohol slurry	35	0.503	0.539	1	7.89	72.0	3	1	2100	"
U-1069	Camphor-alcohol slurry	35	0.510	0.539	1	7.78	71.0	3	1	2100	"
U-1074	Ceremul "C"	35	0.510	0.539	1	7.79	71.1	3	1	2100	"
U-1076	Ceremul "C"	35	0.510	0.539	1	7.80	71.1	3	1	2100	"
U-1078	Ceremul "C"	35	0.505	0.539	1	7.78	71.1	3	1	2100	"
U-1081	Ceremul "C"	35	0.507	0.539	1	7.80	71.2	4	2	2200	"
U-1082	Ceremul "C"	35	0.505	0.539	1	7.85	71.7	4	2	2200	"
U-1083	Ceremul "C"	35	0.506	0.539	1	7.81	71.2	4	2	2200	"
U-1084	Ceremul "C"	35	0.507	0.539	1	7.80	71.2	4	2	2200	"
U-1085	Ceremul "C"	35	0.504	0.539	1	7.90	72.1	4	2	2200	"
U-1086	Ceremul "C"	35	0.500	0.539	1	7.91	72.2	4	2	2200	"
U-1068	Camphor-alcohol slurry	35	0.513	0.539	1	7.78	71.0	5	2	2150	"
U-1072	Camphor-alcohol slurry	35	0.516	0.539	1	7.71	70.4	5	2	2150	"
U-1073	Camphor-alcohol slurry	35	0.514	0.539	1	7.75	70.7	5	2	2150	"
U-1075	Ceremul "C"	35	0.513	0.539	1	7.76	70.8	5	2	2150	"
U-1077	Ceremul "C"	35	0.513	0.539	1	7.77	71.0	5	2	2150	"
U-1079	Ceremul "C"	35	0.513	0.539	1	7.70	70.4	5	2	2150	"
U-1087	Ceremul "C"	35	0.500	0.539	1	7.94	72.5	6	2	2150	"

TABLE FF-3. (Continued)

Green Compacts for Pressure Bonding								Pressure-Bonding Conditions			
Specimen	Binder	Compacting, Pressure, tsi	Height, in.	Diameter, in.	Approximate Height-to-Diameter Ratio	Density, g per cm^3	Density, per cent of theoretical	Tube	Time, hr	Temperature, F	Pressure, psi
					MCW Special Dense UO$_2$						
U-1120	Ceremul "C"	55	0.424	0.539	1	9.47	86.4	7	2	2100	10,000
U-1090	Ceremul "C"	50	0.431	0.539	1	9.34	85.3	7	2	2100	Ditto
U-1091	Ceremul "C"	40	0.470	0.539	1	8.75	80.0	7	2	2100	"
U-1099	Camphor-alcohol slurry	40	0.470	0.539	1	8.53	77.9	7	2	2100	"
U-1100	Camphor-alcohol slurry	50	0.462	0.539	1	8.67	79.1	7	2	2100	"
U-1101	Camphor-alcohol slurry	55	0.458	0.539	1	8.75	80.0	7	2	2100	"
					MCW High-Fired UO$_2$						
U-1129	Ceremul "C"	35	0.493	0.539	1	8.14	74.2	8	2	2100	"
U-1130	Ceremul "C"	40	0.492	0.539	1	8.15	74.4	8	2	2100	"
U-1131	Ceremul "C"	50	0.477	0.539	1	8.34	76.1	8	2	2100	"
U-1111	Camphor-alcohol slurry	40	0.491	0.539	1	8.11	74.0	8	2	2100	"
U-1112	Camphor-alcohol slurry	50	0.481	0.539	1	8.29	75.6	8	2	2100	"
U-1113	Camphor-alcohol slurry	55	0.476	0.540	1	8.34	76.1	8	2	2100	"

FF-8

DEVELOPMENT OF URANIUM CARBIDE-TYPE FUEL MATERIALS

F. A. Rough and W. Chubb

In an attempt to reduce fuel-cycle costs for the production of electrical power from nuclear energy, the AEC has sponsored a series of research programs aimed at the utilization of uranium carbides as reactor fuels. At Battelle, an integrated program of research has been initiated to develop techniques for preparing solid shapes of uranium carbides, to determine their physical and mechanical properties, and to obtain some information on the nature and mechanism of their irradiation damage. In the course of this program, up to the present time, information has been reported on the effects of certain impurities upon the casting behavior, corrosion resistance, and strength of uranium monocarbide. The densities and electrical resistivities of uranium alloys containing from 0 to 9.2 w/o carbon were reported last month in BMI-1357.

During the past month, reactions for the production of powdered or sintered uranium monocarbide from a variety of starting materials have been investigated. Powder containing 3.8 w/o carbon was prepared by reacting uranium metal with methane. Several other reactions appear promising. Studies of the rates of interdiffusion of uranium and carbon in the uranium monocarbide-uranium dicarbide system at temperatures from 1600 to 1980 C have been advanced to the point where it is possible to calculate an activation energy for diffusion of 92,000 cal per mole.

Alternate Fabrication Methods for UC

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

Several methods of producing high-density UC pellets are being investigated. These methods include sintering of UC powders prepared by various techniques as well as reacting uranium with carbon, UC_2 with uranium, and the uranium oxides with carbon at temperatures high enough for densification to occur. The economics of mass production as well as the quality of the UC pellets will be used as basis for evaluation.

Compacts of arc-cast and crushed UC powder of 4.70 and 4.85 w/o carbon were sintered 2 hr at 1850 C. Indications are that the 4.70 w/o carbon UC sintered to a higher density than the 4.85 w/o carbon; however, accurate conclusions could not be made because the pellets were oxidized considerably when removed from the furnace. Additional compacts of the material are being prepared for sintering studies. In addition, a pellet compacted from a stoichiometric mixture of uranium and graphite has been sintered for 4 hr at 1100 C. Considerable degassing was evidenced during heating to temperature. Neither density measurements nor microscopic-examination results are available at this time.

Powder of uranium-3.8 w/o carbon has been prepared by reaction of uranium metal with methane. Compacts of stoichiometric mixtures of this material and carbon are being prepared for sintering studies. In addition, uranium metal is being reacted with methane in an attempt at obtaining stoichiometric UC powder.

An evaluation of the free energy of several possible reactions leading to the formation of UC has been made, and, from this standpoint, the reaction of UC_2 with the uranium metal to produce UC appears promising at temperatures of 1000 K and above. Crushed arc-cast UC_2 and uranium powder are being mixed to investigate this reaction. In addition, mixtures of UC, UC_2, and uranium metal are being prepared for sintering. The bomb reaction of UF_4 or UF_6, calcium, and CaC_2 to UC and CaF_2 appears promising for the formation of UC from the free-energy standpoint. These reactions are being considered for possible investigation.

Melting and Casting Techniques for Uranium-Carbon Alloys

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

Melting and casting procedures for the preparation of simple, cylindrical as-cast shapes of UC in sizes up to 1/2 in. in diameter and 2 in. long have been developed and are capable of consistently producing high-quality homogeneous castings to specifications of plus or minus 0.1 w/o carbon. It is desired to extend these techniques to include the preparation of larger and more varied shapes. The research will include studies of the effects of impurities, alloys, carbon content, starting materials, and molding variations on such characteristics as ease of casting, soundness, surface quality, cast structure, and homogeneity.

Nondestructive methods of evaluation such as 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 of materials will be used to produce UC of different properties. Materials produced by solid-state reaction, gaseous reaction, and direct alloying will be evaluated to determine the effects of production techniques on casting. Alloying additions will be made first with elements that might be expected to contaminate UC during normal processing and second with elements which might be expected to improve the casting properties of UC.

At the present time molds and crucibles are being prepared for melting of UC by skull-melting techniques. A UC skull is being prepared to act as a liner in the skull arc-melt crucible. An induction-heated graphite mold is being prepared for this furnace. The mold-heating device is intended to alleviate the thermal shock that accompanies the casting of UC into massive, cold graphite molds.

FF-10

Metallurgical and Engineering Properties of Uranium Monocarbide

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

This research for the Fuel-Cycle Development Program has to do with the properties of uranium-carbon alloys as influenced by processing and composition variables. Mechanical properties such as modulus, rupture strength, and hardness as well as physical properties such as electrical and thermal conductivity, density, corrosion behavior, and others of specific interest are being and will be studied in an attempt to establish the qualities of these alloys.

Previous work on this program has shown that additions of hydrogen, nitrogen, and oxygen have a considerable effect on the corrosion resistance of UC in water at 60 C. Increased amounts of these additions and silicon, which produced poor castability of UC, were added to charges that were subsequently drop cast into a cylindrical shape (1-3/4 in. long by 3/8 in. in diameter). In evaluating the effects of these impurities on the casting qualities of UC (4.8 to 5.0 w/o carbon) it was found that 200 ppm of silicon was detrimental. No serious casting difficulties were experienced with UO_2 additions up to 1 w/o. UC (4.8 to 5.0 w/o carbon) melted in atmospheres containing hydrogen and nitrogen did not exhibit any great change in casting qualities. However, under conditions involving melt charges containing less than stoichiometric quantities of carbon, UO_2 additions of 10 volume per cent or more, and hydrogen and nitrogen atmospheres caused serious casting difficulties.

Specimens of cast UC have been heat treated at 1500 C for 1 hr and the effects of this heat treatment on mechanical properties are being evaluated. Studies concerned with the compatibility of UC with various materials, and studies of the corrosion rate of UC in NaK at 1500 F are in progress.

Uranium Monocarbide Diffusion Studies

W. Chubb and F. A. Rough

The rates of diffusion of uranium and carbon in uranium carbides are important from the standpoints of understanding sintering phenomena, mechanical properties at high temperature, precipitation phenomena, and diffusion of fission gases in irradiated specimens. To obtain some indication of the magnitude of diffusion rates in these carbides, measurements have been made of the rate of growth of the combined UC and UC_2 layers between a graphite crucible and carbon-saturated uranium metal. Determination of interdiffusion rates by this method is almost complete, and experimental techniques are being developed for determination of self-diffusion rates of uranium in uranium monocarbide.

Measurements of the total thickness of the layers of UC and UC_2 formed between graphite and uranium were successfully completed in three out of four cases of melts held for various lengths of time at 1800 and 1900 C. Two measurements were obtained at 1800 C and the results show good agreement with the assumption that the thickness of the layers varies as the square root of time. The diffusion coefficients calculated from these three melts and from two previous melts made at 1600 and 1980 C are shown in the following tabulation:

Temperature, C	Interdiffusion Coefficient, D, cm^2 per sec
1600	3.6×10^{-8}
1800	2.3×10^{-7}
1800	3.3×10^{-7}
1900	1.6×10^{-6}
1980	2.2×10^{-6}

These results seem to be fairly accurate, since they fall very close to a straight line when log D is plotted versus the reciprocal of temperature. An activation energy for interdiffusion in this duplex, UC plus UC_2, system can be calculated from these data. The result, 92,000 cal per mole, appears to be reasonable for a system such as this one involving solvents having very high melting points. It should be emphasized that these diffusion results are based on the assumption that the phases, UC and UC_2, are sufficiently similar to be considered to be one single solvent.

Techniques for determining rates of self-diffusion of uranium in uranium monocarbide are being investigated. A technique for sandwiching enriched uranium between samples of carbide for diffusion annealing is being tested. A sampling technique involving grinding with carbide grit on glass has had to be abandoned because sodium from the glass will interfere with the activation analysis. Grinding on a carbide grinding stone is being considered.

Future work will include two additional interdiffusion melts at 1600 C, some additional checks on the thicknesses of carbide layers in melts already prepared, further development and application of techniques for determining rates of self-diffusion of uranium in uranium monocarbide, and a search for suitable techniques for determining self-diffusion of carbon in uranium monocarbide.

Irradiation Effects in UC

A. E. Austin and C. M. Schwartz

The present investigation is intended to study the basic effects of neutron irradiation and fission in uranium carbide. For this purpose, it is planned to use X-ray diffraction and scattering measurements and electron microscopy to determine crystal-structure and physical-structure changes.

The X-ray diffraction method requires use of powder materials. The necessary techniques are being worked out and specimen-handling equipment is being designed.

The use of low-angle X-ray scattering is being considered to determine very small voids or gas bubbles. For this purpose alloys of zirconium and uranium carbides are needed to decrease the X-ray absorption and allow thin sections to be prepared. Alloys of 90 w/o ZrC-10 w/o UC, 80 w/o ZrC-20 w/o UC, and 60 w/o ZrC-40 w/o UC have been arc melted and are being examined for homogeneity. Methods of making the required thin sections will be tried.

Techniques have been evolved for preparation and replication of cast UC for electron microscopy. It is planned to try these methods on irradiated arc-cast UC.

GG. VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

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

Battelle is conducting a program to evaluate steam-void content, heat transfer, and pressure-loss relationships in subcooled boiling-water simulated reactor coolant channels. During the past month experiments were performed in an effort to select the best void-detecting apparatus. Reassembly of the heat-transfer loop was initiated.

Tentatively, it has been decided to begin the investigation with a parallel-plate configuration for the test section. Since steam voids will form on the walls, a void-detecting method which may be used perpendicular to the plates is being sought. Beta-ray absorbtion appears promising if thin-walled test sections are used. Bench tests were conducted using the 2.1-Mev beta from yttrium-90 in equilibrium with strontium-90. The maximum range of these betas is about 1000 mg per cm^2. The 2.98-Mev beta from praseodymium-144 (penetration approximately 1600 mg per cm^2) would permit the use of 10-mil-thick steel test sections with a greater than 0.3-in.-thick water gap. Purchase of such a source is being investigated.

In one experimental arrangement, a 1/8-in.-thick anthracene crystal-photomultiplier-tube detector was used for beta counting. In a second setup an end-window Geiger tube was employed. In both cases it appears that Bremsstrahling limits the useful range of detection to approximately 600 mg per cm^2. Methods of increasing this range are being investigated.

The heat-transfer loop has been partially reassembled. The pump, preheat sections, steam condenser, and much of the loop piping have been installed. Most of the power equipment is on hand awaiting installation.

During the coming month experiments to select the void-detection method will continued. The test-section dimensions and design will be specified. Further details of the loop construction, including electrical wiring, will be completed.

H. PHYSICAL RESEARCH

F. A. Rough

Physical research is reported for programs concerned with the thermal migration of hydrogen in zirconium and with the preparation and properties of refractory uranium compounds. The experimental work on the latter program has been concluded.

A diffusion cell for the study of the migration of hydrogen in zirconium under a thermal gradient is currently being redesigned. The changes were dictated by results obtained with the initial cell constructed.

Diffusion rates of hydrogen in beta zirconium at temperatures of 650 to 850 C have been studied and are found to be described by the equation

$$D = 3.11 \times 10^4 \exp(-44,500/RT).$$

Preparation and Properties of Refractory Uranium Compounds

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

In this work, the effect of processing variables on the physical properties of sintered uranium monocarbide compacts is being sought.

The experimental work has been concluded and final measurements of physical properties are being made. A topical report is in preparation.

Thermal Migration of Hydrogen in Zirconium

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

The migration of hydrogen in zirconium under the influence of a thermal gradient is being examined. Preliminary experiments with the first diffusion cell to be fabricated have indicated that some changes in design are necessary, and these are under way. The measurements of the diffusion coefficients for hydrogen in beta zirconium have been completed. Similar measurements will now be made on the delta zirconium hydride.

Thermal Diffusion

The diffusion cell which was constructed initially consisted of a disk of zirconium hydride separated by stainless steel foil from two copper blocks, the whole being pressure bonded in a stainless steel cylinder. The entire assembly was brought to temperature in a tube furnace, and a thermal gradient was imposed on the diffusion cell by an auxiliary heater. Several design changes have been dictated by the results obtained with the first cell. The zirconium hydride will be formed <u>in situ</u> so that better bonding to the cell jacket can be obtained. In order to get more nearly one-dimensional flow of heat, the entire heating system is being redesigned to provide adiabatic conditions. Provision will be made for measuring the heat flow, so that accurate determination of the thermal gradient may be made.

Diffusion Coefficients

Diffusion rates of hydrogen in beta zirconium have been determined in the range 650 to 850 C from permeation-rate data by both the steady-state and time-lag methods. Experiments were conducted with zirconium disks, 0.03 to 0.1 cm thick, which varied in average hydrogen concentration from 9 to 33 a/o.

The diffusion coefficients determined by both methods with samples of differing thickness are in agreement, indicating that the permeation rates measured are diffusion controlled. Within the limits of experimental error, there was no change in diffusion coefficient with concentration. The diffusion of hydrogen in beta zirconium can be described by the equation,

$$D = 3.11 \times 10^4 \exp(-44{,}500/RT),$$

where the activation energy of the diffusion process is 44.5 kcal per g-atom.

The variation of the diffusion coefficient with temperature and with hydrogen concentration will now be determined for the delta zirconium hydride.

I. SOLID HOMOGENEOUS FUELED REACTORS

W. Diethorn and W. H. Goldthwaite

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

LABORATORY EVALUATIONS OF FUELED-GRAPHITE SPHERES

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

Spherical fueled-graphite elements are being evaluated in preirradiation tests. Selected thermal and mechanical properties, microstructure, and uranium surface contamination are being studied.

During July, metallographic examinations were made of two fueled-graphite spheres, each from a different vendor. The composition of the fuel in both types of elements was determined by etching polished specimens. The fuel particles in one sphere consisted of a UC_2 matrix with UC precipitated both in grain boundaries and as bands within the UC_2 grains. This sphere reportedly contained UC fuel. Fuel particles in the other sphere were composed of varying amounts of UC and UC_2 ranging from 20 to 80 w/o UC. This latter sphere was reported to contain UC_2. The fuel particles in both specimens were fragmented and irregular in shape. Fuel particles in the UC sphere were less fragmented. Average particle size was 104 μ in the UC sphere and 63 μ in the UC_2 sphere.

The uranium surface contamination of one vendor's SiC-coated fueled-graphite spheres was investigated further. Previous alpha counts of the coating surface were supplemented by alpha counts of powder samples taken from successive layers of the SiC coating. The measurements indicate that uranium contamination is distributed throughout the coating.

Pyrolytic carbon coatings on some of the fueled spheres supplied recently by a third vendor showed no apparent leakage in the oil test at 400 F. Coating integrity of these spheres is accordingly better than that reported previously for pyrolytic carbon-coated spheres.

A test to determine the tendency of silicon-silicon carbide coatings to weld together at elevated temperatures was invalidated because of air leakage into the apparatus. The equipment has been modified to avoid this difficulty and is being checked.

FABRICATION DEVELOPMENT OF Al_2O_3-CLAD UO_2 FUEL PARTICLES

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

Densely sintered Al_2O_3 claddings for UO_2 fuel particles are being developed in a new program initiated this month. This work is an extension of the work reported in BMI-1321.

Preliminary cladding trials were made, using a pelletizing technique to apply compacted shells of sinterable Al_2O_3 powder to UO_2 fuel particles. The sintered claddings will be evaluated initially by heating the composite pellets in air at 1200 F and measuring any weight gain resulting from oxidation of the UO_2 cores to U_3O_8.

FISSION-PRODUCT RELEASE FROM FUELED-GRAPHITE SPHERES

W. Diethorn

Fission-product release from fueled-graphite spheres of various compositions is being studied by postirradiation heat-treatment experiments (dynamic activation). Materials of potential value as a nuclear fuel are screened in the dynamic-activation studies. The most promising materials are advanced to the dynamic-irradiation experiments.

Dynamic-Activation Experiments

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

During July, equipment for the measurement of fission-gas release from UO_2-graphite spheres was assembled and tested. The purpose of this study is to measure the release-time history of selected specimens as a function of temperature following low-level neutron irradiation. The equipment includes:

(1) A Vycor sweep capsule which can be heated by induction to temperatures up to 2500 F

(2) A glass charcoal trap for continuous monitoring of the sweep gas (helium) coming from the heated, irradiated sphere

(3) A sodium iodide scintillation crystal, attached to a gamma-ray spectrometer, cooled in dry ice, for continuously measuring the fission-gas activity in the charcoal trap

(4) A double-walled irradiation container for the irradiation of the spheres to 6×10^{15} nvt.

Several admixture balls have been irradiated and are currently being studied with this equipment.

Dynamic-Irradiation Experiments

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

Modification of Sweep Capsule SP-3 was completed in July. After very brief operation in the BRR a leak developed in the gas train, and the experiment was terminated. The level of radiation in the gas train during this short exposure was high, indicating considerable release of fission gas, but it was not possible to measure the release quantitatively.

The capsule has been removed to the hot-cell for postirradiation examination. Hot-cell results will be available in August.

A static-capsule experiment, SP-4, is scheduled for BRR insertion on August 24. This capsule will contain coated and uncoated elements. Specimen temperature will be as close to 2000 F as possible with the objective of studying the conversion of UO_2 to UC and the effect of radiation on the shrinkage of the matrix of the coated specimens.

Fabrication of Sweep Capsule SP-5 is well under way. This capsule, containing pairs of FA-6 and FA-11 elements, is scheduled for BRR irradiation in late September.

Work is continuing on the design of a high-flux high-burnup static capsule, SPH-1, for irradiation in the WTR or MTR. Target irradiation date is November 1, 1959.

POSTIRRADIATION EXAMINATIONS OF FUELED-GRAPHITE SPHERES

R. J. Burian and J. E. Gates

Preparations for the hot-cell examination of specimens in SP-3 have been completed.

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

Evaluations of the corrosion which has occurred on various articles of process equipment used at ORNL for studies of the Fluoride-Volatility process are under way as a program of assistance to the Chemical Technology Division. The amount of corrosion and the mechanisms of attack are being determined by metallographic and other methods.

A metallographic examination of specimens from the Volatility Pilot Plant nickel Mark II fluorinator shell, its internal components, and external auxiliary equipment is in progress. Corrosion profiles of the various vessels and piping are being prepared where possible. A considerable amount of intergranular attack has been observed, especially on the submerged inner walls of the "L"-Nickel shell. Presently, an attempt is being made to establish if this attack was the result of sulfur contamination of either the molten salt or the fluorine sparge.

Corrosion profiles are also being determined for the shell and draft tubes of the Unit Operations' INOR-8 hydrofluorinator. In addition, the type of attack occurring on the baffles, gas-distribution plates, thermocouple tube, and salt-outlet pipe of this vessel is being determined.

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

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

The modified dissolution equipment for the study of radiation effects on the dissolution of fuel materials was checked during July by a test run using 25.1 g of stainless steel sheet and 43.6 g of ThO_2-UO_2 pellets. As in all previous simulated dissolution tests with sheet stainless steel, passivation was not encountered. Insoluble residue was 0.19 g. Core dissolution appeared to be quantitative, with no visible insoluble residue.

In a subsequent test, one of the "cold" Consolidated Edison prototype fuel pins was dissolved, following the standard procedure supplied by ORNL. This pin has been heat cycled and possessed an oxidized surface, which may have contributed to the observed passivation. The suggested addition of formic acid to break the passivation was tried. Two increments, each sufficient to make the sulfuric acid solution 0.1 M in formic acid, were ineffective, possibly because of almost immediate volatilization by the hot acid. The passivation was readily broken by contacting a steel rod to the pin, after which dissolution proceeded smoothly. Dissolution of the cladding, except for the end plugs, appeared to be complete in 3 hr. The subsequent core dissolution appeared, as in the simulated test, to be quantitative. The weight of insoluble filter residue was 0.33 g. Analyses of the products are not yet available.

The need for a few additional modifications to the equipment was indicated by the above tests. After these are completed, one or two more "cold" runs in the laboratory are scheduled, following which the apparatus will be transferred to the hot cell for dissolution tests on the irradiated fuel pins, which have now been received.

K. DEVELOPMENTS FOR SRE, OMRE, AND OMR

EVALUATION OF URANIUM MONOCARBIDE AS A REACTOR FUEL

F. A. Rough

Additional information pertaining to the temperature of three capsules presently being irradiated at the MTR is reported. These capsules each contain two cylindrical as-cast specimens of uranium-carbon alloys. Results of detailed evaluations of specimens from three capsules irradiated to burnups of 0.08 a/o uranium at 600 F, 0.19 a/o uranium at 1600 F, and 0.77 a/o uranium at 1830 F are also presented.

The routine evaluation of enriched metal from which a series of cast UC specimens were to be prepared revealed that the material was not suitable for this purpose. Chromium, iron, nickel, nitrogen, and oxygen were present in particularly high concentrations.

Studies concerned with the thermal conductivity of cast specimens of UC over the temperature range 100 to 1100 C have been initiated.

Irradiation of Uranium Monocarbide

D. Stahl and W. H. Goldthwaite

Recent information pertaining to the temperature levels of Capsules BMI-23-3, BMI-23-4, and BMI-23-5, all under irradiation in the MTR, is presented below. Each of these capsules contains two cylindrical specimens of uranium-carbon alloy (approximately 4.8 w/o carbon).

Capsule BMI-23-3. This capsule operated with a core temperature of about 1200 F during Cycle 124 and about 1300 F during Cycle 125 as estimated from temperatures indicated by the two thermocouples which are still operational; these are both located in the NaK space between the specimens and the water-contacting outer-capsule shell.

Capsule BMI-23-4. Core temperatures in this capsule were up from about 1030 F during Cycle 124 to 1120 F during Cycle 125. These are directly measured core temperatures as indicated by a thermocouple inserted into a hollow present in the upper specimen.

Capsule BMI-23-5. There was also a general increase in core temperature from Cycle 124 (approximately 1300 F) to Cycle 125 (approximately 1400 F) in this capsule. In the case of the latter cycle, the levels are estimated on the basis of data provided by thermocouples located near the water-contacting capsule wall. The thermocouple located in the core of the top specimen failed at the end of Cycle 123.

K-2

Postirradiation Examination of Irradiated Uranium Monocarbide

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

A program is currently being conducted which is concerned with the development of uranium monocarbide as a reactor fuel. To date, three separate irradiation experiments have been completed. In the three irradiation tests, cylindrical specimens of uranium-5 w/o carbon have been irradiated to estimated burnup levels of approximately 0.08 a/o of the uranium at a 660 F maximum fuel-core temperature, 0.19 a/o of the uranium at a 1600 F maximum fuel-core temperature, and to approximately 0.77 a/o of the uranium at a 1830 F maximum fuel-core temperature.

An examination of the experimental data obtained on these specimens indicates that UC behaves much better than uranium, or uranium-alloy fuels operating at the same burnup levels and temperatures. Measurements of the fission gas released by UC during irradiation indicate that very little more gas than would normally be released by recoil alone was released from the surface of the specimens.

As a portion of the radiation-effect studies on UC, an experiment was conducted on one specimen from a capsule containing several specimens irradiated to a burnup of about 0.7 a/o of the uranium at 1475 F (and on one unirradiated UC specimen) to determine the effect of a heat treatment on this material. Specifically, the experiment was designed to show any changes in dimensions and density, and to measure any fission gas released during an 8-hr heat treatment at 2000 F. The estimated amount of krypton-85 released from the irradiated specimen during irradiation was 12×10^{-5} cm^3. Measurement of the fission gas released during the heat treatment showed an additional 2.04×10^{-6} cm^3 of krypton-85 released. Calculations show that the total amount of krypton-85 that could be produced in a specimen of this size with an estimated burnup of approximately 0.7 a/o uranium would be 8×10^{-2} cm^3. It can be seen that the amount of krypton-85 released during irradiation, plus the small additional amount released during the heat treatment, is very much less than the total amount of krypton-85 theoretically produced as a result of irradiation. It therefore is evident that a large amount of fission gas was retained in the UC.

Metallographic examination of the heat-treated irradiated specimen disclosed the existence of large numbers of voids throughout the sample. Because the specimen broke into several fragments during the heat treatment, it was impossible to complete any density and dimensional measurements. The unirradiated control specimen showed a small change in density after the heat treatment.

Future work on UC specimens containing 4.6 and 4.8 w/o carbon will be completed after irradiations up to 11 MTR cycles and to total burnups of about 15,000 MWD/T of UC.

K-3 and K-4

Preparation of UC Pins for Irradiation in the SRE

W. M. Phillips, W. J. Hildebrand,
E. P. Foster, and R. F. Dickerson

The radiation behavior of UC prepared by casting is of interest to Atomics International in connection with the development of sodium-cooled nuclear power reactors. At the present time, Battelle is participating in a program to develop this material by preparing about 6 kg of as-cast shapes of uranium monocarbide and evaluating the preirradiation qualities of these castings.

The specific techniques by which the cast specimens will be prepared have been established. A quantity of uranium enriched to the desired level was received and examined by analytical and metallographic techniques. These evaluations established that the uranium was abnormally high in impurities (containing about 800 ppm of chromium, nitrogen, and oxygen, 2000 ppm of nickel, and 5000 ppm of iron), and that it is, therefore, unsuitable as starting material for the specimens desired.

Recently, another shipment of enriched uranium was received, and analytical and metallographic evaluations have been initiated. If this material is reasonably pure, preparation of the specimens will begin.

Thermal Conductivity of Cast UC

E. A. Eldridge, H. Deem, and C. Lucks

Studies of the thermal conductivity of cast specimens of UC over the temperature range 100 to 1100 C have been initiated. Experimental determinations of the thermal conductivity of six cylindrical-shaped specimens will be made.

Prior to testing, the specimens must be prepared by drilling thermocouple holes along their length and bonding to standard shapes of known conductivity. These preparations have been initiated on two of the specimens presently available.

L. TANTALUM AND TANTALUM-ALLOY STUDIES

J. H. Stang

The status of investigations for the LAMPRE Reactor Development Program at Los Alamos is reported in this section. During July emphasis in the material-development research was placed on establishing metallurgical data for as-rolled control specimens prepared from a recent group of six binary tantalum alloys; duplicate specimens were submitted to LASL for plutonium-alloy corrosion evaluations. A third set of duplicates was vacuum annealed and is now being subjected to various examinations prior to the release of specimens to LASL.

The irradiation capsules containing tantalum tensile specimens are about halfway through their exposure in the MTR core.

Development of Container Materials for LAMPRE Applications

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

Twenty-three strip specimens of tantalum and arc-melted binary tantalum alloys in the cold-rolled condition have been sent to LASL for plutonium-fuel-mixture corrosion testing. The alloys contain additions of hafnium, thorium, titanium, tungsten (zone refined), yttrium, and zirconium. Potassium ferrocyanide tests indicated that the specimens were free of surface iron contamination.

Metallographic examination of the as-cast alloys revealed the presence of a second phase in the tantalum-1.5 and -3.0 w/o zirconium alloys, whereas the other materials in this series consist of a single-phase structure. In the 1.5 w/o zirconium alloy, a very heavy second phase was present within the grains. In both alloys, however, a heavy concentration of a second phase within the grains was observed in a region near one surface of the buttons. The alloys containing additions of 1.5 to 6 w/o hafnium were found to contain segregated dendritic areas near both surfaces of the button.

The hardness and fabricability of this series of alloys are given in Table L-1. These data indicate additions of 0.025 through 1.5 w/o hafnium had no significant effect on hardness. However, additions of 3 and 6 w/o hafnium resulted in a marked increase in hardness. The unexpected high hardness of the 3 w/o hafnium alloy may have been caused by unintentional gaseous contamination during melting. The initial addition of 0.025 w/o zirconium and 0.025 w/o thorium resulted in an increase in hardness. Then, increasing additions up to 0.100 w/o caused a decrease in hardness. Reasons for this behavior are not obvious at this time, but they may be apparent from interstitial-contamination analyses which are now under way. Additions of 1.5 and 3 w/o zirconium resulted in a very large increase in hardness. The tantalum-3 w/o tungsten alloy prepared with zone-refined tungsten showed an increase in hardness comparable to those in the first series of alloys prepared. No significant change in hardness was noted with the alloys containing titanium and yttrium additions.

TABLE L-1. FABRICABILITY AND HARDNESS OF BINARY TANTALUM ALLOYS

Specimen	Nominal Composition (Balance Tantalum), w/o	Hardness, VHN As Cast	Hardness, VHN Cold Rolled to 30-Mil Strip	Edge-Cracking Limit[(a)], per cent reduction after intermediate anneal
44	100 Ta	100	178	79
	0			
46	0.025 Hf	106	154	79
47	0.050 Hf	102	150	62
52	0.100 Hf	111	176	75
49	1.5 Hf	103	280	53
50	3 Hf	224	267	71
51	6 Hf	170	246	71
53	0.025 Zr	125	198	79
54	0.050 Zr	113	164	79
55	0.100 Zr	107	175	79
57	1.5 Zr	190	267	54
56	3 Zr	242	255	43
59	0.025 Ti	86	159	79
60	0.050 Ti	111	143	79
61	0.100 Ti	98	156	63
62	0.025 Th	117	163	79
63	0.050 Th	106	163	72
64	0.100 Ti	94	151	72
65	3 W	129	219	80
66	0.025 Y	100	154	75
67	0.050 Y	94	129	79
68	0.100 Y	100	152	73
69	0.200 Y	100	142	79

(a) Amount of reduction when edge cracking began.

In fabricating the alloys with 1.5 to 6 w/o hafnium and 1.5 to 3 w/o zirconium to 0.030-in. strip, severe edge cracking developed. However, the remainder of alloys were rolled to 0.030-in. strip without any serious edge cracking.

Duplicate specimens of the alloys listed in Table L-1 in the rolled condition have been vacuum annealed at 2600 F. These specimens will be forwarded to LASL as soon as it is determined if recrystallization is complete. Chemical-analysis, hardness, and microstructure data will be obtained on both the rolled and the annealed strip material. In addition to the above properties, tensile data are being obtained on annealed tantalum-1.5 to 6 w/o hafnium and tantalum-1.5 w/o and -3 w/o zirconium alloys.

The preparation of a final series of binary tantalum alloys containing minor additions of aluminum, beryllium, cerium, lanthanum, scandium, and uranium and of a tantalum-1 w/o zirconium alloy will be started during the latter part of August.

L-3 and L-4

Effect of Irradiation on Tantalum

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

The irradiation of encapsulated tantalum tensile specimens is continuing at the MTR. Capsule BMI-25-1, being irradiated to a tantalum-to-tungsten conversion of 1.5 w/o, is scheduled for removal at the end of the current cycle (Cycle 125, ending August 10, 1959). Capsule BMI-25-2 is scheduled for three additional cycles to achieve a tantalum-to-tungsten conversion of 3.0 w/o. Tantalum-1.5 and -3.0 w/o tungsten control alloys have been arc melted, rolled into sheet, and fabricated into tensile bars.

M. DEVELOPMENTAL STUDIES FOR THE PWR

R. W. Dayton

The feasibility of preparing Zircaloy-2-clad compartmented flat-plate fuel elements containing UO_2 cores by gas-pressure bonding has been established. A total of 160 compartments in Zircaloy-clad fuel plates have been successfully burst tested in a large number of specimens. These results establish that consistently bonded fuel elements of high integrity can be prepared by this technique. Cores coated with pyrolytic carbon, vapor-deposited chromium, and sprayed and buffed graphite have prevented core-to-cladding reaction. Excellent bonding was achieved with the chromium-coated and pyrolytic-carbon-coated cores and very good bonding with a few particles of contamination was obtained with the graphite-coated cores.

The optimum process developed consisted of taking UO_2 cores coated with vapor-deposited chromium or pyrolytic carbon and Zircaloy strip components and assembling them into a fuel plate for pressure bonding. The cladding was fusion edge welded to form a gastight assembly which was pressure bonded at 1525 F for 4 hr at 10,000 psi to produce a solid-state bond between all Zircaloy components. The coatings on the core virtually eliminated all core-to-cladding reaction during the bonding cycle.

Techniques were developed to coat by vapor deposition of chromium or pyrolytic carbon both blasted and unblasted UO_2 cores. Tests to date have not revealed any significant difference in fuel elements embodying cores with both types of coatings.

Reports are being prepared on the bonding process and on the coating of UO_2 cores. The only additional work planned on this program is the fabrication of irradiation specimens by the optimum process.

Coating of Uranium Dioxide Cores with Pyrolytic Carbon

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

Adherent coatings of pyrolytic carbon between 2 and 5 μin. thick have been deposited on sand-blasted UO_2 cores. The coatings were obtained by the thermal decomposition of methane gas at 1025 C in a sealed fused-quartz reaction tube placed in a temperature-controlled electrically heated furnace. Nitrogen was used as a mixing and flushing gas. Care was taken to keep the system free from air in order to prevent soot formation. The cores were placed in a fused-quartz boat which could be moved into and out of the hot zone without allowing the entry of air. All sides of the cores were coated simultaneously. For the sand-blasted cores, the deposition time was 20 min.

Attempts to apply adherent coatings of this thickness to as-sintered cores were unsuccessful. However, by varying the deposition time, a thin adherent film of pyrolytic carbon was obtained for a deposition time of 2 min. Large numbers of samples of both kinds were prepared as required for pressure-bonding evaluation. The thickness of the coatings was determined by three methods: (1) from the correlation between film resistance and film thickness*, (2) from the correlation between the deposition time and film thickness**, and (3) from multiple-beam interferometry measurements. The quality of the adherence of the carbon coatings to the UO_2 cores was based on a visual examination, smear tests (rubbing the coatings on a clean white sample), and Scotch-Tape tests (placing the sticky side of the tape against the coating and then removing the tape). If the coating appeared bubbled or blistered, it was found to peel off readily. A large removal of carbon by the smear test or the Scotch-Tape test indicated poor adherence. For the appropriate deposition time, coatings on both sand-blasted and as-sintered cores showed good adherence.

The primary conclusion is that adherent coatings of pyrolytic carbon between 2 and 5 μin. thick can be obtained by this process. However, the surfaces of the cores must be roughened in order for a carbon coating of this thickness to be adherent. At present, the only satisfactory method for preparing the surfaces of the cores is sand blasting. The possibility of economically coating UO_2 cores with pyrolytic carbon by the thermal decomposition of methane gas on a production basis is being explored. Letters requesting a cost estimate are to be sent to manufacturers who use this process and who might be interested in doing the work on a production basis. This information will be included in a report being prepared on this program.

Coating of Uranium Dioxide Cores with Vacuum-Evaporated Chromium

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

Adherent coatings of chromium between 25 and 40 μin. thick have been deposited on both sand-blasted and as-sintered UO_2 cores by the process of vacuum metallizing. Pure chromium was evaporated in vacuo from an electrically heated tungsten or molybdenum boat. The chromium then condensed on the cores which were mounted at a suitable distance from the source. A chromium charge weighing between 1.5 and 2.0 g, at a distance of approximately 18 cm from the cores, was found to yield a coating between 25 and 40 μin. thick. Evaporations were carried out at a pressure less than 5×10^{-5} mm of mercury. The cores were mounted in a rotating jig driven by an electric motor located outside the vacuum chamber. Four sides of the cores could be coated in one evaporation with the rotating assembly. Separate evaporations were necessary for coating the ends.

Large numbers of samples were prepared for pressure-bonding evaluation. The thicknesses of the coatings were calculated by measuring the weight increase of the cores. Coatings on both sand-blasted and as-sintered cores showed good adherence. The quality of the adherence of the chromium coatings to the UO_2 cores was based on the results of a visual examination, smear tests (rubbing the coating on a clean white surface), and Scotch-Tape tests (placing the sticky side of the tape against the coating and

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*Grisdale, R. O., Pfister, A. C., and Van Roosbroeck, W., "Pyrolytic Film Resistors: Carbon and Borocarbon", Bell System Tech. J., 30, 271-314 (1951), Figure 12.

**Ibid, Figure 3.

then removing the tape). If the coating appeared powdery, it was found to dust off readily. A large removal of chromium by the smear test or the Scotch-Tape test indicated poor adherence.

The primary conclusion is that vacuum metallizing is a feasible process for obtaining thick and adherent chromium coatings on both sand-blasted and as-sintered UO_2 cores. The possibility of economically coating UO_2 cores by this method on a production basis is being explored. Letters requesting a cost estimate are to be sent to manufacturers who do vacuum coating and who might be interested in doing the work on a production basis. If sufficient information is available from replies to these letters, cost estimates will be included in the report being prepared on this phase of the program.

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

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

Investigations have continued on the fabrication of Zircaloy-2-clad compartmented uranium dioxide fuel elements by gas-pressure bonding. The use of barrier coatings of sprayed and buffed graphite, pyrolytic crystalline carbon, and vacuum-evaporated chromium on UO_2 cores was studied to determine the effectiveness of each of these coatings in preventing core-to-cladding reaction. The possible effect of each of the coatings on the Zircaloy-to-Zircaloy bonds in the specimens was also investigated. The elements prepared for these studies were approximately 3.5 in. wide and 8 in. long with a thickness of 0.150 in., consisting of a 0.100-in.-thick receptacle plate and two 0.025-in.-thick cladding plates. Each element contained thirty-two 0.100 by 0.250 by 1.500-in. cores. Each core was contained in an individual compartment formed by 0.035-in.-wide cross ribs of Zircaloy. The compartmented receptacle plate was assembled entirely from strip components which had been surface finished by machine belt abrasion. In the gas-pressure-bonding technique, the cladding components were fusion edge welded, evacuated, and sealed to form a gastight assembly which was then gas-pressure bonded at 1525 F and 10,000 psi for 4 hr. Recently bonded elements were evaluated in the as-bonded condition only. Previously, some of the specimens were heat treated for 5 min in a salt bath at 1850 F subsequent to pressure bonding; however, results of the program demonstrated that this additional operation is not necessary. The elements were evaluated by internal-pressure burst tests, metallographic examination, and intercompartmental-leakage tests and corrosion tests of purposely defected compartments.

A large number of internal-pressure burst tests were conducted on elements containing cores coated with sprayed and buffed graphite, pyrolytic carbon, or vacuum-evaporated chromium. These elements, which were prepared using improved sealing, cleaning, and rinsing techniques, were consistently found to have strong bonds. A total of 166 consecutive tests of compartments in the 38 specimens bonded resulted in all cases in failure of the cladding material at high pressures with normal cladding deflections, rather than failure of the bonds. Of these tests, 36 were of specimens containing graphite-coated cores, 62 were of specimens containing pyrolytic-carbon-coated cores, and 68 were of specimens containing cores coated with chromium. Approximately eight burst tests for each type of core coating were of portions of specimens prepared early in the series which had received the additional heat treatment at 1850 F for 5 min. Also

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included in these results are tests of 12 compartments in as-bonded elements which contained cores having evaporated-chromium coatings or pyrolytic-carbon coatings that were not sand blasted prior to coating. As discussed in other sections of the report, most of the cores to be coated with pyrolytic carbon or chromium had been sand blasted to obtain the optimum condition for adherency of the coating. However, more recent results indicate that the coatings of chromium applied to cores in the as-sintered condition have satisfactory adherency as compared with coatings applied to sand-blasted cores to the desired coating thickness range of 25 to 50 μin. Therefore, it is concluded that the sand-blasting treatment is not necessary prior to the application of this type of coating. In order to apply adherent coatings of pyrolytic carbon to unblasted cores, it was necessary to reduce the thickness of the coating from about 3 to 5 μin. to about 0.5 μin. The burst tests mentioned above of an element containing these cores indicated that the coatings were adherent and did not result in contamination of the bonds; however, the effectiveness of these thinner pyrolytic-carbon coatings in preventing core-to-cladding reaction will have to be determined based on the results of corrosion tests now in progress of purposely defected compartments.

All of the graphite coatings were applied by spraying a suspension of graphite in water onto unblasted cores. The cakes were then outgassed and buffed, giving a coating of about 5 mg of graphite per in.^{2} of core surface. The outgassing and buffing treatments, which are not necessary for the pyrolytic-carbon and evaporated-chromium coatings, were designed to remove volatile binders and to minimize dusting and flaking of the sprayed graphite coatings. Also bonded in this series of elements was one specimen containing sand-blasted cores coated with pyrolytic carbon which was prepared so as to determine if the Zircaloy bonding surfaces could be contaminated by the core coatings due to mishandling during assembly. During assembly of this element, the cores were purposely shifted in position over the bottom Zircaloy cladding plate which caused them to rub against the relatively rough bonding surfaces. Also, after the cores and receptacle-plate components were in position, the entire top surface of the cores and ribs was hand rubbed, using rubber gloves, in an attempt to transfer carbon from the cores to the surfaces of the ribs. Six burst tests of this element did not result in any bond failures, indicating that at least some mishandling can occur during assembly of cores which have been sand blasted and coated with pyrolytic carbon probably without resulting in poor bonding.

Corrosion tests in 680 F water of defected compartments of as-bonded elements and elements heat treated at 1850 F for 5 min which contain cores coated with pyrolytic crystalline carbon, vacuum-evaporated chromium, and sprayed and buffed graphite have been continued. Specimens in the as-bonded or heat-treated conditions containing cores coated with graphite or crystalline carbon have shown no growth during several weeks of exposure. These results for the graphite-coated cores are corroborated by similar corrosion tests of as-bonded and of heat-treated elements containing cores properly coated with graphite which were prepared during previous phases of this program. The corrosion tests on these elements have been continued and have produced no significant growth during several months of exposure. Specimens in the as-bonded and heat-treated conditions containing cores with chromium coatings 25 to 100 μin. thick have shown no growth during several weeks of corrosion testing. Chromium coatings only 10 μin. thick were not sufficient to prevent core-to-cladding reaction, however, and growth was produced during 3 weeks of exposure. Intercompartmental-leakage tests of all of the elements before and after corrosion testing have revealed no communication between any of the compartments.

M-5 and M-6

The results of metallographic examinations of these elements are in agreement with the results of the burst and corrosion tests. As-bonded, as well as heat-treated, elements containing cores with the three types of coating have excellent metallurgical bonds, showing grain growth across the original interface in all areas with a very minimum amount of bond-line contamination. No core-to-cladding reaction was observed in specimens with core coatings of pyrolytic carbon or graphite. Specimens with 25 to 100-μin.-thick chromium coatings showed no core-to-cladding reaction; however, 10-μin.-thick coatings of chromium permitted some reaction.

It has been concluded from this study that the feasibility of preparing Zircaloy-clad flat-plate fuel elements containing compartmented UO_2 fuel by the gas-pressure-bonding technique has been established. Elements produced by developed procedures behave well in corrosion tests, have consistently strong Zircaloy-to-Zircaloy bonds, possess complete compartment integrity, reveal good dimensional control, and demonstrate strong and ductile cladding. The use of Zircaloy ribs between each of the cores, thus forming individual compartments, minimizes flow of the cladding plates into relatively large void areas in the receptacle plate which could be present due to the use of chipped cores or poor dimensional control of the compartments. Coatings on the cores of sprayed and buffed graphite, pyrolytic crystalline carbon, or vacuum-evaporated chromium are satisfactory to prevent core-to-cladding reaction without introducing contamination on the Zircaloy bonding surfaces of the element. The graphite coatings are not as desirable as the other two types, however, since the coated cores must be carefully handled during assembly to avoid contamination of the bonding surfaces.

A topical report is being prepared to cover this phase of the program. This report will include some additional corrosion data now being compiled.

N. DEVELOPMENTS FOR THE MGCR

W. C. Riley and W. H. Goldthwaite

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

Two methods have been found which apparently eliminate microscopic cracks which appear to originate and terminate at exposed UO_2 grains in fueled BeO pellets. Both involve special treatment of the fuel to cause the shrinkage in the fuel particles during sintering to more nearly approximate that of the matrix.

Attempts to increase the matrix density and thereby decrease the permeability of fueled-graphite materials continued. Repeated pitch impregnation has resulted in some decrease in permeability but not to the extent that appreciable fission-product retention by the graphite would be expected. The use of minor additions has not resulted in significant densification of the graphite.

A method for cladding UO_2 particles with BeO has been devised. The cladding is impervious to the extent that no weight increase is observed after the pellets have been heated in air at 1200 F for 18 hr. Clad particles can be dispersed in either a graphite or a BeO matrix. Experiments have been initiated to determine the time-temperature conditions which will cause reaction of graphite and BeO.

Preliminary neutron-activation experiments have shown that BeO-UO_2 samples exhibit better fission-product retention than available graphite-UC and graphite-UC_2 samples. Fission-product retention appears to be directly proportional to the density of the BeO matrix.

N-2

UO_2 Dispersions in BeO

A. K. Smalley and W. H. Duckworth

Densely-sintered matrices of BeO containing about 20 volume per cent of uniformly dispersed UO_2 particles are being investigated for fuel-element-core applications.

Microscopic examinations of fueled BeO pellets prepared by the method described in BMI-1346 indicated that all pellets contained minute radial cracks. These cracks appeared to originate and terminate at exposed UO_2 grains. The cracks are attributed to the unequal shrinkages of the BeO matrix and to fuel particles during sintering.

An attempt was made to overcome this cracking by increasing the shrinkage of the fuel particles. Experiments were carried out in which particles of either UO_3 or UO_2 plus 10 w/o $Be(OH)_2$ were substituted for the UO_2 particles used previously. The enhanced shrinkage in the fuel resulted from either the reduction of UO_3 to UO_2 or the dehydration of $Be(OH)_2$ during sintering. Using the high-shrinkage fuel and LOH-grade BeO, pellets were prepared by techniques similar to those used for the pellets in which cracks were found.

Microscopic examinations of the sintered pellets showed that they apparently were crack-free. The sintering conditions and bulk densities for these pellets are shown in Table N-1. The bulk densities of pellets in which UO_3 was the starting fuel material were somewhat lower than those of the pellets in which the fuel was UO_2-$Be(OH)_2$. The densities of both groups were lower than the densities of the previously reported pellets in which the fuel was compacted UO_2.

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

	Sintering Conditions(a)		Sintered Bulk Density(b)	
Starting Fuel Material	Maximum Temperature, F	Time at Maximum Temperature, hr	G per Cm^3	Per Cent of Theoretical(c)
UO_2-10 w/o $Be(OH)_2$	2600	1	4.15	89.5
	2800	1	4.42	95.2
	3000	1	4 35	93.7
	3000	4	4.43	95.5
UO_3	2600	1	4.05	87.4
	2800	1	4.17	90.0
	3000	1	4.20	90.6
	3000	4	4.22	90.9

(a) In flowing hydrogen.

(b) Average of five specimens, by xylene absorption and displacement.

(c) Theoretical density taken as 4.64 g per cm^3 for bodies containing 48.5 w/o UO_2 and balance BeO.

Metallographic examinations are being made of the specimens containing high-shrinkage fuel in an attempt to determine whether its use causes major differences in the specimen microstructure. Also, permeability measurements are being made to determine whether the porosity in the pellets is continuous.

UC and UC_2 Dispersions in Graphite

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

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

Last month it was reported that the gas permeability of selected test specimens was decreased by approximately a factor of 2 by impregnation with coal-tar pitch. During July, several of these specimens were pitch-impregnated a second time. This also resulted in a permeability decrease by approximately a factor of 2. However, the measured permeability is still far too high to expect appreciable fission-product retention by the graphite.

The investigation of minor additives to aid in densification of compacts was continued. Nonfueled compacts were formed from petroleum coke and coal-tar pitch. These were prepared by the conventional hot-mixing method, or by mixing at room temperature.

For hot-mixed samples minor amounts of lubricating oil were added to the pitch coke mix at room temperature just prior to forming. No improvement in bulk density of baked compacts resulted from oil additions.

For samples mixed at room temperature, either benzene or carbon tetrachloride was used as a solvent for the pitch to reduce viscosity. After mixing the volatile solvents were removed. Paraffin in amounts of about 3.0 or 5.5 w/o or about 5.0 w/o beeswax dispersed in either carbon tetrachloride or benzene was added to the mix. Again, solvents were removed prior to forming. No apparent increase in bulk density of baked compacts resulted from these additions.

Compacts made from the hot-mixed body had a slightly higher bulk density than those made from bodies prepared by the solvent method. Equal bulk densities were obtained for bodies prepared with benzene or carbon tetrachloride.

In future work, the investigation of methods to decrease the permeability of fuel-containing compacts will be continued.

Cladding of UO_2 Particles With BeO

A. K. Smalley and W. H. Duckworth

Techniques for cladding UO_2 particles with impermeable shells of BeO are being developed.

N-4

During July, a laboratory procedure for applying densely sintered BeO claddings to UO_2-base fuel particles was developed and refined. Briefly, the method consists of the following operations:

(1) Davison ceramic-grade UO_2 was ball-milled for 2 hr with 10 w/o of $Be(OH)_2$ (6.06 w/o BeO). The mixture was treated with 3 w/o of beeswax in a CCl_4 solution, and evaporated to dryness. The dried, wax-containing fuel was compacted at 10,000 psi in a steel die, and the compact was crushed and ground to provide fuel particles in the range of minus 18 to plus 30 mesh.

(2) The fuel granules were placed on a 12-in.-ID rubber-lined dryer, and the dryer was rotated at 33 rpm. The tumbling granules were alternately dusted and sprayed with as-received LOH-grade BeO powder and a slip containing the following:

LOH-grade BeO	98 g
Carbowax 4000	1.75 g
C_2H_5OH	185 ml
H_2O	185 ml
HCl, 12 N	6 ml

When composite pellets of the desired size were obtained, the process was stopped and the pellets were dried at 120 F.

(3) The dried, clad pellets were mixed with 70 volume per cent of Alcoa A-14 minus 100 plus 200-mesh calcined Al_2O_3. The mixture was placed in rubber molds and compacted isostatically at 100,000 psi. The BeO-clad UO_2 pellets were separated from the pressing matrix by screening. Excess Al_2O_3 was removed from the surface of the pellets by tumbling them dry for 2 hr and then rescreening.

(4) The compacted pellets were sintered in a hydrogen atmosphere. They were soaked at 1200 F for 4 hr to allow complete dehydration of the $Be(OH)_2$ in the fuel cones, heated to 2800 F, and soaked for 1 hr.

The sintered BeO-UO_2 pellets were predominantly in the size range of minus 10 plus 16 mesh. Chemical analyses indicated that they contained about 27.4 w/o of UO_2.

Portions of the pellet batch were heated in static laboratory air at 1200 F. No weight gain in the samples was observed after 18 hr at 1200 F. This indicated that the BeO claddings effectively prevented oxidation of the UO_2 to U_3O_8. Therefore, the claddings appear to be sufficiently crack-free and nonporous to prevent diffusion of oxygen into the UO_2 core.

Composite BeO-UO_2 pellets similar to those described above are being prepared for neutron-activation testing.

Carburization Studies in the BeO-Graphite System

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

One of the fuel elements under consideration for the MGCR consists of BeO-clad UO_2 pellets dispersed in graphite. Chemical reaction between the BeO and the graphite matrix may have an adverse effect on the ability of the cladding to retain fission products. Preliminary experiments have been initiated to indicate the time-temperature conditions required to cause chemical reactions, and to indicate the effect of the reaction on fission-product release as determined by neutron activation.

Preliminary Characterization by Neutron Activation

P. Gluck and M. Pobereskin

Recent screening tests on BeO-UO_2, UC-graphite, and UC_2-graphite specimens with respect to fission-product release have been unsuccessful because of failure of fused silica, commonly called quartz, capsules.

Cracking of the capsules was eliminated when pretreated charcoal was encapsulated with the specimens and when the annealing temperature was reduced to 1800 F. In the pretreating operation, the impurities were removed from the charcoal by heat treatment at 300 F for 24 hr, at a terminal vacuum of 5×10^{-6} mm of mercury.

With the capsule-cracking problem solved, the screening studies were resumed with the UC_2-graphite specimens and the BeO-UO_2 specimens. The graphite-matrix specimens were irradiated for 1-3/4 hr, while the BeO-UO_2 specimens were irradiated only for 1-1/6 hr. The approximate fluxes were 6.2×10^{11} and 2.0×10^{12} nv, respectively. The annealing temperature, 1800 F, was the same for both groups. Prior to annealing, the irradiated specimens were individually encapsulated in 7-mm quartz tubing along with 1 cm^3 of the pretreated charcoal. The capsules were sealed and then annealed for 24 hr at 1800 F in a muffle furnace. The liberated fission gases were adsorbed on the activated charcoal at liquid-nitrogen temperature. Gamma-ray spectrographs were obtained from the activities in the charcoals.

The gamma-ray spectrograph revealed that xenon-133 and iodine-131 made up the bulk of the activity. From the data obtained and radiochemical calculations, using the appropriate decay periods, the per cent fission gas released and per cent uranium burnup were calculated and are shown in Table N-2.

TABLE N-2. FISSION-GAS RELEASE FROM UC_2-GRAPHITE AND BeO-UO_2 SPECIMENS HEATED IN VACUUM AT 1800 F FOR 24 HR

Specimen Type	Firing Temperature, F	Weight[a], g	Fuel Content, w/o	Flux, nv	Total Uranium Burnup, a/o	Fission Gas Released, per cent	
						Xenon-133	Iodine-131
UC_2-graphite	2000	0.4144	61.3	6.41×10^{11}	1.52×10^{-6}	1.76	2.11
UC_2-graphite	2000	0.4132	61.3	6.13×10^{11}	1.49×10^{-6}	2.06	2.68
UC_2-graphite	2700-2720	0.5686	61.3	6.24×10^{11}	1.46×10^{-6}	1.10	1.32
UC_2-graphite	2700-2720	0.5985	61.3	6.23×10^{11}	1.46×10^{-6}	1.50	1.95
UC-graphite	2350	0.4262	60.3	6.27×10^{11}	1.47×10^{-6}	0.49	0.64
UC-graphite	2350	0.4280	60.3	6.30×10^{11}	1.47×10^{-6}	0.58	0.89
BeO-UO_2	2600	0.9201	48.25	2.31×10^{12}	3.94×10^{-6}	0.018	0.030
BeO-UO_2	2600	0.9300	48.25	2.10×10^{12}	3.58×10^{-6}	0.026	0.058
BeO-UO_2	2600	0.9376	48.25	2.09×10^{12}	3.57×10^{-6}	0.025	0.048
BeO-UO_2	2800	0.9072	48.25	1.81×10^{12}	3.11×10^{-6}	0.004	0.010

(a) Before and after neutron activation. No change observed.

The fission-gas-release data in Table N-2 show that the BeO-UO_2 materials exhibited better fission-product retention than the graphite-matrix materials. Within the BeO-UO_2 group, the pellets with the highest bulk density retained the fission gases best. There appears to be a good correlation between density and fission-product retention.

STUDIES OF FISSION-GAS RELEASE FROM FUEL MATERIALS

M. Pobereskin

Design, construction, and assembly of equipment continued for neutron-activation and in-pile gas-release studies. Experimental work in these two phases of the study should start in September and November, respectively.

Detailed Neutron-Activation Studies

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

During July progress was made in assembling the apparatus for the postirradiation fission-gas-release study to be conducted in the radioisotopes laboratory. It now appears that the assembly will be completed in the latter part of August or the beginning of September at which time the experimental work will be started.

N-7

In-Pile Studies

N. E. Miller and G. E. Raines

A tentative design has been established for the in-pile capsule. The theoretical analysis indicates that the range of temperature and surface-heat-flux parameters can be obtained with two capsules. The two capsules will differ only in the size of the thermal-barrier gas annulus. The design will be experimentally checked with a thermal mock-up of the capsule.

The operation of the first capsule has been deferred until November 1, 1959.

HIGH-BURNUP IRRADIATION EFFECTS IN FUEL MATERIALS

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

A study of the stability of ceramic fuels under conditions simulating those of the MGCR design is in progress. Fuels being considered are uranium dioxide in beryllium oxide, uranium monocarbide in graphite, and uranium dicarbide in graphite. Two specimens of each type containing about 20 per cent fuel compound, 93 per cent enriched, were irradiated in the BRR for four reactor cycles at surface temperatures of about 1500 F. Each specimen was composed of four pellets approximately 0.222 in. in diameter by 0.250 in. long. The pellets were sealed in stainless steel tubes with 0.012-in.-thick walls under a helium atmosphere. The capsule used in the irradiation was equipped with electrical heaters and thermocouples. The specimens have been recovered from the capsule and the postirradiation examination is nearly complete.

The postirradiation examination included sampling the gas contained in the capsule for fission-gas content, measuring the dimensions and density of the specimens, sampling the gas contained inside each specimen for fission-gas content, measurement of the fuel-pellet dimensions, measurement of the density of the BeO pellets, and metallographic examinations of each fuel material. Radiochemical burnup analyses were planned but could not be accomplished due to time limitations.

The exterior surfaces of the irradiated specimens appeared undisturbed and in good condition. The physical dimensions and relative densities of the specimens were measured and the results indicated that no changes in dimensions or density had occurred. The specimens were then sampled for fission gas. The results of the fission gas analyses are given in Table N-3. Fission gas was not detected inside the capsule, indicating that the specimens were sound. The BeO pellets released about 0.13 per cent of the total fission gas formed, while the graphite pellets containing UC released about 0.4 per cent and those containing UC_2 released about 0.3 per cent at about the same temperature and burnup.

TABLE N-3. FISSION-GAS-RELEASE DATA FROM CAPSULE BMI-31-1

Specimen	Pellet Composition, w/o	Burnup, 10^{19} fissions per cm^3	Surface (Cladding) Temperature[a], F	Total Gas Produced, 10^{-2} cm^3	Total Fission Gas Determined, 10^{-5} cm^3	Fission Gas Lost[b], per cent
Inner capsule	----------No fission gas detected----------					
3	48.25 UO_2 in BeO	8.14	1550	1.82	2.36	0.13
4	66.6 UC_2 in graphite	5.85	1275	1.29	3.9	0.30
9	61.3 UC in graphite	9.24	1560	2.02	8.08	0.40

(a) Estimated from thermocouple readings, values are ±30 F.

(b) Values reported are ±25 per cent.

The pellets recovered from the specimens sampled for fission gas were fractured. This was apparently the result of deformation of the cladding during the puncturing operation. Results of the physical measurements of sound pellets removed from Specimens 1, 4, 8, and 9 are reported in Table N-4. The BeO pellets show essentially no diameter change due to irradiation, while the fueled-graphite pellets show decreases ranging from 1.2 to 2.0 per cent. The relative density of only the BeO pellets from Specimen 1 was measured. The results indicate that the decrease in density was less than 1 per cent, which is within the experimental accuracy of the measurements.

TABLE N-4. CHANGES IN PHYSICAL MEASUREMENTS OF PELLETS

	Preirradiation Measurement, in.		Postirradiation Measurement, in.		Change in Dimensions, per cent	
Specimen and Compositions	Length of Pellet Stack	Diameter of Pellet	Length of Pellet Stack	Diameter of Pellet	Length	Diameter
Specimen 1, UO_2 in BeO pellets						
Pellet 1		0.2220		0.2220	--	0
Pellet 2	0.9980	0.2217		0.2217	--	0
Pellet 3		0.2219	(a)	0.2218	--	Nil
Pellet 4		0.2220		0.2219	--	Nil
Specimen 8, UC in graphite pellets						
Pellet 1		0.2205		0.2162		-1.9
Pellet 2	0.9985	0.2221	0.9997	0.2182	0.12	-1.8
Pellet 3		0.2220		0.2193		-1.2
Pellet 4		0.2210		0.2166		-2.0
Specimen 9, UC in graphite pellets						
Pellet 1		0.2216	Too badly chipped to measure			--
Pellet 2	0.9984	0.2217		0.2179	--	-1.7
Pellet 3		0.2210	(a)	0.2175	--	-1.6
Pellet 4		0.2207		0.2179	--	-1.3
Specimen 4, UC_2 in graphite pellets	Pellets 1 and 2 badly crushed					
Pellet 3	0.9990	0.2235		0.2205		-1.4
Pellet 4		0.2234	(a)	0.2202		-1.4

(a) Length could not be obtained because one pellet or more was damaged during removal.

A metallographic examination has been performed upon pellets from Specimens 1, 5, and 9. In addition, the Type 316 stainless steel cladding from each of the fuel types was examined metallographically to determine if any reaction occurred between cladding and fuel materials. During grinding and polishing, it was noted that the two specimens containing uranium carbide in graphite were pitted by loss of the graphite matrix. Since it was suspected that the water used as lubricant was causing graphite to loosen and wash away, attempts were made to prepare the specimens upon dry wheels. Neither method was successful in retaining all of the graphite, with the result that these specimens showed pitted and mottled matrices. Examination of the fueled portions of the specimens, however, showed no evidence of irradiation damage; the particles of UC and UC_2 were not noticeably different from their preirradiation condition. The UO_2-BeO specimen retained its BeO matrix during metallographic preparation. However, there was considerable void space immediately surrounding the majority of UO_2 particles. Void space was observed previously in low-density unirradiated specimens, and may be due to the method of pellet preparation, or to metallographic grinding and polishing operations. The UO_2 particles themselves did not appear to be affected by irradiation.

The remaining three capsules to be examined in this program have been in the MTR since the beginning of Cycle 125. BMI-31-1a was placed in MTR position A-26-SE, BMI-31-2 was placed in A-14-NW, and BMI-31-3 was placed in A-14-NE. Current information indicates that only one of the three heaters in each capsule is in operation, resulting in lower specimen temperatures than requested. It is believed that incorrect heater hookup is responsible, and this will be corrected if possible.

DIFFUSION OF FISSION PRODUCTS IN CLADDING MATERIALS

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

A program to study the diffusion of fission products in "A" Nickel cladding material is being conducted. Specimens for the study have been impregnated with fission products by irradiation recoil. For this purpose a capsule containing 20 sandwich specimens of 93 per cent enriched uranium foil between nickel foils 1, 5, and 10 mils thick has been irradiated in the Battelle Research Reactor for 3 hr, and is currently being held in the reactor pool to permit radioactive decay for safe handling. The capsule activity is being checked periodically, and, when it is at a safe level, the capsule will be opened and the sandwiches removed for subsequent heat treatment and analysis.

Work is under way to adapt a vacuum hot press to the task of pressure bonding the nickel foils to each other, after removal of the uranium. With this apparatus, an applied load of 6 tons can be administered at about 1600 F, which should be sufficient for good metallurgical bonding of the nickel.

Samples have also been prepared to study the introduction of rare gas into the metal lattice by cathodic bombardment. Two samples of "A" Nickel were subjected to argon bombardment and then surface cleaned to remove surface-absorbed gases. These were then heated to 900 C to drive off any absorbed argon. Due to instrumental difficulties, spectrographic analyses for released argon have not been performed as yet. However, the analyses should be performed during the coming month.

N-10

CARBON-TRANSPORT CORROSION STUDIES

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

In support of the MGCR program, Battelle is investigating the effect of radiation on coolant-core materials proposed for this high-temperature graphite-moderated helium-cooled reactor. Potential problem areas under study are carbon transport, and metal oxidation and carburization.

Hot-cell examination of metal specimens and graphite specimens from Capsule GA-2-2, containing four quartz tori (harps), was continued in June. Weight-change data and the results of cursory visual examination were reported for these specimens last month. Before the capsule was irradiated dosimetry measurements were made in the BRR core position used for this experiment. Results are summarized as follows:

Total fast-neutron flux (fission spectrum)	5×10^{13} n/(cm^2)(sec)
Thermal-neutron flux	4×10^{13} n/(cm^2)(sec)
Absorbed dose rate in graphite (fast neutron plus gamma)	2×10^{8} rads per hr
Ratio of fast-neutron dose rate to gamma dose rate	1

An examination of photomicrographs (100X) shows that all the low-temperature (960 F) specimens were essentially unaffected. The Monel specimen irradiated at 1500 F with no getter in the torus showed intergranular attack or separation to a depth of 1.5 to 2 mils. Getter in the other torus reduced this intergranular attack but the surface zone, to a depth of 2 mils, was different from the interior of the specimen. The nickel specimens at the high temperature showed this same general behavior but to a much smaller degree. No change in surface structure was observed in the niobium-zirconium specimens. Weight change data could not be readily correlated with the metallographic results. In these metallographic examinations control specimens, prepared in the hot cell by the same metallographic techniques, were used for comparison purposes.

Metallographic examination of several graphite specimens from the tori revealed no signs of attack or change in microstructure.

Based on these data from a short in-pile experiment, Monel is attacked more readily than nickel or niobium-1 w/o zirconium in the presence of graphite and good-quality virgin helium at 1500 F. At lower temperatures (960 F) none of the metals are attacked under these conditions. The nature of the attack has not been fully defined. There is no evidence for carbon transport.

Longer irradiation exposures are needed to define the significance of these results to the MGCR. The effect of temperature only should be studied in duplicate tori in order to define the role played by thermal effects.

Additional metallography and microhardness determinations are under consideration in future work with the irradiated metal specimens.

O-1 and O-2

O. ENGINEERING ASSISTANCE TO KAISER ENGINEERS

Reactor-Flow Studies

L. J. Flanigan and H. R. Hazard

Studies of flow in a quarter-scale air model of the Partially Enriched Gas-Cooled Power Reactor are being conducted at Battelle to provide design data for the prototype. Work previously reported includes receipt of the plastic-model parts, assembly of the model, installation of the model in the test facility, and a preliminary run to check general operation and instrumentation.

In July, equipment was set up to inject titanium dioxide into the model for visual flow tracing and to inject and measure gaseous sulfur dioxide in the model to determine the amount of mixing occurring from one location to another.

Two core-support-cylinder configurations have been investigated as part of the program to determine the effect of hole size and location on core-flow distribution, mixing, and pressure drop.

In August, several additional core-support-cylinder configurations, varying in hole pattern, will be studied. Based on the results of these studies, a configuration will be selected for use in subsequent model studies.

P. DEVELOPMENTAL STUDIES FOR THE SM-2

S. J. Paprocki

Studies are being conducted in assistance to Alco Products concerned with the development of fuel, absorber, and suppressor materials for the SM-2.

Test fuel specimens for irradiation in the MTR and ETR have been fabricated. These specimens embody ORNL hydrothermal UO_2, Mallinckrodt spherical UO_2, and a boron burnable poison in the form of a dispersed boron compound.

Studies are in process to develop a fabrication process for the preparation of full-size reference fuel elements. The reference fuel system consists of spherical UO_2 fuel and ZrB burnable poison dispersed in a Type 347 prealloyed stainless matrix and clad with Type 347 stainless steel.

Three noninstrumented capsules containing either seven or eight test specimens each have been forwarded to the MTR for irradiation. These capsules will be inserted in the MTR core and exposed to an unperturbed flux of from 3 to 5 x 10^{14} nv.

Materials Development

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

An investigation is being made to determine suitable materials and fabrication technique for fuel elements containing a burnable poison, suppressor sections which can be attached to or incorporated in the fuel elements, and control rods for the SM-2 reactor. In addition, irradiation specimens are being prepared as required for use in the radiation-testing program.

Development of Fuel Materials

Techniques are being developed for the fabrication of reference fuel elements which contain cores of 26 w/o UO_2 and 0.21 w/o boron in the form of ZrB_2 dispersed in a Type 347 prealloyed stainless matrix (0.030 in. thick) and clad with 0.005-in.-thick Type 347 stainless. Fabrication procedures are being evaluated on the basis of metallographic examination, chemical analyses, and mechanical properties.

Three full-size fuel plates have been hot rolled using cores pressed in a new 3 by 2.581-in. die. It is not expected that the dimensions of these plates will exactly match the final specified size of the SM-2 reference plates. However, since final specifications have not been released it is necessary to use an approximate size in order to establish dimensional control and tolerances. Variations in core and cladding dimensions at various stages in the roll-cladding process are being followed by means of radiographic and metallographic techniques.

P-2

Small-scale specimens are being used to determine the effect of such variables as total reduction. Two series of specimens have been roll clad using total reductions ranging between 5 to 1 and 10 to 1 in thickness. The specimens all contained cores of 28 w/o minus 100 plus 200-mesh high-fired UO_2 and 1.2 w/o ZrB_2 dispersed in a prealloyed Type 347 stainless powder matrix and clad with Type 347 stainless. The plates were hot rolled at 2200 F to a 40 per cent reduction on the first pass followed by 30 cent per remaining pass. After a 30-min anneal at 2200 F they were cold rolled 15 per cent and annealed 1 hr at 2050 F. The plates were evaluated for core and core-to-cladding bond strength by pulling the plate in tension across the thickness direction at room temperature and 600 F. In all the tests fracture occurred in the core, indicating that the core-to-cladding bond was stronger than the core. Previously reported results of the room-temperature tests show a trend to decreasing strength at higher total reduction. Similar results are shown in Table P-1 for the 600 F tests. Individual strengths varied from 10,700 psi for a 10-to-1 reduction to 14,600 psi for a 5-to-1 reduction. Although this spread is not much greater than what would be expected at any one total reduction if a large number of tests were run, there is a definite trend toward decreasing strength at higher reductions. Specimens with a 15-to-1 total reduction are being prepared to complete the series.

TABLE P-1. RESULTS OF TRANSVERSE TENSILE TESTS ON SMALL-SCALE FUEL-PLATE SPECIMENS

Total Reduction Ratio	Boride Mesh Size	Test Temperature, F	Ultimate Tensile Strength, psi
5:1	-200 +270	Room	17,100
5:1	-200 +270	600	14,500
5:1	-200 +270	600	14,600
6:1	-200 +270	600	13,700
6:1	-200 +270	600	13,300
6.7:1	-200 +325	600	11,100
6.7:1	-200 +325	600	11,500
7:1	-200 +270	600	12,200
7:1	-200 +270	600	12,700
8:1	-200 +270	600	12,700
8:1	-200 +270	600	13,100
9:1	-200 +270	600	13,100
9:1	-200 +270	600	13,200
10:1	-200 +270	600	11,500
10:1	-200 +270	600	10,700

A series of tests is also 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. Sintered specimens containing varying amounts of carbon, niobium, and silicon have been examined. Similar specimens are being roll clad at 2000 F and will be evaluated after rolling. The effect of niobium additions was determined in both an elemental Type 347 matrix and an iron matrix. In both cases a slight amount of a secondary phase appeared in the grain boundaries adjacent to ZrB_2 particles when the niobium content was 2 w/o. Chemical analyses also showed a higher boron loss from these specimens. However, oxidation of the ZrB_2 particles decreases even to the 5 w/o niobium level. Silicon additions up to 3 w/o also reduce the oxidation of ZrB_2 particles, but chemical analyses show a higher boron loss with higher silicon contents. The results concerning carbon contents from 0.1 to 1 w/o of the matrix are inconclusive. However, it would not be expected that carbon in the matrix would react with ZrB_2, but if carbon is present in the ZrB_2, it probably exists as B_4C, which will react with the matrix.

A high-purity ZrB_2 containing 19.1 w/o boron, less than 0.1 w/o carbon, and a negligible amount of other impurities has been received and will be used in evaluation tests. Unfortunately the powder is not available in larger mesh sizes than 325, so methods of consolidating the powder to larger sizes are being investigated.

Chemical analyses of heat treated boron-stainless steel alloy foil indicates that heavy boron losses can occur during hot rolling of thin sheets. A loss of only 0.01 w/o boron was reported during melting and fabrication of a cast alloy containing 0.461 w/o boron to 0.010-in. foil, but the reduction below approximately 1/4 in. in thickness was almost entirely by cold rolling. However, when the 0.010-in. foil was heat treated for 2 hr at 2200 F heavy losses of boron occurred in both air and hydrogen. The boron content dropped to 0.245 w/o for the specimen annealed in air and 0.128 w/o for the specimen annealed in hydrogen. The reason for the heavier loss in hydrogen is not clear, but may be due to a shorter diffusion path since no oxide scale formed as in the air-treated specimen. Other samples of the foil are being treated in air, hydrogen, and vacuum in order to gain additional information. The tests to date suggest that, if boron is alloyed with the fuel core matrix, a significant loss due to diffusion through the thin cladding can take place during the latter stages of roll cladding when the cladding is relatively thin.

Fabrication of Irradiation Specimens

The fabrication and the irradiation examination of all of the specimens for the three eight-specimen MTR capsules has been completed and the specimens have been encapsulated and delivered to the MTR. The specimens for the first capsule in the revised irradiation program in the ETR have also been encapsulated. Specimens for six additional capsules scheduled for irradiation in the ETR have been scheduled and are in various stages of completion. All SM-2 irradiation specimens presently scheduled should be completed within the next 2 months. The revised list of specimens is listed in Table P-2.

Development of Control Materials

Subscale control elements containing cores of 33 w/o Eu_2O_3 dispersed in an elemental stainless matrix are being fabricated for use in corrosion tests at Alco Products. These elements are 4 in. long and 1 in. wide, and have cores 0.090 in. thick. Both clad defected plates and bare cores are being prepared. The clad plates will contain a silicon-free stainless barrier foil approximately 0.0005 in. thick between the core and cladding.

Europium oxide of the desired particle size (minus 100 plus 200 mesh) is being prepared by pressing Eu_2O_3 minus 325-mesh particles into a compact containing

TABLE P-2. IRRADIATION SPECIMENS FOR STUDIES OF SM-2 CORE AND VESSEL MATERIALS

Revised June 25, 1959

Material	Fuel	Poison	Stainless Matrix Type	Number of Specimens: MTR Average Burnup	ETR High Burnup	ETR Average Burnup	Remarks
1	Geneva oxide, 26 w/o	B_4C, 0.139 w/o	Prealloyed 302B	2	2	2	SM-1 loading, 0.030-in. core
2	Geneva oxide, 28 w/o	B_4C, 0.27 w/o	Prealloyed 347	2	--	--	SM-2 loading
3	Geneva oxide, 28 w/o	None	Prealloyed 347	2	--	2	--
4	Spherical oxide, 28 w/o	None	Prealloyed 347	2	2	2	--
5	Geneva oxide, 28 w/o	Natural ZrB_2	Prealloyed 347	2	2	2	--
6	Spherical oxide, 28 w/o	Natural ZrB_2	Prealloyed 347	2	2	2	--
7	Spherical oxide, 28 w/o	Natural ZrB_2	Prealloyed 347	2	--	2	One-half SM-2 boron loading
8	Spherical oxide, 28 w/o	Natural ZrB_2	Prealloyed 347	2	--	2	Eu_2O_3 suppressor attached
9	Geneva oxide, 28 w/o	Natural NbB_2	Prealloyed 347	2	2	2	--
10	Deleted						
11	Deleted						
12	Spherical oxide, 28 w/o	Natural ZrB_2	Prealloyed 347	--	--	2	Green compact, hot rolled directly
13	Spherical oxide, 28 w/o	Natural NbB_2	Prealloyed 347	2	2	2	--
14	Spherical oxide, 28 w/o	Enriched boron-10-stainless alloy	Prealloyed 347	2	--	2	Boron-10 in stainless picture frame
15	Deleted						
16	Uranium nitride	Natural ZrB_2	Prealloyed 347	--	2	--	UN equivalent to 40 w/o UO_2
17	Geneva oxide, 40 w/o	Natural ZrB_2	Prealloyed or elemental	--	2	--	--
18	Spherical oxide, 40 w/o	Natural ZrB_2	Prealloyed or elemental	--	2	--	--
			Total specimen spaces	22	18	22	
			Total capsules	3	3	4	

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. Additional work on the preparation of dense particles of Eu_2O_3 of various shapes and sizes is being initiated, but only the material described above will be used for the corrosion specimens.

Core compacts are 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 for 1 hr, pressing at 15 tsi, presintering at 1600 F, and sintering at 2300 F for 2 hr. A roll-cladding temperature of 2200 F will be used.

Encapsulation Studies

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

During July, three noninstrumented irradiation capsules (BMI-32-1, BMI-32-2, and BMI-32-3) were assembled and shipped to the MTR. According to the present schedule, BMI-32-1 will be inserted in the MTR core during Cycle 126 shutdown (August 10, 1959); BMI-32-2 and BMI-32-3 are scheduled for insertion during Cycle 127 shutdown. It may be, however, that all three capsules will be loaded during Cycle 126 shutdown. Table P-3 presents a list of specimens present in these capsules; as mentioned in previous reports, the capsule design is based on a specimen-surface temperature of 650 F in an unperturbed thermal-neutron flux of 5 x 10^{14} nv.

The assembly of one heatered and thermocoupled capsule (designated as BMI-32-4) is nearly complete. This capsule is to be irradiated in a beryllium-reflector position at the ETR. The capsule is the first of seven instrumented capsules that will be irradiated at the ETR. All seven capsules are to be prepared during the remainder of the calendar year.

The construction of nuclear-mock-up capsules to evaluate flux-perturbation characteristics of the MTR and ETR capsules was delayed during July, but emphasis on this phase of the program will be renewed during August. As explained in BMI-1357, experimental perturbation data are desirable in view of a lack of agreement between values obtained by P-3 type calculation and by calculations based on Brad Lewis empirical factors.

TABLE P-3. FUELED CORE COMPOSITIONS FOR SPECIMENS (a) CONTAINED BY CAPSULES BMI-32-1, BMI-32-2, AND BMI-32-3 TO BE IRRADIATED IN MTR CORE POSITIONS

Specimen Location in Capsule, Starting From Top Position	UO_2 in Type 347 Stainless Steel Dispersion, w/o	Poison Type	Nominal Boron Content, w/o
	Capsule BMI-32-1		
1	26(b)	ZrB_2	0.095
2	26	B_4C	0.192
3	26	ZrB_2	0.199
4	26	NbB_2	0.209
5	26	None	--
6	26	None	--
7	26(c)	ZrB_2	0.240
	Capsule BMI-32-2		
1	26	NbB_2	0.204
2	26	ZrB_2	0.095
3	26	Boron-10 (in cladding)	0.022
4	24.2	B_4C	0.021
5	26	None	--
6	26	ZrB_2	0.192
7	26(c)	ZrB_2	0.240
	Capsule BMI-32-3		
1	26	Boron-10 (in cladding)	0.022
2	26	B_4C	0.192
3	26	None	--
4	26	NbB_2	0.209
5	26	NbB_2	0.202
6	26	ZrB_2	0.199
7	24.2	B_4C	0.021
8	26	ZrB_2	0.192

(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.

(b) Uranium-235 contents are 0.514 g for the 26 w/o cores and 0.476 g for the 24.2 w/o specimens.

(c) These are double-length specimens in which a UO_2-fueled core and a Eu_2O_3 (suppressor material) core are both present; each of these cores is of regular size and, in the fabrication process, are laid end-to-end and clad.

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

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 BRR.

The six fuel pins from Capsule BMI-27-2 (three solid UO_2 and three annularly loaded UO_2 cores) have been examined visually. Four of these pins failed during irradiation while two appear undamaged. Extensive examination is now in progress.

During the past month neutron-flux and power-distribution studies were conducted, and reactivity worths of mock-up control blades were measured in the ML-1 critical assembly. Also, a GCRE-IB fuel element was introduced to the center of the ML-1 core for neutron-flux mappings within and around the I-B element.

Encapsulation Studies

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

Irradiation of Clad UN and UO_2 Dispersions

Capsule BMI-28-1 was returned to the Battelle Hot-Cell Facility during July and is awaiting opening. This capsule was irradiated at the MTR for about three and one-half reactor cycles. The one UO_2- and three UN-bearing clad coupons it contains were maintained at approximately 1650 F during the exposure.

Irradiation of Clad Pin-Type Specimens Containing Dense UO_2

Capsule BMI-27-2 was received at the Battelle Hot-Cell Facility on July 20 for postirradiation analyses. It was found, upon opening the system, that four of the six specimens (Inconel-clad UO_2-fueled pins) present were severely damaged. As mentioned in previous reports, temperatures of some of the specimens surfaces probably reached 1700 F during the irradiation (five cycles at MTR), but this level did not appear to be exceeded, except perhaps during brief periods at reactor startup. However, in view of the condition of the specimens, the irradiation is being reviewed to establish the operating-temperature history in more detail than previously has been done.

The status of Capsule BMI-27-1, being irradiated in MTR Position A-7-NW, remained unchanged during July. As before, bottom-specimen temperatures, estimated from readings of thermocouples located in the vicinity of the top two specimens (about 500 F), are in the 1400 to 1600 F range. It is estimated that the bottom-specimen fission-burnup level has reached about 6 a/o. As things now stand, it is unlikely that this capsule will be repositioned in an attempt to achieve a more uniform axial temperature pattern. In fact, consideration is presently being given to discharging the capsule during August in view of the damage suffered by specimens from BMI-27-2 (which, according to best estimates, accumulated a maximum fission burnup of about 4 a/o).

Irradiation of Specimens Containing MCW Spherical UO_2 Dispersed in Stainless Steel

Two irradiation capsules, designated as BMI-33-1 and BMI-33-2, were shipped to the MTR on July 22 with the request that they be inserted during Cycle 127 shutdown (August 31, 1959). Each of these capsules contains four stainless steel-clad specimens with fueled cores consisting of a dispersion of 30 w/o highly enriched UO_2 in Type 318 stainless steel; in half of the specimens, ORNL hydrothermal UO_2 is present and in the other half, MCW spherical UO_2 is present. The objective of the experiments is to compare the irradiation stability of the dispersions containing the two different types of UO_2. Irradiation parameters include a specimen-surface temperature of 1650 F and a fission burnup of 6 and 9 a/o, respectively, for the two capsules. In the flux requested, approximately 1×10^{14} nv, 6 a/o burnup will be achieved in two normal MTR cycles; 9 a/o burnup will require three normal cycles.

As mentioned in BMI-1357, the capsule design is similar to those previously employed in Battelle irradiation programs. It is based on a double-wall configuration in which the four vertically aligned specimens are immersed in NaK and thermal contact between the inner and outer capsule shells is provided by fins. Thermocouples and dosimeters are located adjacent to the specimens.

Irradiation of Specimens Containing UO_2 in Graphite

Capsule BMI-29-1 is at NTRS awaiting insertion in the MTR. As mentioned in previous reports, this capsule contains six metal-clad 1/4-in.-OD pins containing 8 w/o UO_2 (highly enriched) in graphite; cladding materials are Hastelloy X, Inconel 702, and Carpenter 20 Cb. The target specimen-surface temperature is 1750 F; auxiliary heaters are present to assist in maintaining this level during the exposure.

Q-3

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

Preparations for elevated-temperature irradiation-damage studies of SiC-coated graphite-UO_2 (8 w/o highly enriched UO_2) bodies continued during July. This involves the irradiation for approximately 5500 hr of three similar capsules in a core-face position of the Battelle Research Reactor. Each capsule will contain a specimen assembly consisting of one unfueled and two fueled disks (1-3/8 in. in OD by 1 in. long) sealed in a metal can. The evaluation of irradiation damage to the fueled bodies and the determination of the corrosive attack of GCRE reference gas on the specimen-can materials (now Inconel 702 and Hastelloy X) at a nominal temperature at 1700 F are two specific objectives of the program.

Thermal-mock-up studies are under way (1) to assess problems involved in capsule-temperature control (which will be accomplished by regulating the composition of the gas mixture present in the thin annulus between the water-contacting outer shell and the canned specimen assembly), and (2) to ascertain the importance of axial heat flow from the two fueled disks to the central nonfueled disk in the individual assemblies. In the mock-up, sheathed resistance-wire heaters provide the heat source. Heat is transferred across a 40-mil annulus connected to argon and helium cylinders via individual lines. Gas composition is regulated by a delivery- and bleed-valve system actuated at appropriate intervals by a control thermocouple located adjacent to the heater coils.

In experiments thus far completed, the pressure of the heat-transfer gas mixture was maintained at 110 psi since consideration was initially given to pressurization in the final in-pile systems. The best temperature control achieved was ± 25 F at a control level of 1000 F. Atmospheric-pressure experiments were next scheduled, but, before these could be initiated, the system heaters failed and, at present, necessary repairs are being made; in addition, the system design is being modified to achieve increased flexibility. When tests are resumed, control-temperature levels will be in the 1500 F range with atmospheric-pressure gas-mixture conditions.

As described in BMI-1357, GCRE reference gas pressurized to 400 psi will flow slowly during the irradiations through small tubes welded into the canned-disk assemblies. Flow-control problems presented by this system have been evaluated by introducing cylinder nitrogen into a small furnace-heated (to 1700 F) U-tube. Valving consisted of (1) a precision cylinder-to-tube regulator, (2) a second regulator for throttling, and (3) a manually adjustable needle valve at the exit. Pressure control achieved with this arrangement was quite acceptable inasmuch as a flow rate of 0.067 lb of gas per hr produced a pressure drop of only 6 psi from a 400-psi static condition. This flow rate is the maximum anticipated for the individual capsule systems during their in-pile exposure.

As an adjunct to these corrosion-gas experiments, investigation was made of thermocouple placement within the heated tube and its effect on the accuracy of the temperatures monitored. It was found that couples held against the inside wall by a clip device were more responsive to wall-temperature variations than couples suspended in the gas stream. Also, the former thermocouples were less affected by variations in gas-flow rate. With the wall temperature held at 1700 F, they indicated a temperature of about 1650 F when the flow rate was 0.080 lb per hr. These results justify the use of clip-positioned thermocouples in the final capsule design.

The final experimental activity now under way involves nuclear-mock-up tests to assess capsule flux-perturbation characteristics. Three complete mock-up assemblies and the necessary holding frame were fabricated and exposed at the BRR during July. In the upper and lower units, the fuel is simulated by an aluminum braze alloy; in the central unit, graphite specimens are fueled with the proper amount of highly enriched UO_2. This series of tests, along with the attendant dosimeter analyses, will be completed during early August.

During July, detailed analyses were made of the thermal resistance between the coated graphite bodies and their containing cans, taking into account clearances developed at operating temperatures from unequal thermal expansions, dilution of the original helium cover gas by fission gas release and consequent reduction in heat-transfer capacity, and eccentricity effects. A summary of typical results is presented in Table Q-1. It may be seen that variations in the temperature drop from the fuel to the can can be sizable unless initial fits are closely controlled. A large variation here, of course, will reduce the degree of specimen temperature control offered by variable gas-mixture composition. Fortunately, however, information from AGN, who will supply the test-specimen assemblies, indicates that an initial press-fit assembly can be achieved without undue difficulty.

Effects of Irradiation

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

The irradiation in Capsule BMI-27-2 of six Inconel-clad specimens containing solid and annularly loaded UO_2 has been completed. These specimens were irradiated in contact with NaK to approximate burnups of 4 per cent of the uranium-235. Preliminary examinations of the thermal history of these specimens as obtained from thermocouples during irradiation indicated that the surface temperatures ranged from approximately 1100 to 1700 F. The postirradiation examination planned for each specimen included sampling for fission gas released, visual inspection, making of stereomacrographs, density determinations, dimensional measurements, burnup determinations by isotopic analysis, and metallographic examinations. In addition, it was planned to remove the solid pellets from four specimens and obtain stereomacrographs, densities, and dimensions of each pellet. The irradiated capsule has been received at Battelle and the recovery and examination of the specimens have been initiated.

The capsule was received and opened at the Battelle Hot Cell Facility during July, 1959. Four of the six specimens contained in the capsule had failed in such a manner that measurements and densities will be impossible to obtain. Metallographic examinations and burnup analyses on these specimens will be conducted. Two specimens from the central section of the capsule, both loaded with solid UO_2, appear to be in good condition and will receive a complete examination. Of the four specimens which

TABLE Q-1. CALCULATED SPECIMEN-TO-CAN SURFACE TEMPERATURE GRADIENTS FOR A SELECTED RANGE OF PERTINENT VARIABLES(a)

Diametral Clearance at Room Temperature, mils	Amount of Fission Gas Release Into Space Between the Specimen and Encapsulating Can, per cent	Eccentricity at Temperature, mils	Temperature Gradients, Specimen Surface to Can, F
-0.6 (interference fit)	0	0	165
	0	2	150(b)
	0	4	90(b)
	50	0	280
+2.0	0	0	205
	0	2	193(b)
	0	4	150(b)
	50	0	345

(a) Assumed specimen conditions include diameter (room temperature) = 1.3754 in., surface temperature = 1604 F, heat-generation rate = 3000 Btu/(hr)(in.), and coefficient of expansion = 3.3×10^{-6} per F; the assumed coefficient of thermal expansion of the can material is 9.2×10^{-6} per F (for Hastelloy X).

(b) Isothermal surfaces were assumed for the nonconcentric cases.

failed, the top specimen and the bottom specimen containing solid UO_2 appear to have failed because of melting or alloying of the Inconel cladding. The second specimen from the top, containing annularly loaded UO_2, also appears to have failed in this manner. The remaining annularly loaded specimen, located next to the bottom specimen, was ruptured but shows no evidence of melting.

Preliminary examinations have not indicated the cause of failure of the specimens. A complete analysis of the capsule design, the irradiation history, and the appearance of the specimens is being made in an attempt to discover the reason for the failures.

GCRE Critical-Assembly Experiments

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

Experiments are continuing with the ML-1 critical-assembly mock-up. During the past month neutron-flux and power-distribution studies were conducted and reactivity worths of mock-up control blades were measured.

The ML-1 reactor is designed to produce a nearly uniform radial power distribution. To investigate power uniformity 1/4-in.-wide aluminum strips (catcher foils) were wrapped on each of the four fuel cylinders at a fixed axial position in all fuel elements of a symmetry sector of the core. The maximum deviation from core-averaged power was less than 18 per cent. The percentage contribution of the individual cylinders to fuel-element power varied little throughout the core. Table Q-2 shows the maximum and minimum power contributions for each of the four cylinders.

TABLE Q-2. PER CENT OF TOTAL FUEL-ELEMENT POWER SUPPLIED BY INDIVIDUAL FUEL CYLINDERS

Fuel Cylinder	Power Contribution, per cent Maximum	Power Contribution, per cent Minimum
Inner	12.00	10.00
Second	18.10	15.90
Third	27.70	25.15
Outer	48.05	43.20

Neutron-flux data throughout the core were also obtained and are being analyzed.

Special studies using a GCRE-IB fuel element (19-pin uranium dioxide-pellet-fueled assembly) were conducted to investigate the neutron-flux pattern within and around the IB element. For these studies the IB element replaced a standard GCRE-I multicylinder element at the center of the core. The exchange represented no change in reactivity. The flux data are being analyzed.

Shutdown-reactivity values are being measured for mock-up scissor-action control blades. The first set of measurements were made with stainless steel, approximately 3 in. wide by 12 in. long and tapering in thickness from 5/8 in. to 1/8 in. at the tip (core end). The reactivity measurements included five angles of separation between the two blades: zero, 6-1/3, 30, 60, and 110-1/2 deg. It was not possible to open the blades fully (180 deg) because other components of the critical-assembly mock-up interferred. The data are being analyzed.

In a second set of experiments the stainless steel blades were covered with 20-mil-thick cadmium sheet. This latter study is still in progress.

During the coming month the control-blade studies will be concluded. In addition, experiments concerning the reactivity worth of a production GCRE-I fuel element in the ML-I critical assembly will be conducted. These studies will attempt to detect variations in construction or in fuel loading of the production element. Analysis of data from all experiments will continue during the next month.

IN-PILE-LOOP PROGRAM

G. A. Francis

During the period covered by this report activity continued on both the Battelle Research Reactor and Engineering Test Reactor in-pile recirculating gas-loop programs. At the BRR, irradiation of a fuel specimen was completed and a flux run was made. At the ETR, preparation for insertion of the in-reactor loop piping was continued. The activities associated with the two loop programs are described in the following sections.

BRR Loop Program

S. J. Basham and W. H. Goldthwaite

Irradiation of the IB-1αT specimen was started during June. The specimen consists of 19 Inconel-clad UO_2 fuel pins in the form of a fuel subassembly. The irradiation was terminated on July 18 because of high fission-gas activity at the exhaust stack.

The element had been irradiated for a total of 165 hr at that time. The element power transmitted to the flowing gas (at an average test-section temperature of 650 F) was approximately 33 kw.

At the time of test termination, 267 thermal cycles had been accumulated. They were as follows:

Q-8

Controlled thermal cycles	248
Slow thermal cycles	1
Loop shutdowns	3
Loop-caused reactor scrams	6
Reactor scrams	9

During the run, eight of the fuel-assembly thermocouples failed. All but one of these failures occurred during thermal cycling of the element. The controlled cycles were between 1000 and 1600 F at the hottest measured fuel-specimen surface.

Gas samples which were taken from the primary-coolant system throughout the run showed gaseous fission products to be present. The amount increased during the irradiation and showed either surface contamination or a cladding defect. The data are being analyzed at this time and will be reported next month.

After the specimen was removed from the loop, a nuclear mock-up of the unit was inserted in the loop for flux mapping. The results of the flux study will be reported next month.

A survey of the loop for activity which might have resulted from the fission gas release showed no abnormal amounts. It appears that the unit should still be serviceable by use of previously established techniques.

Plans call for the start of irradiation of the IB-1βT during August. The β-unit is similar to the α-unit with respect to general configuration and materials but features stepped and finned heat-transfer surfaces.

ETR Loop Program

J. V. Baum and E. O. Fromm

Activities at the reactor site have continued with the cold check of the out-of-pile installation and preparation for insertion of the in-pile tube.

The problems associated with the operation of centrifugal blowers in parallel were studied both analytically and experimentally. Operation of the system with the individual blower outlet valves throttled to give a 20-psi differential between headers has shown that stable operation can be achieved at the loop design flow. Since all anticipated loop operation is at flow rates far below loop design and this might introduce new stability problems, single-blower operation has been requested for initiation after the first cycle of irradiation of the first test specimen. A change of the loop control system to a flow basis has been requested for the same time.

Q-9 and Q-10

The checkout of removal equipment has started but is not complete. The initial plan has been altered to eliminate assembly of the complete apparatus on top of the reactor. Scheduling has necessitated this action.

At present, the out-of-pile portion of the loop has been released to the reactor operating staff. Operators are becoming familiar with the system by training on all shifts.

Plans call for the reactor to be down during the first two weeks of August. The Battelle in-pile piping should be installed during the early part of the shutdown. The hot check will be performed with a dummy specimen in place during the next reactor cycle.

RWD/CRT:all