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PROGRESS RELATING TO CIVILIAN APPLICATIONS
DURING MARCH, 1961

by

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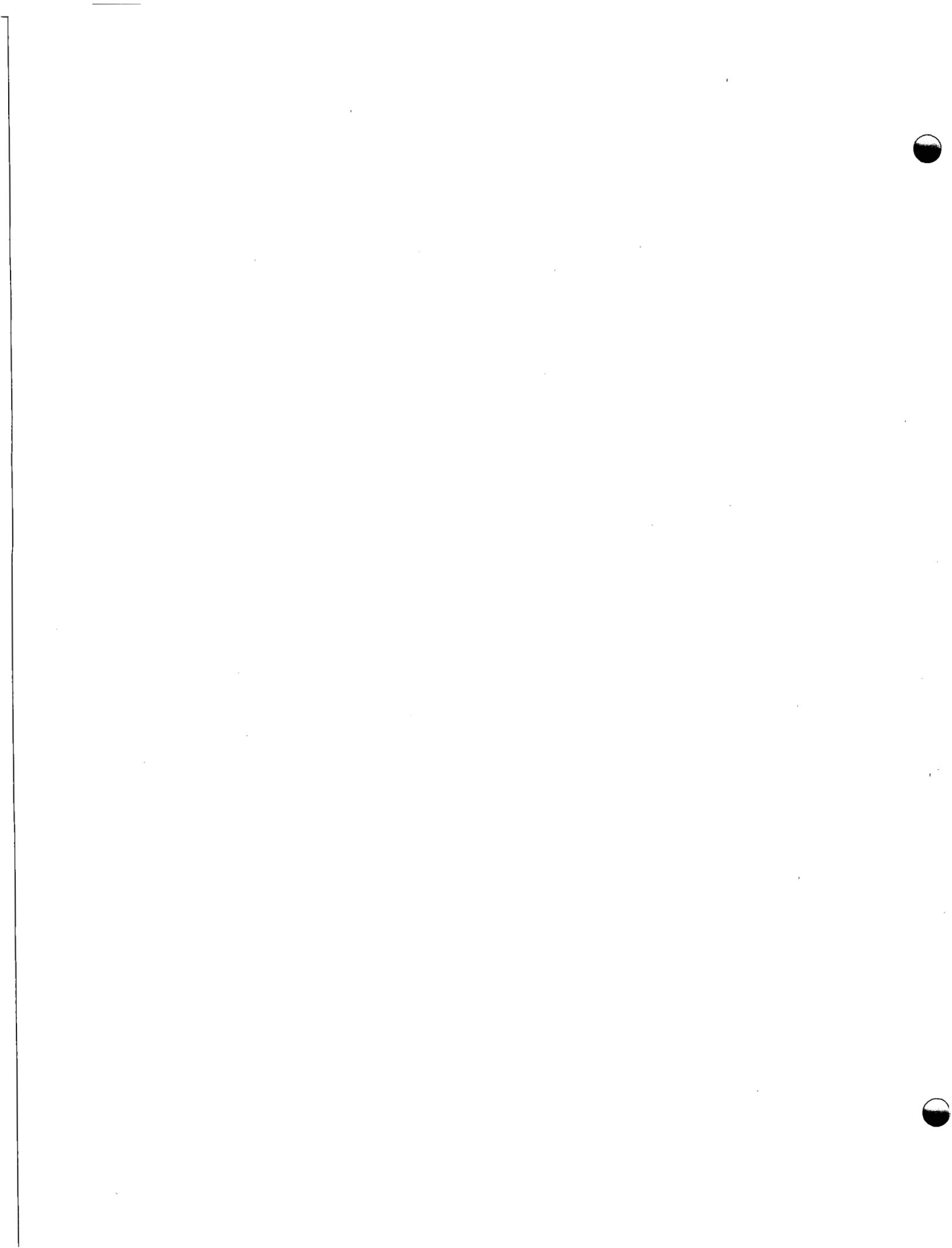
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REPORTS RELATING TO CIVILIAN APPLICATIONS
ISSUED DURING MARCH, 1961

- BMI-1489 (Rev.) "Progress Relating to Civilian Applications During December, 1960", by Russell W. Dayton and Clyde R. Tipton, Jr.
- BMI-1500 "Development of Container Materials for LAMPRE Applications", by David C. Drennen, Merritt E. Langston, Charles J. Slunder, Joseph G. Dunleavy, and A. M. Hall.
- BMI-1501 "ML-1-1B Critical-Assembly Studies", by Joseph W. Ray, Richard G. Matthews, William S. Hogan, David A. Dingee, and Joel W. Chastain, Jr.
- BMI-1502 "Thermal Diffusion in a Solid Solution of Hydrogen in Beta Zirconium", by John W. Droege.
- BMI-1503 "Numerical Solution of Reactor Stress Problems", by Robert F. Redmond, Lewis E. Hulbert, and Richard W. Clark.
- BMI-1504 "Progress Relating to Civilian Applications During February, 1961", by Russell W. Dayton and Clyde R. Tipton, Jr.



REACTOR MATERIALS AND COMPONENTS (AEC-DRD)

R. F. Dickerson

The general objective of the research tasks reported in this section is to provide basic structural material and component support to the over-all Atomic Energy Commission reactor development program. These studies are concerned with the effect of ultrahigh pressures and high temperatures on the structure of materials, the effect of irradiation on the mechanical properties of Type 347 stainless steel and Zircaloy-2, the development of high-strength niobium-base alloys for use in the EBR, the friction and wear of sliding surfaces in sodium environments, the determination of oxygen in sodium at concentrations below 10 ppm, and the development of a thermal-neutron flux-sensing device.

High-Pressure High-Temperature Solid-State Studies

W. B. Wilson and C. M. Schwartz

Objective

To determine if ultrahigh pressure can be utilized to produce new materials.

Program Approach

The effects of pressure on reactions of uranium oxide with other oxides are currently under investigation. Emphasis, at present, is being placed on the study of reactions between U_3O_8 and Al_2O_3 .

Progress Reported Previously

The development and calibration of an apparatus capable of pressures above 150,000 atm and temperatures above 2000 C have been reported in BMI-1328. The apparatus has been employed to study the effects of pressure on the uranium-oxygen system. The results have been noted in previous reports.

The U_3O_8 - Al_2O_3 reaction is being studied. Two new compounds, "X" and "Y", are produced in this system under the combined influence of high pressure and temperature. The "Y" phase is a high-temperature product, and little or no detailed study of this material has been undertaken to date. Recent work has been directed toward obtaining single crystals of the "X" phase to confirm earlier indexing of the powder patterns. As reported last month (BMI-1504), while conducting this work, a single crystal of the gamma high-pressure form of U_3O_8 was obtained. The study of the "X" phase crystal was deferred to permit analyses of this U_3O_8 crystal.

Work This Period

The single crystal of gamma U_3O_8 was mounted and oriented for examination on the Weissenberg X-ray camera. Rotation and Weissenberg patterns were recorded. This analysis has shown that gamma U_3O_8 is monoclinic with $a_0 = 5.66$ A, $b_0 = 5.05$ A, and $c_0 = 12.40$ A with $\beta = 98$ deg. The values given are not precision parameters but represent a first attempt at refinement for reflections computed from the powder patterns.

A unit-cell volume of 329 A³ was obtained from these values. Since the composition or oxygen/uranium ratio was determined as 2.66 for this phase, it is possible to determine the density by assuming integral values of formula units per unit cell.

This establishes a density of 9.34 g per cm³ for seven uranium atoms per unit cell, and 10.6 g per cm³ for eight atoms per unit cell. Lower and higher values, obtained from other integral multiples, have been discarded since they fall outside the range of reasonable densities based upon U₃O₈ and UO₂, respectively. An experimental density of 9.2 g per cm³ ± 10 per cent was previously reported which was known to be unreliable because of the small amount of material available.

From these data it thus appears that gamma U₃O₈ has either seven atoms of uranium in the unit cell and a density of 9.34 g per cm³ or eight atoms and a density of 10.6 g per cm³. Both seven and eight atoms may be placed in monoclinic unit cells. However, consideration of the X-ray extinctions show that only eight atoms per unit cell are possible to produce the observed diffraction effects. On this basis, only space group C₂^h appears suitable from the standpoint of observed extinctions and cell content. The true density thus appears to be 10.6 g per cm³, suggesting that the earlier experimental value was too low, possibly because of the presence of some normal alpha U₃O₈ of density 8.34 g per cm³.

Plans for Future Work

Work will continue toward determination of a possible trial structure of gamma U₃O₈. Single-crystal work on the "X" phase, the high-pressure reaction product of U₃O₈-Al₂O₃, will be resumed.

Irradiation-Surveillance Program on Type 347 Stainless Steel

W. E. Murr, R. Leiberman, F. R. Shober, and R. F. Dickerson

Objective

To determine the effects of fast-neutron irradiation (neutrons having energies greater than 1 Mev) on the mechanical properties of AISI Type 347 stainless steel.

Program Approach

Subsize tensile, cyclic-strain fatigue, and impact specimens of Type 347 stainless steel are being irradiated in capsules in core positions of the ETR. Evaluation of irradiation-induced changes will be based on results from mechanical tests made after exposures of about 0.5 to greater than 3.0 x 10²² nvt (fast). The information thus obtained will be used to predict the safe operating life of this material.

Progress Reported Previously

The total accumulated fast-flux exposures for capsules currently operating in or removed from the ETR through December 14, 1960 (end of Cycle 33) were reported previously in BMI-1489. The results from Charpy and subsize Izod specimens irradiated to exposures of about 5.7 x 10²¹ nvt and tested at room temperature and -300 F are reported in BMI-1448 (Rev.). A definite loss in impact strength was noted in the irradiated specimens, particularly for those specimens tested at liquid-nitrogen temperatures.

Work This Period

Ten capsules currently located in a core-filler piece in the I-13 position of the ETR have continued to receive fast-neutron exposures. During Cycles 34 and 35, the capsules accumulated 3499 and 1479 megawatt-days of exposure, respectively. At the present time, the capsules are operating in Cycle 36, which is scheduled to terminate on about April 17, 1961. The additional fast-flux exposure to the specimens in I-13 during Cycles 34 and 35 will be reported after dosimeter wires, exposed during these cycles, have been analyzed.

Negotiations have been initiated to obtain sections of the Type 347 stainless steel J-10 pressure tube recently discharged from the ETR. This tube was installed in ETR immediately prior to Cycle 21 (September, 1959) and was removed after Cycle 35 (March, 1961). During this time, the maximum flux positions of this loop received an estimated exposure of about 7.5×10^{21} nvt while operating at a calculated surface temperature of about 600 F. Portions of this tube will be machined into tensile specimens for testing at room and elevated temperatures.

Work aimed at evaluating the $\text{Fe}^{54}(\text{n},\text{p})\text{Mn}^{54}$ reaction for use as a fast-flux monitor is continuing. Recent dosimetry measurements made on nickel and iron wires irradiated in the ETR during Cycle 32 have shown that estimates of fast-neutron flux based on the Mn^{54} reaction are higher than those based on the $\text{Ni}^{58}(\text{n},\text{p})\text{Co}^{58}$ reaction even after the Co^{58} results are corrected for the $\text{Co}^{58}(\text{n},\gamma)\text{Co}^{59}$ reaction. The analysis of iron and nickel dosimeter wires irradiated in three quadrants of the core-filler piece for ETR Cycle 32 are shown in Table A-1. Measurements based on the Mn^{54} analysis were compared by radioassaying the Mn^{54} in the presence of Fe^{59} , and by separation of the Mn^{54} before assay. As shown in Table A-1, the separation procedure gave lower fast-flux values and is considered more accurate than the simultaneous counting method.

TABLE A-1. COMPARISONS OF ESTIMATES OF FAST FLUX IN AN ETR CORE-FILLER PIECE DURING CYCLE 32 OBTAINED FROM NICKEL AND FROM IRON DOSIMETERS

Quadrant in Core-Filler Piece in Which Wires Were Located	Estimated Fast-Neutron (>1 Mev) Flux, nv			
	Based on $\text{Ni}^{58}(\text{n},\text{p})\text{Co}^{58}$ Reaction		Based on $\text{Fe}^{54}(\text{n},\text{p})\text{Mn}^{54}$ Reaction	
	Uncorrected(a)	Corrected(b)	Uncorrected(c)	Corrected(d)
South-4	5.98×10^{13}	1.26×10^{14}	1.74×10^{14}	1.54×10^{14}
East-4	6.90×10^{13}	1.15×10^{14}	2.08×10^{14}	1.71×10^{14}
West-4	4.32×10^{13}	8.84×10^{13}	1.43×10^{14}	Lost

(a) Uncorrected for thermal burnup of Co^{58} .

(b) Corrected for thermal burnup of Co^{58} .

(c) Simultaneous counting of Mn^{54} and Fe^{59} without ion separation.

(d) Ion-exchange separation of Mn^{54} .

The total estimated fast-flux exposure of specimens irradiated in Capsule BMI-24-14 was earlier reported as about 5.7×10^{21} nvt. This capsule, which was continuously irradiated from Cycle 4 through Cycle 28, contained specimens subsequently tested to provide the data given in BMI-1448 (Rev.). Recently obtained flux data from specimens of this capsule have substantiated the previously quoted flux exposure levels. Stainless steel filings from the fractured area of two Charpy specimens were analyzed for Mn^{54} by ion-exchange separation of the radioisotope. Average fast-flux values of 2.4 and 2.6×10^{14} nv were obtained on the two specimens. The integrated fast-neutron flux to which the specimens were exposed was then determined by multiplying the effective irradiation time by average fast flux. Values obtained for the two specimens examined were 5.5 and 6.0×10^{21} nvt, as compared with 5.7×10^{21} nvt determined by nickel N_L dosimetry.

Plans for Future Work

Studies of fast-flux determination techniques will continue. Future work will include a comparison of fast-neutron flux measurements using nickel and cadmium-shielded nickel, iron, and copper. The cadmium shield surrounding the nickel will eliminate the loss of Co^{58} through the (n, γ) reaction. The copper reaction to receive cursory examination for possible fast-flux dosimetry is the $\text{Cu}^{63}(n, \alpha)\text{Co}^{60}$ reaction. Experiments will be run on simultaneously exposed wires in the ETR beginning with Cycle 37.

It is expected that the requested sections of the J-10 loop will be received at Battelle during April. Tensile specimens will be machined from the material, and tensile tests will be conducted at room temperature and 600 F. Two sections of the loop have been requested, one section which operated in contact with the ETR process water at about 120 F and a second, or inner, section which operated near 600 F. In addition to the tensile tests, annealing studies, metallography, and hardness studies are planned on portions of both sections.

The ten capsules in the ETR will continue to receive exposure. It is planned to discharge one capsule for examination after a fast-neutron exposure of about 5.7×10^{21} nvt, while the remainder of the capsules will receive exposures over a range from 1.0 to 3.0×10^{22} nvt.

Development of High-Strength Niobium-Base Alloys

J. A. De Mastry, F. R. Shober, and R. F. Dickerson

Objective

To develop high-strength niobium-base alloys compatible with sodium and uranium-plutonium fuels for cladding applications at 800 to 1000 C in the EBR II.

Program Approach

Additions of 2.37 w/o chromium, 3.34 w/o zirconium, and 5.21 w/o vanadium were made to niobium. The effects of these additions on the hot and cold fabricability, the tensile and creep strength at 650, 800, and 1000 C, and the recrystallization temperature are being determined.

Progress Reported Previously

Ingots weighing 4 to 5 lb of each composition were consumably arc-melted, forged to 0.50-in.-thick slabs at 540 C, and then rolled at room temperature to 0.030-in.-thick sheet.

The results of tensile testing were reported in BMI-1469. Preliminary creep data were reported in BMI-1480. All alloys are superior to Type 304 stainless steel in tensile and creep properties at the temperatures considered. Chromium appears to be the most potent strengthener on a weight percentage basis. Room-temperature hardness measurements of all three alloys after a 1-hr anneal at 1038 C indicated that stress relief had occurred. Higher annealing temperatures, >1038 C, did not reduce these hardnesses appreciably.

Work This Period

The stresses to produce deformation rates of 0.001, 0.01, and 0.1 per cent per hr at 650, 800, and 1000 C are shown in Table A-2. Most of the test specimens behaved in a normal manner. However, the niobium-3.34 w/o zirconium alloy showed different creep characteristics than the other alloys tested at 650 C. At 650 C, the alloy either failed on loading or continued in test indefinitely at a very low creep rate.

The difference in stress between failing on loading and continuing indefinitely is very small, and essentially the same stress could produce either. In the case of the short-time failure, the load applied was approximately equal to the ultimate tensile strength of the alloy. In the specimens which did not fail immediately, large initial elongations (3 to 10 per cent) were noted. At 650 C, the amount of strain hardening introduced by the amount of deformation in the zirconium alloy is apparently sufficient to produce appreciable strengthening. The test temperature of 650 C is well below the recrystallization temperature for this alloy, and a portion of this added strength could be expected to be retained. Therefore, the stress-versus-minimum creep rate curve is very flat. This results in practically no difference in the three stresses that produce creep rates of 0.001, 0.01, and 0.1 per cent per hr. At temperatures approaching the recrystallization temperature, the effect of strain hardening would be expected to be annealed out quite rapidly. At the higher test temperature, 800 and 1000 C, where lower initial deformations (<0.5 per cent) were noted, the difference in stresses to produce the previously indicated creep rates are greater in magnitude. The data in Table II-2 show that the niobium-5.21 w/o vanadium alloy possesses the highest creep strength at 650, 800, and 1000 C.

TABLE A-2. STRESSES TO PRODUCE DEFORMATION RATES OF 0.001, 0.01, AND 0.1 PER CENT PER HR AT 650, 800, AND 1000 C IN THE BINARY NIOBIUM ALLOYS

Test Temperature, C	Alloy Composition (Balance Niobium), w/o	Stress to Produce Creep Rates Shown, psi		
		0.001 Per Cent per Hr	0.01 Per Cent per Hr	0.1 Per Cent per Hr
650	2.37 Cr	47,800	53,200	58,700
	3.34 Zr	41,400	41,800	42,300
	5.21 V	52,700	58,800	64,700
800	2.37 Cr	35,000	37,800	40,600
	3.34 Zr	31,300	40,200	42,200
	5.21 V	40,000	42,500	45,000
1000	2.37 Cr	10,800	17,000	23,200
	3.34 Zr	9,000	15,300	20,800
	5.21 V	15,500	20,300	25,000

Specimens of all three alloys which were cold worked 90 per cent (reduction in thickness) and annealed for 1 hr were completely recrystallized between 1092 and 1149 C. The degree of recrystallization was determined metallographically.

Plans for Future Work

Work on this project has been concluded.

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

F. R. Shober, P. B. Shumaker, and A. P. Young

Objective

To compare the creep resistance of Zircaloy-2 at 650 F in a fast flux of 1×10^{14} n/v with its creep resistance at 650 F in the absence of a fast flux and to study strain

aging of Zircaloy-2 to elucidate the extent to which it may influence creep strength at 650 F.

Program Approach

In-reactor and out-of-reactor creep experiments are planned in which total deformations of sheet specimens in 1000-hr creep tests will be compared. Total deformation will also be indicated during the test by a differential transformer, the output of which will be recorded. The effects of stress under these creep conditions will be investigated by incorporating three different widths in the gage section of the test specimens. The in-reactor portion of these tests is to be conducted at the WTR and the out-of-reactor part at Battelle. Evidences of strain aging are being investigated by means of short-time tensile tests and deformation studies performed at room and elevated temperatures.

Progress Reported Previously

An instrumented capsule, described in BMI-1430, has been designed. Certain modifications have been made with regard to the annulus between the cast aluminum blocks containing the electrical heaters and the capsule wall. The MgO used initially has been replaced by 15 equally spaced vertical copper tubes to increase the thermal conductivity of this section of the capsule.

Work This Period

Preparations for the first in-pile experiment at the WTR were continued. Replacement of the loading bellows which failed during assembly of the first irradiation capsule was completed early in March. However, the failure (in the lead tube) of two electric-heater circuits after subsequent reassembly forced postponement of the scheduled March insertion into the WTR. The capsule has been completely reassembled; final checkouts of all circuitry are now in progress. Arrangements have been made to initiate the irradiation in the WTR with Cycle 13, which is scheduled to start about April 25.

Additional tensile tests have been made at room temperature and at 400, 450, 550, and 650 F as part of the strain-aging experiments. Results obtained are inconclusive. They do indicate, however, that the yield-point phenomenon can be influenced by prior heat treatment and rate of straining. Quenching from the beta phase (1850 F) after holding there for 15 min and then tensile testing at 650 F produced a more pronounced yield point than shown by material vacuum annealed at 1380 F and furnace cooled. It appears that the yield point is a function of the amount of alloying elements retained in solution at the test temperature.

A refined preparation technique has yielded a much smoother surface on zirconium for electron-microscopy examination. A similar technique is being tried on Zircaloy-2 for the same purpose.

Plans for Future Work

Assembly of the control creep experiment (out-of-pile capsule) will begin early in April. It is anticipated that this experiment can begin early in May. On-site surveillance of the in-pile experiment is planned, and efforts will be made to return the irradiated capsule to the BMI Hot-Cell Facility by mid-June. Tensile tests are planned for heavily cold-worked Zircaloy-2 and for other heat-treated conditions.

Friction and Wear of Sliding Surfaces in Sodium Environments

W. A. Glaeser and J. W. Kissel

Objective

To develop general principles applicable to selection of materials and design of mechanical parts involving sliding surfaces in high-temperature molten sodium.

Program Approach

The friction and wear behavior of various materials resistant to corrosion in liquid sodium is being studied by measuring the stick-slip friction characteristics of a ball sliding on a flat in liquid sodium and under a carefully controlled environment. Interpretation of stick-slip traces is supplemented by analysis of surface films formed during exposure to sliding in sodium. Sliding experiments are also being carried out in flowing sodium to determine the effect of oxygen content on film formation.

Progress Reported Previously

The friction and wear behavior of molybdenum, tungsten, Inconel X, Stellite 3, tungsten carbide, and titanium carbide have been investigated, and in each case, except for the two carbides, improved operation resulted when sodium was present between the rubbing surfaces. In the case of molybdenum sliding against itself, the improved behavior has been definitely correlated with the presence of a Na_2MoO_4 film.

Mathematical analysis of the stick-slip friction traces, obtained during friction experiments, revealed that viscous action is occurring during sliding in liquid sodium. These methods were shown to provide a means of detecting changes in the viscous properties of the rubbing surfaces. As a result of these preliminary successful analyses, the slow-speed stick-slip apparatus has been modified to further facilitate the detection of surface viscosity changes.

Work This Period

Experimentation involving the slow-speed stick-slip friction apparatus as well as studies of the compositions and properties of surface films produced on specimens in special environments were temporarily suspended so that full effort could be directed toward early completion of the high-purity flowing-sodium experiments.

The latter program has involved the construction of a rotating-specimen friction tester and a pumped sodium loop which incorporates both hot-trap and cold-trap purification facilities. Assembly and checkout of this apparatus is nearing completion.

Plans for Future Work

Experiments will be conducted with the high-purity flowing-sodium system to establish the validity of the theory that reacted surface films on sliding parts can be formed in molten sodium of extremely low oxygen content. Some information with respect to the likelihood of regeneration of ruptured films is also anticipated. Realization of these objectives will establish the validity of applying results obtained via the stick-slip laboratory apparatus - an oxygen-contaminated system - to actual large-scale sodium systems.

Development of Miniature Heating Elements for a
Power-Balancing-Type Thermal-Neutron Flux Sensor

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

Objective

To develop miniature heating elements suitable for use in a power-balancing-type thermal-neutron flux sensor.

Program Approach

Two distinct approaches are being used toward the development of a miniature heater. One is based on the fabrication of a film-conductance element and the other a buried, sheathed-wire heater. The ultimate element is to be about 1/8 in. in diameter and 1/2 in. long, have a nominal resistance of approximately 100 ohms, and be capable of thermal cycling and sustained operation at a maximum temperature of 1500 F in a reactor-core radiation environment. The element must also contain a specific quantity of uranium in the range of 1 to 10 mg and a thermocouple junction.

Progress Reported Previously

Imbedded-Wire Element. Sheathed differential thermocouple units were fabricated by a high-temperature-helium pressure-bonding process. Evaluations of several Chromel alloys were begun to determine their relative suitability as heaters for extended operation at 1500 F.

Film-Conductance Element. Two approaches to the fabrication of high-temperature electrical end contacts were studied. A titanium-nickel eutectic alloy bond between a titanium end cap and a pyrolytic carbon film promises success. Molybdenum-manganese metallizing appears suitable for connecting titanium end caps to palladium resinate films.

Work This Period

Imbedded-Wire Element. Close inspection of the pressure-bonded differential thermocouple assemblies revealed serious faults. The stainless steel sheath material flowed into cracks between the alumina insulators and shorted out the thermocouples, making the units useless. Grave doubts have also been raised concerning the fabrication of the complete imbedded-wire element from the standpoint of uniformity in distribution of the uranium dioxide in the cermet. Separation of the stainless steel and UO₂ powders could well result in nonuniform heat generation in the sensing element which then would lack thermal similarity to the electrically heated balance element. This could seriously affect performance of a finished sensor. Because of these difficulties, alternate wire element designs are being considered for development in this phase of the program.

Two Chromel alloys have been experimentally evaluated as potential heater wires. A 20-mil sheathed Chromel P thermocouple wire was coiled on a ceramic form and self-heated to 1600 F in a vacuum for 300 hr. The lead-in wire failed twice, and the test was terminated after the second failure. The resistance of the coil remained constant, however, for the duration of the test, indicating that a sheathed thermocouple may be used successfully as an electrical heater. Three bare coils of Chromel A wire were evaluated. All of these were preoxidized and coiled on ceramic forms with adjacent turns touching. One survived cycling between 1000 and 1500 F for 10,000 cycles and then between 1200 and 1800 F for an additional 10,000 cycles in open air with no appreciable resistance change. This coil was then subjected to self-heating in air for 300 hr at 1500 F, after which the test was terminated without failure.

Examination revealed a heavy oxide coating on the coil. The second and third coils similarly prepared and electrically heated were subjected to 70 hr at 1600 F and 1000 cycles between 1300 and 1600 F in a vacuum, with no apparent resistance change. Both were removed from test when a short developed in the lead wires of one of the coils. Heavy oxidation was not present. These last three experiments suggest that the films formed on Chromel A wire during preoxidation does serve as a potential high-temperature electrical insulator.

Film-Conductance Element. The titanium-nickel-alloy bonding method has proved successful for the production of electrical contacting to pyrolytic carbon films. Six pyrolytic carbon elements have been subjected to laboratory testing.

Three elements were encapsulated in an low-pressure inert atmosphere and placed in a furnace at 1500 F. Two showed a drastic decrease in resistance in the first 30 hr of a 500-hr exposure. Initial examination after exposure revealed a metallic appearance to the film, and it is strongly suspected that the external heating vaporized a small portion of the container or end contact material which subsequently condensed out on the carbon film, increasing the conductivity greatly. The third unit exhibited a drop then a rise in resistance to a stable value for the last 355 hr of the 500-hr exposure. This performance is explained by the possible presence of a small quantity of air within the capsule which reacted at test temperature with the film until the oxygen was completely consumed.

Three additional elements were evaluated in an inert atmosphere using electrical self-heating to attain the test temperatures. Two of them were thermally cycled between 1000 and 1500 F. The resistance of both remained essentially constant throughout the experiment. One sustained 1000 cycles and the other 3000 cycles. The third element was self-heated to 1500 F and has dropped only slightly in resistance during the first 212 hr of exposure. This element is continuing on life test.

Satisfactory end contacts for palladium-film heaters were not obtained, and further work suspended because of success with the carbon films.

Plans for Future Work

Imbedded-Wire Element. Bare wire designs will be investigated further to determine their relative stability as a function of atmosphere, temperature, and thermal cycling. A heater coil formed from 13-mil sheathed Chromel P will also be evaluated.

Film-Conductance Element. Final laboratory evaluations of pyrolytic carbon film heaters will be made using uranium dioxide containing alumina substrates. Plans and preparation for reactor testing will be advanced.

Determination of Oxygen in Sodium at Concentrations Below 10 PPM

E. H. Hall, J. McCallum, and D. R. Grieser

Objective

To demonstrate the feasibility of a method for the determination of oxygen in sodium at concentrations below 10 ppm with a sensitivity of ± 1 ppm.

Program Approach

Two approaches are presently being investigated - one is the application of a mass spectrometer, the other the technique of coulometry. The mass spectrometric method is described in BMI-1398 and the coulometric method in BMI-1469. Feasibility of these techniques for the detection of small quantities of sodium oxide in sodium is being sought.

Progress Reported Previously

Mass Spectrometry. A new heater was built for heating the sodium sample oven to higher temperatures than previously attainable. Final checkout was made.

Coulometry. Coulometric runs in sodium were made along with amalgamation sampling of the sodium before and after running. The results were inconclusive as to feasibility of this technique for oxygen monitoring.

Work This Period

Mass Spectrometry. The high-temperature oven for use in the decomposition of sodium oxide was heated to 1000 C in a checkout run. The oven was then charged with HgO and sodium. The system was evacuated and the oven heated to about 200 C. The reaction:



proceeded smoothly as evidenced by the evolution of mercury which was monitored by the mass spectrometer. The resulting solution contained about 1 per cent Na₂O in the sodium based on the ratio of reactants used. When the evolution of mercury ceased, the oven temperature was raised until the excess sodium was distilled away. The oven was finally heated to 780 C. Only small peaks for NaOH were observed and no oxygen peaks were obtained. At this temperature high-resistance leakage from the ion source to ground was again encountered, which caused the run to be terminated.

Coulometry. In the light of the probable deleterious effect of free calcium upon the operation of the coulometric technique and the inconclusive gettering behavior noted throughout the investigations, this approach has been abandoned.

Plans for Future Work

Mass Spectrometry. A modification of the ion-source structure is being made to eliminate the leakage paths. The HgO-sodium run will be repeated and the Na₂O residue heated to a higher temperature.

Arc Spectrography. An approach based on arc spectrography is being given serious consideration for experimental evaluation. This technique has been applied successfully to the determination of oxygen in metals other than sodium, i. e., titanium, steel, zirconium, etc. Experience has been gathered with this approach, and no major modification of existing equipment appears necessary in order to perform feasibility studies.

STUDIES OF FUELS (AEC-DRD)

R. F. Dickerson

The development of new and the improvement of existing fuel materials require continued research effort. The research reported in this section is supported by the AEC Division of Reactor Development for the purpose of contributing to the technology of both existing and new fuel materials. These studies of new materials are concerned with the development of niobium-base-uranium alloys for high-temperature applications, the study of plutonium-containing alloys and compounds as possible intermediate-temperature fuels or "spike" fuels for power reactors, the development of uranium-containing thorium compounds for use in thermal breeder reactors, and a study of the radiation stability of aluminum-uranium fueled glass fiber fuel elements.

As a contribution to the technology of existing fuel materials, one program is reported which has as its objective the establishment of an understanding of the important causes of fission-product release from refractory fuels while in another the irradiation behavior and phase stability of metastable beta-phase uranium alloys are being studied.

Development of Niobium-Uranium Alloys

J. A. DeMastry, D. P. Moak, A. A. Bauer, and R. F. Dickerson

Objective

To determine the fabrication characteristics, physical and mechanical properties, corrosion behavior, and irradiation stability of high-strength niobium-uranium alloys.

Program Approach

The high-temperature strength characteristics of niobium-uranium alloys indicate that they may possess high-temperature irradiation stability. Niobium alloys containing additions of 10, 20, 30, and 50 w/o uranium are being studied to determine those properties of interest for potential application of the alloys as fuel materials. The properties of interest are fabricability, tensile strength, creep strength, physical properties, corrosion resistance in various selected media, and irradiation stability.

Progress Reported Previously

The initial phase of this investigation was reported in a topical report, BMI-1400. The results of physical-property studies were reported in BMI-1434 and BMI-1448. Planning and construction of capsules for irradiation tests have been completed and were reported in BMI-1464.

Difficulties in the fabrication of alloys containing 30 w/o or more uranium have been encountered. A program concerned with the preparation of niobium-uranium-alloy ingots with structures amenable to fabrication was undertaken. Alloys containing nominally 10, 20, 30, and 50 w/o uranium and a test ingot of unalloyed niobium were prepared by skull melting utilizing the consumable-electrode procedure.

Work This Period

Chemical analysis of the skull-cast material is complete and results are shown in Table B-1. The uranium content is lower by about 5 w/o than expected in all cases. In order to minimize the amount of niobium used, a single niobium skull was built up

and used to melt all the alloys in order of increasing uranium content. Apparently dilution of the charge composition by the skull occurred to a greater extent than was anticipated. While the nominal compositions desired were not achieved by melting, the alloys prepared will permit the variation in properties as a function of composition to be determined. Random gas analyses indicate no serious impurity pickup during melting.

TABLE B-1. RESULTS OF CHEMICAL ANALYSES OF SKULL-CAST NIOBIUM-URANIUM ALLOYS

Uranium Content (Balance Niobium), w/o		Impurity Content, ppm			
Nominal	Analyzed	Melting-Stock Niobium		As-Cast Ingot	
		Oxygen	Hydrogen	Oxygen	Hydrogen
10	4.38	85	1	160	3
20	14.3	85	1	--	--
30	25.0	85	1	--	--
50	46.7	85	1	115	2

Ingot slabs, 2 by 2 by 1/2 in. of nominal niobium-10 and -20 w/o uranium composition were successfully reduced 85 per cent by rolling in air at 1000 F. The nominal niobium-30 w/o uranium alloy was reduced 80 per cent by rolling in air at 1000 F. Fabrication of an alloy nominally containing 50 per cent uranium was unsuccessful at 1000 and 1850 F. Creep and tensile specimens were machined from the fabricated material.

Creep tests have been initiated on the niobium-5 and -15 w/o uranium alloys and specimens of niobium-25 w/o uranium are being machined for test. It is tentatively planned to establish the stresses necessary to produce deformation rates of 0.1 and 0.001 per cent per hr at 1600 and 2000 F. No results have been obtained as yet.

Small slabs (2 by 1 by 1/2 in.) of the niobium-47 w/o uranium alloy have been enclosed in molybdenum packs for investigation of fabricability by rolling at 2000 to 2400 F.

Table B-2 lists the results of chemical analyses of the irradiation specimens. All compositions are well within experimental error. The carbon and nitrogen contents are relatively high for these alloys. This is probably due to high carbon and nitrogen content of the enriched uranium. Analysis of the enriched uranium is not complete.

TABLE B-2. RESULTS OF CHEMICAL ANALYSES OF IRRADIATION FUEL PINS^(a)

Nominal Composition (Balance Niobium), w/o	Enrichment, per cent	Analyzed Content, w/o		Impurity Content ^(b) , ppm			
		Uranium	Zirconium	Oxygen	Hydrogen	Carbon	Nitrogen
10 U	93.19	10.4	--	265	18	580	210
20 U	37.5	21.1	--	87	16	460	120
30 U	30.22	30.4	--	142	8	320	60
20 U-10 Zr	30.22	19.9	9.9	187	4	270	90
20 U-20 Zr	30.22	19.7	20.0	232	5	250	80

(a) Average of three analyses.

(b) Niobium alloy stock contained as follows: 94 ppm oxygen, 55 ppm nitrogen, 75 ppm hydrogen, and 43 ppm carbon.

Control samples of the compositions which will be irradiated have been enclosed in stainless steel capsules containing sodium. These capsules are being heat treated at 1600 and 1800 F for a period of time which will approximate the irradiation time (2400 hr). It is hoped that this test will allow a more thorough evaluation of the results of irradiation by giving an indication of the effect of temperature alone on the irradiation samples.

Plans For Future Work

Creep tests will be continued until the stresses to produce creep rates of 0.1 and 0.001 per cent per hr at 1600 and 2000 F are established.

Heat treatments of enriched irradiation control samples at 1600 and 1800 F will be continued until a 2400-hr period has been accumulated.

Irradiation studies of fuel pins will begin as soon as suitable reactor space is available.

Development of Uranium-Containing Thorium Compounds

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

Objective

To investigate thorium and thorium-uranium compounds with the aim of developing high-temperature fuels for thermal breeder reactors and of determining those properties which will indicate their suitability for various reactor applications.

Program Approach

High-melting compounds having high thorium (uranium) density and low thermal-neutron cross section, namely, Th_2Al , ThBe_{13} , ThB_4 , ThC , ThC_2 , ThN , ThS , Th_3Si_2 , and ThSi , and these same compounds with a 10 a/o addition of uranium, are being evaluated from the standpoint of preparation, corrosion resistance, and mechanical and physical properties.

Progress Reported Previously

Satisfactory cylindrical arc castings of Th_2Al , ThC , ThC_2 , Th_3Si_2 , and ThSi and these compounds with a 10 a/o uranium addition have been obtained. Arc-melted buttons of the beryllides and borides have been made; however, satisfactory arc-cast cylinders have not been obtained. Measurements show that the castings have densities within 2 per cent of theoretical. Dry-air and CO_2 oxidation tests at 1200 F showed that the borides and beryllides were very resistant to attack by these media. Corrosion tests in 350 C Santowax indicated that the aluminides, beryllides, and dicarbides were the most resistant in this medium. Water corrosion tests at 90 C show that the borides and beryllides are superior, although the aluminides and silicides had negligible weight losses. The carbides reacted very rapidly in water at 90 C. Oxidation and corrosion data were reported in BMI-1480, BMI-1489, BMI-1496, and BMI-1504.

Work This Period

The corrosion resistance of the thorium-base compounds being studied was determined in NaK at 1200 F. The results of a 1-month test are given in Table B-3. Although the results are for a single test, the corrosion rates obtained can be used for qualitative comparisons. The beryllides performed well in NaK, the carbides had relatively high weight losses, while the weight changes of the other compounds were intermediate. The weight changes noted are believed to result primarily from reaction with the oxygen contained in the NaK.

TABLE B-3. CORROSION OF THORIUM AND THORIUM-URANIUM COMPOUNDS
BY NaK AT 1200 F

Nominal Compound Composition	Weight Change, mg/(cm ²)(day)
Th ₂ Al	-0.029
(Th ₉ U) ₂ Al	+0.034
ThB ₄	+0.016
(Th ₉ U)B ₄	Broke in pieces
ThBe ₁₃	-0.0013
(Th ₉ U)Be ₁₃	-0.0044
(Th ₉ U)C	-0.045
(Th ₉ U)C ₂	-0.015
Th ₃ Si ₂	-0.0009
(Th ₉ U) ₃ Si ₂	+0.022
ThSi	+0.015
(Th ₉ U)Si	+0.0085

Thermal-shock tests performed by quenching specimens from 1000 C showed that all of the compounds being investigated withstood rapid cooling without cracking.

Hot-hardness measurements of thorium-uranium base compounds have been initiated, but significant data have not been obtained yet.

Plans For Future Work

Studies planned for the future include determinations of thermal expansivity, thermal conductivity, and strength characteristics.

A Study of Plutonium-Containing Fuel Alloys

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

Objective

To investigate those plutonium alloys which appear to have potential as fuel for relatively high-temperature propulsion applications or for possible use as "spike" fuel elements in power reactors.

Program Approach

Plutonium-containing alloy systems have been selected for study on the basis of the behavior of similar uranium-alloy systems. The compositions selected are based on the niobium, thorium, uranium-molybdenum, and uranium-niobium systems. These systems are being studied at the plutonium-lean end. Initial evaluation is based on metallographic studies and hardness measurements. Those alloys exhibiting desirable phase distribution and evidence of strength on the basis of hardness measurements will be subjected to more detailed property studies.

Progress Reported Previously

As the microstructures of binary niobium-plutonium alloys appeared unfavorable, attempts were made to tie up the plutonium as a high-melting alloy phase or compound by ternary alloying with zirconium and silicon (BMI-1464). Niobium-10 w/o plutonium-10 and -30 w/o zirconium alloys were found to be three-phase, with two of the phases being present at the grain boundaries. X-ray diffraction analysis of a niobium-25.6 w/o plutonium-6.1 w/o silicon alloy resulted in tentative identification of its two-phase structure as niobium and Nb₅Si₃; the plutonium apparently is in solution in the Nb₅Si₃ (BMI-1504).

A thorium-10 w/o plutonium-10 w/o zirconium alloy was harder and more resistant to room-temperature oxidation than binary thorium-plutonium alloys containing 5 to 30 w/o plutonium (BMI-1489). On the basis of hardness data, alloys appear to recrystallize within 1 min at 700 C (BMI-1504).

Work This Period

Hardness data suggest that complete recrystallization of 80 and 90 per cent cold-reduced thorium-5 and -10 w/o plutonium alloys occurs within 1 min at both 650 and 700 C. An 80 per cent cold-reduced thorium-10 w/o plutonium-10 w/o zirconium alloy exhibited a continuous decrease in hardness with increasing time up to 10 min at 650 and 700 C. The rate of decrease in hardness was more rapid at 700 C. The effect of additional time at these temperatures will require investigation. Metallographic examination of these alloys will be necessary also to establish whether recrystallization is occurring.

Cold-worked thorium-30 w/o plutonium alloys disappeared when heat treated in a lead bath at 650 C. This suggests either a reaction between the alloy and molten lead or a solidus temperature below 650 C. While no reaction between lead and the thorium-5 and -10 w/o plutonium alloys was noted, it is possible that the solubility of alpha thorium in lead is increased appreciably by larger (30 w/o) additions of plutonium. To further support this theory, no evidence of melting was noted when the thorium-30 w/o plutonium alloy was heat treated under vacuum at 800 C.

X-ray diffraction analysis indicated the primary phase in a cast niobium-10 w/o plutonium-10 w/o zirconium alloy to be niobium-zirconium solid solution with a lattice parameter of 3.22 A. The lattice parameter of unalloyed niobium is 3.294 A. No X-ray patterns for the two secondary grain-boundary phases were observed. The X-ray diffraction technique employed is not sensitive to phases comprising less than 10 per cent of the alloy, which is the case with these particular unknown phases.

Plans For Future Work

Recrystallization studies of cold-worked thorium-base alloys will continue with emphasis on metallography to determine the degree of recrystallization.

Attempts at identifying the unknown grain-boundary phases in the niobium-plutonium-zirconium alloys will be made by etching away the primary phase, hence increasing the relative quantity of grain-boundary phases. Also, effects of heat treatment on the microstructures will be investigated. Additional niobium-plutonium-silicon alloys are being prepared for investigation in an effort to more completely understand the phase relationships in the niobium corner of the system. Of particular interest is the relationship between plutonium and Nb_5Si_3 .

Compatibility of PuO_2

S. J. Paprocki, D. L. Keller, and W. M. Pardue

Objective

To study the compatibility of PuO_2 with various metallic and ceramic materials.

Program Approach

Heat treatments of powder mixtures and abutted surfaces of PuO_2 with metal and ceramic materials are being conducted. The appearance of new phases will be followed by X-ray diffraction measurements, metallographic examination, and changes in the physical appearance of the compacts.

Progress Reported Previously

A preliminary screening procedure using calculations of standard free energies to indicate metal PuO_2 reactions is summarized in BMI-1455. The as-received PuO_2 particles average 9.4μ in diameter. Ten volume per cent of an extraneous phase was tentatively identified as Pu_2O_3 . Air oxidation of the loose powder for 3 hr at 600 to 800 C eliminates this phase and increases the average particle size to 13.1μ . Sintering in dry hydrogen or dry argon at 1700 C causes a chemical reduction to hexagonal Pu_2O_3 (oxygen:plutonium ratio of 1.55) with melting and plastic flow exhibited by the pellets. This reduction is dependent upon heating rate as well as moisture and oxygen content of the atmosphere. From available thermodynamic data, it has been calculated that a moisture content of approximately 1 per cent (+42 F dew point) would be required to prevent the continuous reduction of PuO_2 in a flowing hydrogen atmosphere at 1727 C. At a lower temperature, 1527 C, in hydrogen, a moisture content of less than 0.09 per cent (-5 F dew point) would be expected to promote reduction of PuO_2 .

In dry argon, the heating rate is the predominantly controlling variable. Since the as-received PuO_2 contains 10 to 15 volume per cent of Pu_2O_3 and the flowing argon contains some small quantity of moisture and oxygen, a low heating rate will permit oxidation of the Pu_2O_3 to PuO_2 before the melting point (1610 C) of the lower oxide is reached.

These calculations have been correlated with experimental observations. Rapid heating of PuO_2 pellets to 1700 C in dried argon resulted in melting, while pellets soaked overnight in argon at 1000 C and sintered for 3 hr at 1700 C in argon which was not dried before use showed no melting. Pellets sintered in hydrogen for 1.5 hr at 1600 C, or below the melting point of Pu_2O_3 , were quite sound. Earlier sintering in dry hydrogen at 1700 C resulted in gross melting and plastic flow.

In view of all evidence, it is concluded that melting is caused by the prior existence of Pu_2O_3 or the reduction of PuO_2 to Pu_2O_3 in dry sintering atmospheres followed by subsequent heating to a temperature in excess of the melting point of Pu_2O_3 .

Compatibility determinations on PuO_2 pellets which were completely encased in cold-pressed metal powders indicate that PuO_2 is stable in stainless steels and Inconel at either 1900 or 2000 F for a 2-hr period in hydrogen. Types 304, 316, 318, and 347 stainless steels were used. On the basis of preliminary runs, MgO appears to be the most suitable container material for PuO_2 at 1700 C.

Work This Period

Continued studies of the compatibility of PuO_2 and the various stainless steels and Inconel investigated earlier were performed. Examination after sintering for 2 hr at 2150 F in hydrogen did not reveal any PuO_2 -metal reaction zones.

Compatibility determinations were initiated for PuO_2 pellets completely encased in cold-pressed MgO, ZrO_2 , Al_2O_3 , molybdenum, and tungsten powders. All of these duplex compacts were sintered 4 hr at 2900 F in hydrogen and then held at 2500 F for 20 hr to insure maximum possibility of reactions. Visually, all compacts appeared free of reaction. The MgO, ZrO_2 , and tungsten compacts all exhibited some cracking. This is associated with the low sintering temperature relative to the respective melting points of the materials involved, since the cracks were of a cleavage type, indicating poor bonding.

Plans For Future Work

Examination of the refractory material- PuO_2 specimens which were heat treated during the past month will be undertaken. In addition, compatibility determinations of niobium, titanium, vanadium, and Zircaloy-2 with PuO_2 will be made.

Fission-Product Release From Refractory Fuels

J. B. Melehan, S. D. Beck, and F. A. Rough

Objective

To establish an understanding of the important causes of fission-product release during irradiation of refractory fuels, starting with uranium dioxide, and to obtain engineering information which will aid in the design of fuel elements utilizing these fuels.

Program Approach

The principal effort is devoted to a study of the continuous release of fission products during irradiation. This work is supported by studies of characterization of the materials and postirradiation diffusion measurements.

Progress Reported Previously

The results of in-pile fission-gas-release experiments suggested a surface-area-dependent mechanism for release from UO_2 which did not vary significantly up to a temperature of 1800 F. A mathematical analysis of release by simultaneous diffusion and surface-recoil mechanisms was developed for the case of a fuel having flat-plate geometry.

Work This Period

Additional efforts at characterizing sintered UO_2 bodies by quantitative metallography are under way. The objective is to determine the open surface area through a process of impregnation of the open pores.

It has already been mentioned that the principal problem encountered during in-pile operation of the r-f induction furnace has been the failure of induction coil and coil leads as the result of high-voltage electric arcing.

Several rubber and plastic insulating materials have not been successful in solving this problem. Glass and Vycor, although their characteristic brittleness is a drawback, appear to be two of the more promising candidates as electrical insulators. Their electrical properties are not significantly changed by fast-neutron and gamma irradiation until irradiation dosages are in the range of 10^{19} to 10^{20} nvt. Also, annealing of damage in glass can be expected at elevated temperatures. Consequently, a Vycor electrical insulating cover was fabricated to contain the induction coil. Similarly, the induction-coil leads are to be enclosed in Vycor tubing, the objective being to enclose in a continuous glass insulating cover that part of the induction coil and the coil leads which are located in the region of highest neutron and gamma-ray flux. An out-of-pile test of this modification of the coil and its insulation is presently being prepared, although a direct indication of its in-pile behavior will require operation under actual in-pile conditions.

It appears that the best possibility for solving the problem of high-temperature thermal insulation for the susceptor is to design segmented insulators so as to avoid the high thermal stresses which develop in massive single-piece insulators. Enclosure of the induction susceptor in small ceramic insulating pellets or loosely compacted insulating powder is an approach which merits some consideration. While alumina is an effective thermal insulator to 3000 F, higher temperature ceramics, such as zirconia, will be required for thermal insulation above 3000 F. Zirconia is not only more stable mechanically and chemically, but it retains its electrical insulating properties above 3500 F.

In a parallel effort, the feasibility of a resistance furnace for 3000 to 4000 F operation is being considered.

Plans For Future Work

Future work will continue in the solution of the problems of electrical and thermal efficiency of the induction furnace, electrical insulation of the induction coil to prevent failure by electrical arcing, and improvement of mechanical stability of ceramic thermal insulating components. These efforts will be made with the objective in mind to extend the furnace capability to above 3000 F.

Radiation Stability of Aluminum-Uranium Fueled Glass Fiber Fuel Plates

R. J. Burian and J. E. Gates

Objective

To determine the effects of reactor irradiation on the mechanical properties of a material composed of aluminum-50 volume per cent uranium-fueled glass fiber.

Program Approach

Tensile and bend-test specimens prepared from fuel plates fabricated from aluminum-50 volume per cent uranium-fueled glass fiber will be irradiated to a burnup of about 25 per cent of the uranium-235 at temperatures below 100 C. The radiation stability of the material will be evaluated by a comparison of mechanical properties before and after irradiation.

Progress Reported Previously

The program was initiated this report period.

Work This Period

Specimen design was determined and negotiations were initiated with the Clevite Corporation to obtain specimens of the selected designs. The tensile specimens will be nominally 2-1/2 in. long and 1/2 in. wide with a 3/4-in. long by 3/8-in. -wide gage section. The rectangular bend-test specimens will be 2-1/8 in. long and 3/8 in. wide. The specimens will contain a 0.020-in. -thick core with 0.020 in. of cladding on both surfaces except for three unclad tensile specimens. The core will contain 50 volume per cent glass fibers fueled with 50 w/o fully enriched U₃O₈. The specifications for the nine specimens are as follows:

- (1) Two tensile specimens clad with either 1100 or 6061 aluminum, except for bare edges.
- (2) Two tensile specimens clad with aluminum-unfueled glass fiber, except for bare edges.
- (3) Three tensile specimens without cladding.
- (4) Two bend-test specimens clad with either 1100 or 6061 aluminum, except for bare edges.

Capsule design has been initiated and negotiations for reactor space are in progress. Samples of the three types of material to be irradiated have been obtained for NaK-compatibility tests. Specimens will be heated in NaK at about 150 C for about 2 weeks prior to examination.

Plans For Future Work

The NaK-compatibility tests will be performed. Preirradiation examination of the irradiation specimens will be completed and the specimens encapsulated. Irradiation of the specimens will begin in May or June.

Irradiation Study of Metastable Beta-Phase Uranium Alloys

M. S. Farkas, B. D. Hamilton, A. A. Bauer, and R. F. Dickerson

Objective

To study the irradiation behavior and phase stability during irradiation of metastable beta-phase uranium alloys.

Program Approach

Uranium alloys with additions of 0.3 w/o chromium and 0.3 w/o chromium-0.3 w/o molybdenum designed to retain the metastable beta phase have been developed by Nuclear Metals, Inc. Eighteen specimens of these alloys, prepared from uranium of 10 per cent enrichment, in a variety of conditions of heat treatment, are to be irradiated in two temperature-controlled capsules. The specimen compositions and heat-treatment conditions were reported in BMI-1489.

Capsule design is intended to maintain the specimen center-line temperature at 850 F at an effective flux of about 10^{13} nv. The specimens in the first capsule are to be irradiated in the BRR to a burnup of 0.01 to 0.05 a/o over a 2 to 3-day period. These specimens are to be examined after irradiation, primarily to determine if the beta phase has been retained during irradiation. The specimens in the second capsule are to be irradiated under similar conditions to a burnup of about 0.3 a/o to investigate the irradiation stability of the alloys.

Progress Reported Previously

The design of the two capsules to contain the 18 uranium-alloy pins during irradiation was completed. These capsules are double-walled NaK-containing systems utilizing electrical-resistance heaters to control specimen center-line temperatures. A natural-uranium pin is included in the bottom of each capsule. The adjacent radial heat-transfer path is specially tailored in an attempt to maintain the center-line temperature of the natural pin approximately the same as that of the enriched pins.

Work This Period

The fabrication of capsule components was completed and capsule-loading operations were begun.

X-ray diffraction examinations were conducted on the surfaces of all irradiation specimens. The objective was to make certain that beta-phase decomposition to alpha had not occurred during the period the samples were stored after preparation and heat treatment. It was determined that beta remained the major phase in all samples heat treated to retain beta, although varying minor-phase amounts of alpha were also detected. The results indicated that significant beta-phase decomposition had not occurred.

Plans For Future Work

Density measurements and photomicrographs of the specimens to be irradiated will be obtained prior to encapsulation. One low-burnup capsule is tentatively scheduled for insertion in the BRR during the week of April 17. The capsule will be irradiated

B-10

for approximately 3 days to achieve the desired burnup level of 0.01 to 0.05 a/o. The high-burnup capsule is scheduled for insertion into the MTR on May 1, 1961. Approximately five MTR cycles (75 days) will be necessary to obtain the reference specimen burnups of 0.3 to 0.4 a/o.

GENERAL FUEL-ELEMENT DEVELOPMENT (AEC-DRD)

S. J. Paprocki

The studies reported in this section are being performed for the AEC Division of Reactor Development.

Techniques have been developed for the fabrication of cermet fuels containing 80 volume per cent of UO_2 and 20 volume per cent of the metals chromium, molybdenum, niobium, and stainless steel. In comparison to bulk UO_2 , these materials possess much improved thermal-shock resistance, thermal conductivity, and rupture strength. Based on these measured improved properties, an irradiation program has been formulated to correlate these results with the irradiation behavior of bulk UO_2 .

The cermet fabrication studies have been extended to include UN and UC fuels. Although the thermal conductivity of these materials is superior to UO_2 , an additional benefit is obtained in the cermet structure because the metal phase in the cermet can be metallurgically bonded to the cladding, eliminating the cladding-to-core void. In addition, there is a significant improvement in structural strength and thermal-shock resistance.

The techniques of hot pressing and gas-pressure bonding are increasing in importance as methods for bonding of nuclear fuel and structural components, and a study is being conducted to obtain a better understanding of the kinetics and mechanism of these bonding processes.

In some reactor applications, tubular fuel elements offer distinct advantages over flat-plate fuel systems; however, established techniques for the fabrication of tubular fuel elements are restrictive and of relatively high cost. A study is being conducted to evaluate new techniques for fabrication of tubular elements with emphasis on simplicity and low cost.

Irradiation of Cermet Fuels

S. J. Paprocki, G. W. Cunningham, D. E. Lozier, and E. O. Fromm

Objective

To investigate the irradiation stability of UO_2 -refractory metal (chromium, molybdenum, and niobium) cermet fuels.

Program Approach

Two capsules containing clad (with 35 mils of Type 430 stainless steel or niobium) cylindrical specimens of UO_2 -refractory metal fuels are being prepared for high-temperature high-heat-flux irradiation at the Materials Testing Reactor. Three refractory-metal matrices, chromium, molybdenum, and niobium, are involved; each is combined with 80 volume per cent minus 100 plus 200-mesh spherical UO_2 enriched to 20 per cent. Each capsule will contain six specimens, two each of the three materials indicated above, and will be equipped with thermocouples for specimen-temperature monitoring. The two capsules will be irradiated in similar thermal-neutron fluxes but for different periods of time to achieve two levels of specimen burnup

(approximately 2 per cent of the total uranium atoms present during three normal MTR cycles in the one case and approximately 5 a/o during eight normal cycles in the other).

Progress Reported Previously

Except for certain heat-transfer details, the capsule design for the irradiations was completed. The design is based on a double-wall configuration with the specimens immersed in NaK for heat-removal purposes.

The governing thermal parameters include specimen center-line temperatures of approximately 2650 F for the molybdenum- UO_2 and niobium- UO_2 and 2350 F for the chromium- UO_2 , and specimen-surface heat fluxes of approximately 10^6 Btu/(ft²)(hr). The three materials have different thermal conductivities, and, consequently, it has been necessary to vary specimen diameters to satisfy the desired combinations of thermal conditions.

Cermets containing 80 volume per cent minus 100 plus 200-mesh spherical UO_2 and molybdenum or chromium have been clad with Type 430 stainless steel by an isostatic pressure of 10,000 psi at 2300 F for 3 hr. The specimens were tested by thermally cycling between 200 and 1850 F for 25 times and heat treating for 168 hr at 1800 F. Cracks resulting from a reaction zone in the molybdenum-containing cermet have been prevented by including a 5-mil molybdenum barrier foil as part of the 35-mil cladding in this cermet.

Work This Period

The heat-transfer studies of the capsule design were completed during March and the fabrication of capsule components continued. The final heat-transfer studies produced certain adjustments in the radial zones adjacent to each of the three sets of encapsulated specimens. Since the fission-heat generation rates among the specimen pairs will be different owing to singular combinations of specimen size and effective flux, it has been necessary to tailor each radial zone individually. The final design will feature special radial heat-flow elements as follows:

<u>Specimen Pair</u>	<u>Design of Adjacent Radial Zone</u>
Molybdenum- UO_2	A molybdenum sleeve in the NaK annulus, and metallic fins for thermal contact between the inner and outer shells; total fin thickness is 0.105 in. per axial in.
Chromium- UO_2	Metallic fins as above; total fin thickness is 0.055 in. per axial in.
Niobium- UO_2	A 0.008-in. helium annulus

Dies for the three different core sizes were machined and dummy specimens pressed and clad by pressure bonding. The specimen containing the molybdenum metal phase was 0.004 in. oversize in diameter after bonding. The chromium-containing cermet was within tolerance, and the cermet containing the niobium phase was oversize; however, the niobium on the coated UO_2 to be used in the irradiation specimens is more ductile and should be densified to the correct dimensions by pressure bonding.

Plans For Future Work

The capsules will be assembled as soon as parts are fabricated and specimens become available. It is anticipated that both capsules will be ready for irradiation in June, 1961.

Specimens containing 20 per cent enriched UO_2 will be prepared for the first capsule loading.

Fabrication of Cermet Fuel Elements

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

Objective

To develop fabrication techniques for producing UN cermets containing a minimum fuel loading of 60 volume per cent to densities of 90 per cent of theoretical or greater as well as to conduct a cursory evaluation of fabrication techniques for producing UC cermets.

Program Approach

Specimens are being fabricated by the gas-pressure-bonding technique. Cermet evaluations are being made on the basis of microstructure and density. In addition, metal-ceramic compatibility results are being obtained from fabricated cermets.

Progress Reported Previously

Cermets of 80 volume per cent UN dispersed in molybdenum, chromium, and Type 316 stainless steel were gas-pressure bonded for 3 hr at 2400 F under a helium pressure of 10,000 psi. The cladding was machined from the cermets. Measured densities were 90.4, 85.0, and 95.1 per cent of theoretical, respectively. These densities compared favorably with previously reported densities measured by the point-counting technique on specimens gas-pressure bonded in compartmentalized tubes.

Work This Period

Green-pressed cores of 80 volume per cent UN dispersed in molybdenum and chromium have been gas-pressure bonded for 3 hr at 2500 F under a helium pressure of 10,000 psi. A niobium tube 0.470 in. in ID was used as a container. After cladding removal the cermet densities were 90.9 and 91.0 per cent of theoretical, respectively, for the molybdenum and chromium matrix cermets. These specimens are being prepared for thermal-conductivity measurements.

Green-pressed cores of minus 100-mesh UN and UC have been loaded in 0.470-in. -ID niobium tubes. The UC contained 5.2 to 5.5 w/o carbon, estimated from the microstructure. The tube containing the UC cores was loaded with alternate green-pressed cores of molybdenum, rhenium, and tungsten between each UC core. The tube containing the UN cores was loaded with green-pressed cores of molybdenum, rhenium, tungsten, and tantalum between each UN core. All cores were approximately 1/2 in. in length. After loading each tube was outgassed at 1500 F to remove the Carbowax from the UN and UC cores which had been added to prevent oxidation during handling. The tubes were sealed for gas-pressure bonding by electron-beam welding 1/8-in. -thick niobium end plugs in the tubes. After gas-pressure bonding for 3 hr at 2600 F under a helium pressure of 10,000 psi the tubes were sectioned through the fuel and metallographically polished along the length dimension to expose the metal-fuel interface.

Metallographic examination of the UN-metal interfaces has not been completed at this time. Microscopic examination of the UC and metal cores revealed densities of 90 per cent of theoretical or greater in all cases. The molybdenum-UC interface contained two definite reaction zones, one metallic in appearance approximately 0.002 in. in thickness adjacent to the molybdenum core and a gray zone 0.002 to 0.0025 in. in thickness adjacent to the UC. The gray interface is believed to be UO_2 . The metallic-appearing interface is probably a result of interdiffusion of carbon from the UC to the molybdenum, resulting in the formation of molybdenum carbide. In addition, the dark-gray phase assumed to be UO_2 is extended along grain boundaries within the uranium carbide to a depth of approximately 0.01 in.

At the rhenium-UC interface a dark-gray phase assumed to be UO_2 existed in a zone approximately 0.0005 in. in thickness. The amount of U_2C_3 structure present in the UC core increased in amount nearer the interface, suggesting that possibly oxygen from the rhenium substituted for carbon in the UC. No reaction zone was detected in the rhenium, further indicating their compatibility.

The tungsten-UC interface revealed a gray phase adjacent to the UC (apparently UO_2) and two white zones between the gray zone and the tungsten core assumed to be tungsten carbide and tungsten-tungsten carbide solution. The white zones were each approximately 0.0005 in. in thickness while the gray reaction zone varied from approximately 0.0005 in. to 0.002 in., blending into a three-phase region assumed to be UO_2 , U_2C_3 , and UC.

It seems plausible that the dark-gray phase at the interface and within the UC fuel is UO_2 as only approximately 700 ppm of oxygen would have to be contained in the metal powder cores to form the maximum thickness of dark-gray phase observed. In addition, since the oxides of the refractory metals used are volatile below the densification temperature, one would expect them to be dispersed through the porosity within the UC for a considerable depth.

Plans for Future Work

Examination of the UN-metal interfaces will be completed. In addition, similar UC- and UN-metal compatibility specimens, as described in this month's report, will be made using green-pressed cores which have been vacuum sintered at 2600 F before gas-pressure bonding.

Factors Affecting Pressure Bonding

D. C. Carmichael, G. W. Cunningham, and J. W. Spretnak

Objective

To determine the mechanism of solid-phase bonding of metal components under the application of heat and pressure.

Program Approach

Hot pressing or gas-pressure bonding under various conditions of temperature, pressure, and time are being used to bond specimens for study. The specimens are of different designs and preparations and are composed of two components of the same metal. Evaluation of the bonds is based chiefly on metallographic examination, using special techniques. Observations are made of the extent of contact between mating surfaces and of the behavior of grain boundaries, dislocations, and interfacial voids which affect the elimination of the interface during bonding.

Progress Reported Previously

The mechanism for the self-bonding of copper has been established and the effects of pressure, temperature, time, grain growth, impurities, and surface roughness have been investigated. In order to produce a metallurgically sound bond, i. e., no discontinuities or changes in the structure in the bond region, the first requirement is the application of sufficient pressure to place the surfaces in intimate contact. For copper, it was found that the pressure required could be related directly to Meyer hot-hardness values.

Grain growth across the bond interface and thus elimination of the interface has been found to be dependent upon the presence or absence of microvoids in the bond region. Microvoids can be created during bonding by insufficient flow of metal or by condensation of excess vacancies at impurity nuclei. Decreasing the number and size of microvoids to a level permitting grain growth depends upon the availability of vacancy sinks in the metal. In copper, the most effective sinks were found to be grain boundaries.

Work This Period

The pressure-bonding mechanism is now being studied in more detail and in other metals than copper. Initially, the mechanism and the pressure-hot hardness relationship for obtaining intimate contact are being investigated for other selected metals, including high-temperature metals. Modification of the vacuum hot-pressing equipment for operation at higher temperatures has been completed. Thus far, temperatures up to 2400 F have been utilized in bonding specimens of niobium. Some materials for specimens have been obtained and others are being procured. Meyer hot-hardness values for some of the materials are being determined.

Plans For Future Work

During the coming months the hot-hardness at various temperatures for metals of interest will be measured and additional specimens will be prepared for investigation of the mechanism of pressure bonding.

Development of Low-Cost Tubular Fuel Elements

S. J. Paprocki, R. J. Carlson, and E. G. Smith, Jr.

Objective

To develop techniques for fabricating low-cost tubular fuel elements of consistent quality.

Program Approach

Several approaches are under current investigation. One of these involves the assembly of concentric core and cladding components followed by densification, bonding, and sizing by techniques such as explosive forming or pressure bonding. Various ways to locate the core powder mixtures are being considered, including tamp packing into the annulus between the concentric cladding tubes and the use of binders to form the mixture into sheet which can then be formed into a tubular shape.

In another approach, multiple-cored fuel plates are fabricated in a conventional manner with each core running the length of the plate. Subsequent to rolling, two such plates are lined up core-for-core and joined together by a technique such as seam welding in such a manner that the areas between the cores can be expanded or opened up to

form tubular shapes, with the cores from each plate making up half of the circumference of the tubular cross section. Hydraulic, pneumatic, or mechanical methods may be used to open up the tubular sections.

Progress Previously Reported

Early studies using a plastic binder such as Acryloid have shown that relatively large amounts were required to obtain the desired working characteristics in a stainless steel-UO₂ powder mixture (20 w/o using minus 325-mesh stainless steel powder and 10 w/o using minus 100-mesh material). Recent experiments indicate that essentially no binder residue remains after sintering at 1600 F for 1 hr followed by 2300 F for 1 hr in hydrogen.

Explosive-compaction studies using both formed binder-powder mixtures and loose-packed powders indicate that areas of low density exist in the core annulus prior to densification. Although appreciable density was obtained in the samples, areas of surface roughening were observed on the ID and OD of the tubes. Sections of the as-compacted tubes were heat treated for 2 hr at 2450 F in hydrogen to enhance the core-cladding bond and improve the over-all density of the core.

Expanded multiple-tube assemblies have been prepared using both spot welding and seam welding as a means of joining the two fuel plates prior to expansion of the individual fuel sections by mechanical means. Negotiations are under way for the procurement of seam-welding equipment for further development of this joining technique.

Fabrication studies of spot-welded specimens to produce a recrystallized structure have been completed. Hot rolling at 2200 F results in a complete recrystallization of the weld with grain size a function of reduction per pass, large reductions giving the smaller grain size. Cold rolling breaks down the weld nugget to some degree, but further heat treatment is necessary to recrystallize the cast structure.

Work This Period

Heat treatment of the cold-rolled spot welds at 1800 F for 1 hr followed by air cooling resulted in almost complete recrystallization of the weld zone. Cold rolling would be preferred to hot rolling as no elaborate surface preparation would be required to prevent the plates from sticking together over the core sections.

Explosive-forming studies have indicated the necessity for further improvement in loading techniques to produce a uniform density in the core prior to further densification.

Variations in fabrication schedules and heat treatments have been made with the plastic-binder approach to improve on the uniformity of the density obtained in the green fabricated sheet. Samples were prepared using minus 100- and minus 325-mesh stainless steel powder. Different initial thicknesses of material were made so as to employ different reductions when cold rolling to sheet of 0.024 to 0.027 in. thickness. The strips were subsequently sintered using two schedules. One consisted of heating to 1600 F for 1 hr followed by 2 hr at 2450 F while the other involved heating at 2450 F for 2 hr only. Radiographs taken before and after the sintering step were not sensitive enough to reveal any density variation in the strips. The strips containing the minus 100-mesh stainless powder given the two-step sintering cycle exhibited the best ductility, as they could be formed easily to any shape. Further, the strips given the largest cold reduction showed the best surfaces, both in the green and in the sintered conditions.

There were, however, bubble formations on some of the strips which were believed to be caused by the binder coming out of the strip. The two-step sintering schedule reduces the amount of bubble formation by reducing the rate at which the binder is removed.

Hydrastatic pressing has been considered for obtaining sufficient green density prior to subsequent gas-pressure bonding. Tubular specimens containing a loose-powder dispersion of stainless steel and UO_2 have been prepared and subjected to a hydrostatic pressure of 100,000 psi. Sectioning of the specimens showed that some densification of the powder core had taken place. However, it was not possible to polish a metallographic specimen as the powder was pulled out of the annulus. This may be indicative of little bonding among the powder particles, which is not unexpected, since the pressing was accomplished at room temperature.

Plans For Future Work

Metallographic examination of explosively compacted and heat-treated specimens will be completed. Emphasis on loading procedures to obtain uniform core densities will be continued with both the loose-powder and plastic-binder loading techniques. Extrusion of plastic-binder mixtures will begin after procedures for obtaining uniform green densities have been established.

Development of seam-welding technology for joining multiple-core flat plates will begin as soon as equipment is obtained.

GAS-PRESSURE BONDING OF CERAMIC, CERMET, AND DISPERSION FUEL ELEMENTS (ARC-FUEL CYCLE)

S. J. Paprocki, S. W. Porembka, D. L. Keller,
E. S. Hodge, and C. B. Boyer

Objective

To develop and refine the gas-pressure-bonding process as a method for fabricating low-cost ceramic, cermet, and dispersion fuel elements.

Background

Through the application of gas-pressure bonding, both fuel densification and cladding are accomplished in a single bonding operation. The technique appears to be a promising method for producing quality fuel elements at a reduced cost. The work is being carried out in support of the AEC Fuel-Cycle Development Program.

Program Approach

Major emphasis has been placed on the uranium dioxide ceramic fuel system as clad with Type 304 stainless steel. Past investigations have involved studies of initial oxide compacting techniques, characteristics of pressure-bonded oxides, and various basic fuel-element geometries.

Progress Reported Previously

The evaluation of various types of uranium dioxide powders has been established through studies of their initial compacting and pressure-bonding characteristics. Of the oxides considered, the best characteristics for successful pressure bonding have been demonstrated by the mixtures of ceramic-grade and fused oxides. The effects of bonding parameters on the properties of Type 304 stainless steel cladding have also been defined. The results of these studies are summarized in BMI-1475. Preliminary process specifications for pressure bonding several basic Type 304 stainless steel-clad UO_2 fuel-element shapes have been partially developed. The program is now directed to the fabrication of fuel rods for irradiation testing in the VBWR.

Work This Period

The uranium dioxide and stainless steel tubing required to produce the fuel rods for irradiation in the VBWR have been ordered by the Atomic Products Division of the General Electric Company; however, none of this material has been received by Battelle. The compacting dies have been fabricated for both the UO_2 and stainless steel powders, using the established tubing tolerances. Pellets of UO_2 and stainless steel fabricated in these dies meet the specifications for loading into the ordered tubing.

Plans for Future Work

The preparation of several small-scale rods of the VBWR design will be fabricated upon receipt of the stainless steel tubing. These rods will provide basic deformation information required to pressure bond the full-scale assemblies.

DEVELOPMENT OF URANIUM CARBIDE (AEC-FUEL CYCLE)

F. A. Rough and W. Chubb

Uranium carbides and their alloys are being developed for the AEC as a part of the Fuel-Cycle Development Program. The objective of this program is to lower the cost of power produced from nuclear energy by lowering the costs of manufacturing, using, and reprocessing reactor fuels. Foremost among the various ways in which these costs can be reduced are: (1) lowering the cost of fabricating the fuel, (2) increasing the temperature at which the reactor coolant and fuel operate, (3) increasing the life of the fuel in the reactor, and (4) decreasing the cost of reprocessing the fuel. Uranium carbides seem capable of producing dramatic savings in connection with the second and third of these items, and the costs of fabricating and reprocessing of fuel elements using uranium carbides will probably be quite similar to the costs of fabricating and reprocessing fuel elements containing uranium oxide or uranium metal.

To assist in the accomplishment of the aforementioned objective, an integrated program of research on the preparation and properties of bulk uranium carbides is in progress at Battelle. This program includes: (1) development of skull-melting and casting techniques to provide a method for commercial production of cast uranium carbides in large quantities and sizes, (2) determination of the mechanical, physical, and chemical properties of uranium carbides at room and elevated temperatures, (3) measurement of the rates of diffusion of uranium and carbon in uranium carbides to provide a basic understanding of the behavior of these materials under a variety of different conditions at elevated temperatures, and (4) irradiations designed to produce basic information on the nature of radiation-damage processes in the structure and upon the integrity of uranium carbides. These studies have now been in progress for 2 years. During the next 6 months, efforts will be devoted primarily toward developing procedures and specifications for large-scale production of carbide castings, measurement of the high-temperature mechanical and physical properties of uranium carbides, and determination of the fundamental mechanisms causing damage to uranium carbides during irradiation in a nuclear reactor.

Melting and Casting Techniques for Uranium-Carbon Alloys

B. C. Boesser, E. L. Foster, and R. F. Dickerson

Objective

To develop techniques amenable to large-scale production of high-quality cast shapes of uranium carbide.

Program Approach

Skull-type arc-melting procedures are being applied to the melting and casting of uranium carbide. The effects of process variables such as the quality of the charge materials, the furnace atmosphere, the arc current and voltage, the grade of graphite used for the electrode tip, the skull size, and the mold configuration are to be investigated in the interest of producing high-purity castings with good compositional control. Evaluations of compositions and homogeneity are based on chemical analysis. Casting soundness is determined by radiography.

Progress Reported Previously

Casting research covering the first 1-1/2 years of this program is summarized in BMI-1370 and BMI-1488. Research in the several months since BMI-1488 was written has dealt with the effect of vacuum-outgassed commercial-grade graphite (used both as a charge addition and as an electrode tip) upon the control of the composition of the product. It was found that five castings produced using outgassed graphite varied less than ± 0.1 w/o carbon while castings produced from the same grade of graphite that had not been outgassed varied by ± 0.3 w/o carbon. Vacuum-outgassed AGOT reactor-grade graphite, used both as a charge addition and as an electrode tip, resulted in castings which varied by about ± 0.3 w/o carbon.

Work This Period

Seven replicate melts using outgassed TSF reactor-grade graphite have been completed. The carbon analyses of these castings are as follows:

<u>Casting</u>	<u>Carbon, w/o</u>
1	5.61
2	5.48
3	5.36
4	5.31
5	5.28
6	5.06
7	4.58(a)

(a) Analysis is being rechecked.

Observations made during this series of melts may help explain the variation in composition after analyses of the skull are obtained. It was noted (1) that the variation in carbon content between subsequent melts was small and (2) that electrode-tip erosion was minor. In the prior melts involving AGOT graphite, erratic and excessive electrode-tip erosion caused wide variations in the carbon content of the melts and, presumably, the skull. During the present series of tests involving TSF graphite, but the same skull as before, the carbon content of castings decreased as shown above with each successive melt, and it is believed that further melting would have resulted in both skull and casting compositions near the charge composition of 4.2 w/o carbon. However, further analyses are needed to elucidate the above trend of small, successive decreases in carbon content.

Plans for Future Work

The electrodes from the above melts will be weighed, and the skull will be analyzed for carbon and oxygen to obtain a carbon balance for these melts. A series of melts using helium or argon atmospheres will be made to determine the effect of the atmosphere on compositional control, casting purity, castability, and the amount of molten metal available from a given skull at a constant power input. Melts will also be made using commercial UC as a charge material. Also, a series of melts will be made using outgassed commercial graphite as a charge addition and as an electrode tip in an effort to duplicate the previous run which resulted in a carbon variation of only ± 0.1 w/o.

Mechanical and Physical Properties of Uranium Monocarbide

B. C. Boesser, R. W. Endebrock, E. L. Foster, and R. F. Dickerson

Objective

To obtain measurements of mechanical and physical properties such as strength, hardness, thermal conductivity, and resistivity at room and elevated temperatures and to determine the effect of alloy additions and heat treatments on the properties of uranium carbides.

Program Approach

Specimens of uranium monocarbide are to be prepared for measurements of certain mechanical and physical properties. Equipment is being modified to permit property measurements in the 1000 to 1600 C temperature range. Test environments will be limited to either vacuum or helium. In the interest of improving the properties of UC, research concerned with alloying, coating, and heat treating is also being performed.

Progress Reported Previously

Mechanical- and physical-property data obtained during the last 1-1/2 years are presented in two reports, BMI-1370 (August 21, 1959) and BMI-1488 (December 27, 1960). It has been found that the strength and hardness of uranium carbides in the as-cast condition are relatively insensitive to composition in the range of 4.8 to 9.0 w/o carbon. When heat treated to produce U_2C_3 , the 7.0 w/o carbon alloy has a hardness of about 1100 KHN as compared to about 700 KHN for UC and 500 KHN for UC_2 . Moisture adversely affects the strength and integrity of uranium carbides. Alloying with Mo_2C , NbC, VC, ZrC, and TiC results in improved strength, hardness, and corrosion resistance. A promising protective coating for UC appears to be a copper-20 w/o silicon alloy applied by dipping at 2450 F. Various heat treatments have been given to specimens with carbon compositions of 4.8, 7.0, and 9.0 w/o. Results of these heat treatments were described in BMI-1469.

A UC-20 w/o vanadium alloy has been prepared exhibiting phases identified by X-ray diffraction as UC, uranium, and vanadium. Various ternary alloys of UC with combinations of NbC, VC, vanadium, niobium, and molybdenum which have been prepared are to be evaluated in terms of electrical resistivity, corrosion resistance, and metallographic structure. Heat treatments of as-cast uranium-7.0 w/o carbon alloy specimens to determine the transformation rate to the U_2C_3 structure indicated that treatments of less than 15 min are required for transformation in the 1400 to 1700 C temperature range. Hot-hardness measurements from 1000 to 1500 C have been obtained for uranium-5.0 w/o and -9.0 w/o carbon alloys.

Work This Period

Hot-hardness measurements obtained on two 7.0 w/o carbon specimens and one 4.8 w/o carbon specimens are given in Table F-1. It is interesting to note that the hardness values of the 4.8 w/o carbon specimen are lower than those previously reported for 5.0 w/o carbon, indicating the need for additional replicate tests of hot hardness. Specimens were prepared for investigating the high-temperature thermal-expansion properties of alloys containing 4.8, 7.0, and 9.0 w/o carbon; a 5.0 w/o carbon specimen has been prepared for high-temperature thermal-conductivity measurements.

TABLE F-1. HOT HARDNESS OF URANIUM-CARBON ALLOYS

Temperature, C	Diamond Pyramid Hardness ^(a)		
	4.8 w/o C	7.0 w/o C, specimen 1	7.0 w/o C, specimen 2
1000	400	(b)	(b)
1100	100	300	100
1200	30	50	43
1300	16	17	37
1400	12	8	11
1500	10	2	7

(a) Hardness reported represents an average of five measurements at each temperature.

(b) Impressions were not readable at 1000 C because of the extreme hardness of the specimen and the low indenter load.

Plans for Future Work

Replicate hot-hardness measurements of uranium-carbon alloys containing 4.8, 7.0, and 9.0 w/o carbon will be obtained to determine the reliability of prior measurements. Preparations are being completed for obtaining high-temperature thermal-conductivity, electrical-resistivity, and thermal-expansion measurements on these alloys.

Studies of Diffusion in Uranium Monocarbide

W. Chubb, R. W. Getz, and F. A. Rough

Objective

To obtain the rates of interdiffusion of carbon and uranium and the rates of self-diffusion of uranium and carbon in uranium monocarbide.

Program Approach

Rates of interdiffusion are determined by measuring the rate of growth of the uranium carbide layers between graphite and liquid uranium saturated with carbon. Rates of self-diffusion of uranium in uranium monocarbide are determined by measuring the gamma activity of samples ground from a diffusion couple consisting of a foil of enriched uranium sandwiched between two rods of depleted uranium monocarbide. Rates of self-diffusion of carbon in uranium monocarbide are being determined by measuring the beta activity of carbon-14 in diffusion couples prepared from two rods of uranium monocarbide, one of which contains carbon-14.

Progress Reported Previously

Determination of the rates of interdiffusion of carbon and uranium has been completed; the results were reported as the equation $D = 148 \exp(-79,000/RT)$, and details of the procedure are given in BMI-1370. Determination of the rates of self-diffusion of uranium in uranium monocarbide has been completed; the results were reported as the equation $D = 0.034 \exp(-86,000/RT)$, and details of the procedure are given in BMI-1488. Preparations were made to evaluate the initial couple used to determine the rates of self-diffusion of carbon in uranium monocarbide. Details of the procedure are given in BMI-1473, BMI-1480, and BMI-1489.

Work This Period

The rates of self-diffusion of carbon in uranium monocarbide were investigated by annealing one couple for 60 min at 1500 C and another for 15 min at 1800 C. Metallographic examinations of the interface between the two lengths of uranium monocarbide of each couple indicated in both cases that the interface was free of contamination and that there was limited grain growth across the interface. Beta-activity measurements made on these couples and on the initial couple which was annealed for 15 min at 1600 C were used to calculate diffusion coefficients. The values of the coefficients for self-diffusion of carbon in uranium monocarbide obtained to date are presented in Table F-2.

TABLE F-2. DATA OBTAINED IN THE DETERMINATION OF THE RATE OF SELF-DIFFUSION OF CARBON IN URANIUM MONOCARBIDE

Temperature, C	Self-Diffusion Coefficient, cm ² per sec
1500	1.2×10^{-8}
1600	3.3×10^{-8}
1800	2.1×10^{-7}

An additional casting of uranium monocarbide containing carbon-14 is being prepared for use as half of each of several couples to be employed in the determination of the rates of self-diffusion of carbon in uranium monocarbide.

Plans for Future Work

Additional couples are to be annealed at 1200 and 1400 C in the determination of the rates of self-diffusion of carbon in uranium monocarbide.

Effects of Radiation on UC

D. G. Freas, A. E. Austin, J. E. Gates,
C. M. Schwartz, and F. A. Rough

Objective

To study and understand the important mechanisms of damage occurring in uranium monocarbide as a result of irradiation.

Program Approach

Specimens of uranium monocarbide (UC), uranium sesquicarbide (U₂C₃), and uranium dicarbide (UC₂) are being irradiated in the Battelle Research Reactor at surface temperatures ranging from 500 F or less to 1600 F and to burnups ranging from 0.01 a/o to 0.7 a/o of the uranium. The specimens are contained in six NaK-filled capsules. Evaluation of these irradiated specimens will be based on their dimensional stability and microstructural stability, and on measurements of their electrical resistivities and lattice parameters.

Progress Reported Previously

Specimens of hyperstoichiometric UC, composed of natural uranium plus 5.0 w/o carbon, were irradiated in Capsules 1 and 2 to burnups of 0.01 and 0.03 a/o of the uranium at surface temperatures below 500 F. Changes in the dimensions of these

specimens were found to be small, and only slight decreases in density were measured. The electrical resistivity increased from an unirradiated value of about 35 microhm-cm to about 90 microhm-cm at 0.01 a/o and 0.03 a/o burnup. Since the resistivity values do not show a hyperbolic-type increase with increasing burnup, it is believed that saturation of the lattice by displaced interstitial atoms and vacancies was attained during these short irradiations. X-ray diffraction studies of the crystal lattice of specimens from both capsules also indicated that saturation of displaced interstitial atoms and vacancies had occurred. A metallographic examination of specimens at both levels of burnup failed to reveal any structural changes. Electron-microscope examination of a specimen with 0.01 a/o burnup of the uranium disclosed no changes as compared to an unirradiated control specimen.

Capsule 4, which contained four uranium-5.0 w/o carbon specimens, two in the as-cast condition and two which had been heat treated to relieve casting stresses, and two heat-treated (transformed) specimens of uranium-6.7 w/o carbon, was discharged from the reactor after the detection of fission products leaking from it into the reactor coolant. The specimens in this capsule were all prepared from 10 per cent enriched uranium. It was estimated that these specimens had experienced a uranium burnup of approximately 0.1 a/o. The specimen-surface temperatures were intended to be in the 1400 to 1600 F range, but a high-temperature excursion occurred during operation as a result of temperature-control problems with the particular capsule that was used.

Work This Period

The irradiation of Capsule 3, which contained four uranium-5.0 w/o carbon specimens, two in the as-cast condition and two which had been heat treated to relieve casting stresses, and two heat-treated (transformed) specimens of uranium-6.7 w/o carbon has been completed. The postirradiation examination of specimens from this capsule was initiated about 1 month after discharge from the reactor. The examination of specimens from Capsule 4, which had been discharged earlier from the reactor, was initiated at the same time.

During opening of Capsule 4, it was found that a large hole had been melted through the lower portion of the inner stainless steel capsule as a result of overheating of the capsule. There was a complete loss of the NaK coolant. No intact uranium-carbide specimens were recovered from this capsule. A melted section of the capsule body, the remains of one unknown specimen, and a section believed to be a portion of the niobium specimen basket are being examined by metallographic techniques in an effort to determine the characteristics of this failure.

Upon opening Capsule 3, which was irradiated at less than 500 F, it was found that all of the uranium carbide specimens were intact and in good condition. Physical measurements of the specimens showed that only very minor dimensional changes had occurred during irradiation. Measurements indicated that only slight decreases in the density of the uranium-5.0 w/o carbon specimens had occurred. Other measurements indicated increases in electrical resistivity of the specimens of about 300 per cent as compared with preirradiation data.

Density measurements of the uranium-6.7 w/o carbon specimens indicated that a small increase in density may have occurred. Small increases in density would be expected if transformation to U_2C_3 occurred.* The preirradiation electrical-resistivity data on these specimens suggest that transformation had not occurred during the preirradiation heat treatment; however, the postirradiation electrical-resistivity measurements indicate an increase of about 400 per cent as compared to the preirradiation data. Metallographic and X-ray diffraction studies, presently being performed, will assist in evaluating these results.

*Rough, F. A., and Chubb, W., "Progress on the Development of Uranium Carbide-Type Fuels, Phase II Report on the AEC Fuel-Cycle Program", BMI-1488 (December 27, 1960), p 39.

The irradiation of the remaining two capsules, containing specimens of uranium-5.0 w/o carbon, uranium-6.7 w/o carbon, and uranium-8.5 w/o carbon alloys, is continuing in the BRR. These specimens all contain 10 per cent enriched uranium.

Plans for Future Work

Upon completion of irradiations of the two capsules now in the BRR, postirradiation examinations of the specimens will be undertaken to study the dimensional stability, microstructural stability, electrical resistivity, and lattice parameters.

GROWTH OF UO_2 SINGLE CRYSTALS (AEC-DR)

J. D. Jackson, C. A. Alexander, H. L. Goering,
and F. A. Rough

Objective

To prepare large single crystals of UO_2 .

Background

While a great deal of research has been and is being done on UO_2 , the fundamental physical behavior of stoichiometric and high-purity UO_2 remains to be determined. The present program is being supported by the AEC Division of Research with the aim of securing a limited quantity of high-purity single crystals suitable for such fundamental property determinations.

Program Approach

Various high-temperature sources are being evaluated to melt UO_2 (melting point about 2760 C) and to control its solidification to form large single crystals.

Progress Reported Previously

The following fusion methods have been investigated:

- (1) Electron-beam melting
- (2) Resistance heating
- (3) Arc melting
- (4) Plasma-jet impingement
- (5) Induction heating
- (6) Radiation heating from resistance element.

Most of the experimental program has employed the last three methods. The resistance furnace using a tungsten heating element appears to give the greatest promise for success, and the present objective is to improve the furnace to reach and maintain a temperature of 2800 C.

Work This Period

During March, effort was expended in improving the temperature capabilities of the tungsten resistance furnace. Particular emphasis was placed on determining the reasons for the asymptotic behavior of the power versus temperature characteristic of the furnace.

This behavior could possibly be explained by the Ramsauer effect, by which high-temperature gases become electrically conductive at potentials below their ionization potential. Helium, however, has never been observed to behave in this manner. Hence, helium was run as the furnace atmosphere under conditions similar to those under which asymptotic temperature-power characteristics had been observed in presence of argon.

In a run with a system helium pressure of 25 mm of mercury, it was observed that, at an estimated temperature of 2700 C, the power versus temperature plot indicated asymptotic behavior. Reduction of the system pressure to 0.4 mm of mercury at the fixed power setting resulted in a temperature rise to 2760 C in 2 min. The experiment then ended abruptly as the furnace shorted. It would appear from this run that thermal convection by the gas is sufficiently high even at 25 mm of mercury to cause the observed behavior. It is thought that electrical conductivity is not a factor. Hence, it is indicated that future runs must be conducted at submillimeter ranges of pressure in order to reach

G-2

the melting point of UO_2 . Since the vapor pressure of UO_2 at its melting point is considerably greater than this pressure it is concluded that a capped crucible must be employed to prevent the boiling of UO_2 which was reported last month in BMI-1504 from a previous vacuum run.

Plans For Future Work

Experiments will be continued to reach higher temperatures in the tungsten resistance furnace. The experiments will be run in vacuum using a capped crucible to prevent UO_2 losses. Modifications of existing shielding are planned to reduce radiation losses.

RADIOISOTOPE AND RADIATION APPLICATIONS (AEC-OID)

D. N. Sunderman

Three research programs are under way at Battelle as part of the program of the Office of Isotopes Development. The first is concerned with the development of technology necessary to the application of intrinsic radioactive tracers to industrial process control. In the intrinsic-tracer concept a radioactive tracer is added directly to a process stream in the same physical and chemical form as the element or compound whose concentration is to be controlled. As physical or chemical separation steps are performed upon this material the radioactivity level of the stream continues to be a measure of the concentration of the single species tagged with radiotracer. Since measurement of radioactivity is less strongly influenced by the chemical and physical composition of the stream, systems of simpler design may be used to monitor and control the industrial process.

The second program is a study of the influence of structural factors on graft polymerization by ionizing radiation. Graft polymerization has been demonstrated capable of modifying substantially the physical properties of polymeric materials, and broad interest is being given this process industrially. However, it is necessary to learn more concerning the influence of molecular structure upon the ability to form graft copolymers before the full capabilities of this process can be achieved. In this study a series of substituted methacrylate polymers is subjected to the ionizing radiation from cobalt-60, and the product is analyzed by electron paramagnetic resonance, chemical, mass-spectrometry, gas-chromatographic, and infrared techniques. Information is being obtained on rate of free-radical formation and decay as well as on the mechanism of the reaction induced.

The third program is directed toward the development and evaluation of safety performance standards for sealed radiation sources. There are presently no detailed formal standards to insure the safe performance of sealed radiation sources. The ever-increasing usage of these sources industrially has indicated a need for such standards both to provide for public safety and to encourage further application of these devices. Such safety performance standards should be sufficiently rigorous to assure that a source conforming to them would not constitute an unusual hazard under either normal or abnormal conditions of use. Establishment of definite criteria should also assist in creating an atmosphere that will encourage increased industrial utilization of such sources.

Use of Intrinsic Radioactive Tracers for Process Control

J. L. McFarling, P. Gluck, J. F. Kircher, and D. N. Sunderman

Objective

To develop the technology essential to the application of intrinsic radiotracers to the control of certain industrial chemical process operations.

Program Approach

Several potential intrinsic radiotracer control applications are being evaluated according to criteria established earlier in the program. One important phase of this evaluation is to demonstrate the application of intrinsic radiotracers on a laboratory-scale model of the system being studied. The particular application chosen for intensive study is the use of intrinsic radiotracers to control precipitation-filtration

purification operations. In conjunction with the experimental program, an evaluation of the possible hazards to consumers from use of intrinsic radiotracers for process control is also under way, using the steel industry as an example.

Progress Reported Previously

The removal of impurity iron from a nickel-refinery stream was the first intensively studied process. The use of iron-59 tracer as an aid in efficiently removing iron from a mixed cobalt-nickel solution has been demonstrated with the process model. It was shown that using radioactive iron could improve the accuracy and shorten the response time of the control system. It was also shown that there were no significant radioactivity contamination hazards in the proposed application.

The second process application studied experimentally was the use of manganese-54 to facilitate removal of impurity manganese from process water. Results showed good general agreement between the filtrate radioassay and chemical analysis, with the radioassay being considerably more rapid.

In the hazards-evaluation study, it was concluded that there would be negligible contamination of drinking water and food from steel products containing the maximum allowable level of radioisotopes.

Work This Period

The planned experimental runs using the process-model system to study manganese removal have been completed. The results of the final experiments followed closely those previously reported.

Experimental data from the last run completed appear in Table H-1. It can be seen from the table that there is some difference between the filtrate count observed during the run and a later recount of the acidified filtrate samples. This discrepancy is thought to be largely due to the uncertainty in the background count during the experimental run because of buildup of tracer activity on the walls of the counting cell.

The evaluation of technical hazards to sensitive industries using steel products containing radiotracer residues has been completed. The results indicate that such industries as those concerned with photographic film and nucleonics instruments are the most critical areas.

Plans for Future Work

The third process chosen to demonstrate radiotracer control of impurity removal is the use of cadmium-115m to aid in removing cadmium from zinc electrolytes. This process, which is widely used in electrorefining of zinc, involves the deposition of cadmium metal from solution by exchange with zinc dust. Initial work on this system has begun.

Graft-Polymerization Studies

F. A. Sliemers, E. Gulbaran*, W. B. Gager,
J. F. Kircher, and R. I. Leininger

Objective

To determine the influence of structural and compositional factors on the behavior of polymers subjected to gamma irradiation and to correlate the radiation-induced changes which occur with the ability of the polymer to form graft copolymers.

*I. A. E. A. Fellow.

TABLE H-1. EXPERIMENTAL DATA FROM STUDY OF MANGANESE REMOVAL USING MANGANESE-54

Initial manganese concentration in feed = 2.93 ppm.
 Initial manganese-54 count in feed = 23,000 cpm.

Sample	Elapsed Time Since Start of Run, min	Apparent Filtrate Activity During Run, cpm \pm σ	Filtrate Activity From Later Recount, cpm \pm σ	Apparent Manganese Concentration in Filtrate, ppm		
				By Chemical Analysis of Filtrate	Calculated From Activity During Run	Calculated From Activity Recount
1	175	5310 \pm 80	5625 \pm 27	0.71 \pm 0.01	0.675 \pm 0.038	0.715 \pm 0.032
2	200	7220 \pm 104	6570 \pm 29	0.785 \pm 0.01	0.922 \pm 0.050	0.836 \pm 0.037
3	240	5830 \pm 96	5125 \pm 26	0.62 \pm 0.01	0.745 \pm 0.043	0.653 \pm 0.029
4	265	4950 \pm 90	4130 \pm 25	0.50 \pm 0.01	0.630 \pm 0.038	0.525 \pm 0.024
5	335	450 \pm 49	385 \pm 11	0.034 \pm 0.01	0.057 \pm 0.006	0.049 \pm 0.003
6	375	295 \pm 55	210 \pm 10	0.011 \pm 0.01	0.038 \pm 0.006	0.027 \pm 0.002
7	400	180 \pm 53	85 \pm 10	>0.005	0.023 \pm 0.006	0.011 \pm 0.002

Program Approach

It is anticipated that a mechanism of radiation attack on the various polymer systems can be established. Polymers are irradiated in vacuo to various total doses. Free-radical site concentrations are determined both as a function of dose and as a function of time, using electron paramagnetic resonance (EPR) measurements. Vapor-phase chromatography and mass spectrometry are used to determine quantitatively the volatile fragments of the irradiation. Grafting techniques are employed to correlate free-radical site concentrations and grafted copolymer.

Progress Reported Previously

A number of polyalkylmethacrylates have been examined extensively. The data suggest that: (1) free-radical formation is accomplished by means of a whole or partial ester scission, (2) the size and configuration of the hydrocarbon tail of the ester group influence the efficiency of site formation, (3) site formation is accompanied, in some cases, by a scission on the polymer backbone, and (4) the amount of grafted copolymer increases with increased free-radical concentration. More recently, the work has been extended to other polymer systems.

Work This Period

The study of the effect of polymer structure on the efficiency of free-radical site formation has been continued. Site measurements versus dose have been made for poly-ter-butylmethacrylate, polyacrylic acid, and atactic and isotactic polypropylene. These results are listed in Tables H-2 through H-4.

Site formation with dose in poly-ter-butylmethacrylate is of the same order of magnitude as found in the sec- and isobutyl polymers, but is considerably higher than in the n-butyl polymer. Although the ease of removal of hydrogen to form free radicals follows roughly in the ratio of 33:3:1 for tertiary:secondary:primary, apparently the proximity (to the carboxyl group) of the removable hydrogens must also be a factor. This would explain why more free radicals are formed by the ter-butyl than by the n-butyl polymer and why more are formed by the sec-butyl than by the isobutyl polymer.

The polyacrylic acid was prepared by irradiation of the monomer in bulk (no diluent was used). Irradiation of the polymer shows that the efficiency of site formation is greater than for any of the methacrylic acid esters or for polymethyl acrylate. A similarly high efficiency was noted earlier for polymethacrylic acid. Again, the proximity of the hydrogen to the carboxyl group probably accounts for the differences.

The atactic and isotactic fractions from a commercial polypropylene were prepared by extracting the polymer with n-heptane for 24 hr. The extractable material is, for the most part, atactic polymer. Sites-versus-dose data for the two fractions, although scanty, reveal no marked differences. Neither forms sites as readily as the polymethacrylates. Additional data will be obtained on these materials.

The volatile products from the irradiation of polycyclohexylmethacrylate and polyacrylic acid have been measured quantitatively by means of mass spectrometry and vapor-phase chromatography. Analysis of the data from these studies has not been completed.

Plans for Future Work

During April, the anticipated program of work will include: (1) analysis of the volatile fragment data obtained for polycyclohexylmethacrylate and polyacrylic acid, (2) initiation of an investigation of site formation in polyisopropenyl acetate, polymethacrolein, and polymethacrylamide, and (3) volatile-product analyses of poly-ter-butylmethacrylate and polymethacrylic acid. Following completion of the

TABLE H-2. SITES VERSUS DOSE FOR POLY-TER-BUTYLMETHACRYLATE

Dose, rads	Sites Per Monomer Unit x 10 ³ (a)
1.8 x 10 ⁵	0.56
4.6 x 10 ⁵	1.27
9.2 x 10 ⁵	2.60
3.1 x 10 ⁶	7.17
4.8 x 10 ⁶	9.31
9.2 x 10 ⁶	10.30
1.8 x 10 ⁷	9.53
2.8 x 10 ⁷	5.84

(a) By EPR measurements.

TABLE H-3. SITES VERSUS DOSE FOR POLYACRYLIC ACID

Dose, rads	Sites Per Monomer Unit x 10 ³ (a)
2 x 10 ⁵	0.50
5 x 10 ⁵	2.06
1 x 10 ⁶	2.36
3.4 x 10 ⁶	9.16
4.8 x 10 ⁶	10.5
1 x 10 ⁷	13.4
2 x 10 ⁷	20.5
3.3 x 10 ⁷	23.1

(a) By EPR measurements.

TABLE H-4. SITES VERSUS DOSE FOR ATACTIC AND ISOTACTIC POLYPROPYLENE

Dose, rads	Sites Per Monomer Unit x 10 ³ (a)	
	Atactic	Isotactic
2 x 10 ⁵	N.M.(b)	--
5 x 10 ⁵	0.11	N.M.
1 x 10 ⁶	N.M.	0.037
3.5 x 10 ⁶	N.M.	0.12
4.8 x 10 ⁶	0.21	--
6 x 10 ⁶	--	0.13
1 x 10 ⁷	N.M.	0.40
3 x 10 ⁷	--	0.79

(a) By EPR measurements.

(b) Concentration of sites not measurable from EPR spectrum.

volatile-product analysis of the ter-butylmethacrylate polymer, a paper will be written covering that portion of the work relating to the four polybutylmethacrylates.

Development and Evaluation of Safety Performance
Criteria for Sealed Radiation Sources

C. W. Townley, R. B. Price, J. M. Fackelmann,
C. L. Selander, R. A. Ewing, and D. N. Sunderman

Objective

To develop and evaluate safety performance criteria for sealed radiation sources.

Program Approach

During Phase I of this program, sealed radiation sources and their applications are being surveyed to accumulate information on current source design, fabrication techniques, composition, and applications. Upon evaluation of these data, safety performance criteria and a classification scheme will be established, and appropriate test procedures selected and developed to measure performance against these standards.

Progress Reported Previously

Questionnaires were sent to all organizations in North America known or believed to manufacture sealed sources. A similar questionnaire was sent to manufacturers of devices containing sealed sources.

Current regulatory status was investigated by contact with Federal, state, and municipal agencies. The status of efforts by advisory technical groups and insurance and standards associations with interests in radiation safety was surveyed to establish the current state of efforts in this area.

Preliminary studies were initiated to develop performance criteria for sealed sources and to determine the applicability of standard test procedures employed in other industries.

Work This Period

Two-thirds of the questionnaires sent to sealed-source manufacturers and one-third of those sent to device manufacturers have been returned. The information received on source design, fabrication techniques, composition, testing procedures used, and applications is being compiled. Letters were sent to the companies that have not responded urging them to return the questionnaire.

A meeting of prominent sealed-source and device manufacturers and the Office of Isotopes Development and Division of Licensing and Regulation of the Atomic Energy Commission was attended on March 1, 1961, in Washington, D. C. The purpose of the meeting was to explain the safety performance criteria program to the manufacturers and to obtain their cooperation in the conduct of the program. In general, it was indicated that the manufacturers were willing to cooperate and would furnish the information necessary for the conduct of the program.

The Nuclear-Chicago Corporation and Minnesota Mining and Manufacturing were visited to obtain more detailed information on source designs, fabrication techniques, current testing procedures, and failure-experience data than that furnished by replies to the questionnaires. The Underwriter's Laboratories in Chicago, Illinois, was visited to discuss problems in testing and developing performance criteria.

The search for currently available standard test procedures which might be applicable to the testing of sealed sources has been continued.

Plans for Future Work

More visits are planned to a number of source and device manufacturers to obtain more information about source technology, current testing procedures, and previous failures. When sufficient information in these areas has been accumulated, safety performance criteria and a source-classification scheme will be established, and the applicability of potential test methods will be evaluated.

JOINT U.S. -EURATOM PROGRAM

H. M. Epstein and D. L. Keller

Studies concerned with obtaining information on void-content and heat-transfer relationships in forced-convection two-phase flow in light-water systems and with the development of uranium mononitride as a possible fuel material are under way for the Joint U.S. -Euratom Research and Development Board. Both programs are directed toward obtaining data that will be useful in efforts to achieve more economical nuclear power.

The most important property of two-phase flow for the reactor designer is the boiling void distribution since the amount and distribution of the vapor will have a major effect on the reactor kinetics as well as on the critical dimensions. Since no general correlation of the void distribution has been proven reliable over the wide range of reactor variables, data from the study of boiling void distribution will be interesting to those working in this field.

Uranium mononitride is a high-melting-point compound possessing a particularly high uranium density. As a dispersion fuel in stainless steel its behavior during irradiation is similar to UO_2 . However, little is known about its irradiation resistance in bulk form or about practical methods of fabricating the material into dense bodies. Therefore, the program on uranium mononitride is concerned with establishing the basic feasibility of this material as a reactor fuel. It will include an evaluation of most promising methods of fabricating UN, the determination of several physical and mechanical properties of UN, and the preparation, irradiation, and evaluation of UN bodies (both clad and unclad) of three different enrichments.

VOID-DISTRIBUTION AND HEAT-TRANSFER STUDIES

R. O. Wooten, F. G. Peter*, J. J. Foglia,
H. M. Epstein, and J. W. Chastain

Objective

To investigate axial void distribution and heat transfer for rectangular coolant passages over a range of experimental conditions of interest in light-water reactors.

Program Approach

A positive-displacement pump is used to circulate distilled water in a closed, pressurized loop consisting of piping, test section, preheater, condenser, sump, and accumulator. The void distribution in the test section is measured using a beta-ray attenuation technique. The experimental void fractions are correlated in terms of the significant heat-transfer parameters such as mass flow rate, fluid temperature, heat flux, slip ratio, and quality.

Progress Reported Previously

A correlation of the Bankoff flow parameter, K , in terms of the Reynolds number was developed, and tests were made which confirmed the behavior of the correlation at low Reynolds numbers (45,000).

Several runs taken at pressures around 900 psi to re-examine the variation in the slip ratio indicated that the slip ratio exhibits a scatter of ± 25 per cent with a-c power on the test section.

*Euratom scientist.

A 40-kw direct-current motor generator was installed to investigate the influence of a-c versus d-c power on the test section.

Work This Period

The experimental program was concluded, and a topical report is being prepared.

Plans for Future Work

The topical report will be issued when completed.

DEVELOPMENT OF URANIUM MONONITRIDE

Powder-Metallurgy Studies

S. J. Paprocki, D. L. Keller, J. Stoecker*, and E. O. Speidel

Objective

To develop a method for fabricating high-density uranium mononitride bodies suitable for property measurements and irradiation testing.

Program Approach

Cold pressing and sintering, isostatic hot pressing by the gas-pressure-bonding technique, and rotary swaging are the primary fabrication techniques being investigated.

Progress Reported Previously

Little densification of UN powder by vacuum sintering has been effected at temperatures low enough to prevent decomposition of the UN. At apparent temperatures of 1800 C and above, a free-uranium liquid phase occurs.

Rotary swaging of pellets in stainless steel cans consolidates the powder up to 85 per cent of theoretical density, but no interparticle bonding is achieved.

The amount of densification achieved by isostatic hot pressing of UN powder for 3 hr at 10,000 psi is related to the temperature of pressing. Up to 87 per cent of theoretical was obtained in stainless steel cans at a temperature of 1290 C, 93 per cent was obtained in niobium at 1425 C, and 95 per cent in molybdenum at 1480 C. The dense cores pressed in molybdenum were extensively worked, caused possibly by thermal-expansion differences between UN and molybdenum.

Uranium mononitride is not nearly as susceptible to attack by atmospheric moisture as is UC.

Work This Period

Two 95 to 96 per cent dense specimens suitable for irradiation testing have been fabricated by isostatic hot pressing for 3 hr at 1480 C and 10,000 psi in niobium cans. The niobium can as well as the reaction zone between the UN and niobium have been removed by grinding on a diamond wheel.

A small amount of an as yet unidentified phase is dispersed throughout the specimen. This phase is not continuous but occurs as nodules at the UN grain boundaries. The porosity of the specimen seems predominantly associated with this phase. Selective etching failed to identify it. A piece of the dense material analyzed 5.44 w/o nitrogen, which is somewhat lower than the 5.57 w/o nitrogen content of the starting powder. This second phase appears to be nonmetallic and is apparently unaffected by a 6-hr vacuum anneal at 1650 C.

*Euratom scientist.

Uranium nitride powder consolidated by rotary swaging in Type 304 stainless steel before being isostatically hot pressed for 3 hr at 1290 C and 10,000 psi produced cores measuring 13.7 g per cm³ or about 96 per cent of the UN theoretical density. A very deeply pitted surface occurs when the stainless cladding is ground off. Microscopic examination shows that the cores are not 4 per cent porous as expected, but instead contain extrinsic phases which might have been liquid. A liquid phase does occur during vacuum annealing at 1650 C.

Some values for the solubility of hydrogen in UN at a 1-atm pressure have been obtained in a micro-Sievert apparatus and are shown in Table I-1. The sample was three pellets of pressed UN powder which had been vacuum sintered 1 hr at 1600 C. The total weight of the 74 per cent dense pellets was 4.4 g.

TABLE I-1. SOLUBILITY OF HYDROGEN IN UN AT VARIOUS TEMPERATURES UNDER A 1-ATM HYDROGEN PRESSURE

Temperature, C	Solubility, ppm (± 3)
300(a)	15 ^(a)
350	20
400	20
450	18
500	16
800	9

(a) Not at equilibrium.

Equilibrium was not reached after 7 days at 300 C. Times of about 20 hr at 400 C and 2 hr at 500 C were necessary to obtain equilibrium. After these measurements the sample was allowed to absorb 14 ± 3 ppm hydrogen as measured by the apparatus. Vacuum-fusion analyses of the three pellets showed hydrogen contents of 12, 16, and 16 ppm. The data did not permit the calculation of diffusion coefficients.

Plans for Future Work

Irradiation specimens are being made by isostatic hot pressing in niobium cans at 1480 C and 10,000 psi. The can will be removed and the specimens ground to 0.3 in. in diameter and 3/4 in. in length. Those to be clad will be reloaded in 15-mil-thick Type 304 stainless steel cans with 1/8-in. end plugs and hot pressed at 1150 C to effect intimate contact between the cladding and the core.

Melting and Casting Studies

R. W. Endebrook, J. Bugl*, E. L. Foster, and R. F. Dickerson

Objective

To produce by melting and casting techniques a limited number of dense UN bodies to be used for irradiation tests and for the determination of physical and mechanical properties.

*Euratom scientist.

Program Approach

The conversion of uranium metal to the mononitride in a nitrogen atmosphere is the basis of an arc-melting process under study. Calculations from existing thermochemical data indicate that the UN system is sensitive to changes in pressure and temperature above and below the melting point of UN. Consequently, the program requires that pressure relationships must first be established for the production of sound bodies, and then bodies must be prepared for irradiation studies and physical-property determinations. A high-pressure melting technique appears to be a requisite in this program. Results of experiments are evaluated in terms of the degree of UN stoichiometry and homogeneity attained.

Progress Reported Previously

Reaction of uranium and nitrogen is virtually complete after about 10 min of arc melting in nitrogen at a 3-atm pressure. The nitrogen content of buttons was shown to vary widely with arc amperage and, therefore, temperature. By reducing the arc amperage at the end of the melt the nitrogen content of the button was raised approximately 15 per cent. An investigation of the effect of pressure showed that the nitrogen content varied from 3.15 w/o in a button produced at 1/4 atm to 4.82 w/o in one produced at 3 atm. Logarithmic extrapolation of the data indicated that stoichiometric material probably can be produced by direct arc-melting procedures at about 10 atm of nitrogen pressure. However, exploratory tests in the 10 to 24-atm pressure range and using the same power conditions as tests made at 3 atm did not increase the nitrogen content of buttons significantly above that of buttons prepared at 3 atm. As pressure was raised, the distance from the electrode to the melt became less, the melting area became smaller, and smoke identified as 70 to 80 per cent UN₂, 10 to 15 per cent UN, and 5 to 10 per cent of a phase believed to be tungsten nitride, completely obscured the arc.

Further investigations of furnace variables have indicated that placing insulation around the arc zone and hearth improves stoichiometry of buttons somewhat. A high-frequency arc maintained during direct-current melting as an aid in arc stabilization and gas ionization proved to be of no value for improving the nitrogen content of a button. Attempts to produce a single-phase alloy of UN and UC at a 1/2-atm nitrogen pressure by introducing enough carbon to react with the uranium phase did not eliminate the second-phase uranium.

Work This Period

As indicated previously, logarithmic extrapolations of pressure-composition curves indicated that melting of uranium under less than 10 atm of nitrogen should yield stoichiometric UN. By melting UN powder rather than uranium under 10 atm of nitrogen, consolidation might be expected without decomposition or generation of UN_x smoke. Consequently, a 50-g compact of UN (5.47 w/o nitrogen) was prepared and melted under 10 atm of pure nitrogen at 325 amp and 42 v.

The arc was visible for 2 min after which there was a rapid accumulation of smoke which obscured the arc. After 3 min the smoke cleared, and the arc was clearly visible for the remainder of the 5-min melt cycle. The button was doughnut shaped, with very little material remaining at the center. Much of the original powder compact had broken up into particles that were not consolidated during melting. Material taken from the center of the button was found to contain 4.75 w/o nitrogen; samples taken 3/8 to 1/2 in. from the center were found to contain about 5.2 w/o nitrogen and 2.2 w/o tungsten.

During the experiment there was considerably less nitride smoke formed than in the case where uranium was melted under 10 atm of nitrogen. Moreover, the composition of the smoke differed in that the major phase as determined by X-ray diffraction

was about 60 to 80 per cent U_2N_3 , a second component was about 15 per cent UN_2 , and a third component was about 10 to 20 per cent of a phase believed to be tungsten nitride.

The results of this experiment suggest that the arc was intensely hot because of (1) the insulating effect of the high-pressure nitrogen and the smoke and because of (2) the nearness of the electrode to the button. The central portion of the button where the heat was concentrated decomposed into nitrogen and uranium, part of which vaporized and part of which became second-phase material in this portion of the button. That portion of the button which was further from the center was rich in nitrogen and was diluted with tungsten from the electrode.

The effect of nitrogen pressure on arc length indicates the importance of arc potential on melting characteristics. Consequently, the remaining efforts during the month were devoted to increasing the arc potential. Although initial attempts to form buttons by reacting uranium under 10 atm of nitrogen at 300 amp and 100 v were limited to melting cycles of less than 2 min, results have been encouraging in that uniform melting of buttons has occurred and has produced buttons with a nitrogen content of 5.0 w/o. Arc distances of more than 1 in. were attained, and very little melting of the tungsten electrode was observed.

Plans for the Future

More research on the effect of arc potential is planned. In addition, additives, such as hydrogen in the nitrogen atmosphere or tin in the uranium charge, are to be investigated as aids for producing a longer, more stable arc.

Irradiation of UN Specimens

P. B. Shumaker, G. E. Raines, and J. H. Stang

Objective

To investigate the integrity of UN bodies irradiated to high burnups in capsules designed to minimize specimen temperatures in-pile.

Program Approach

Six noninstrumented capsules are being prepared for irradiation of UN specimens containing depleted (approximately 0.3 per cent uranium-235) and natural (approximately 0.7 per cent uranium-235) uranium and uranium enriched approximately 5 per cent. The capsules will be irradiated at the Materials Testing Reactor for approximately 6 months in thermal-neutron fluxes selected to provide specimen-effective fluxes of approximately 1×10^{14} nv. This exposure will produce in the specimens (1) uranium-235 burnups of approximately 60 per cent, (2) a fairly wide range of fractional burnup of all the uranium atoms originally present, and (3) a fairly constant amount of matrix hydrogen as a result of the (n,p) reaction with nitrogen. The combination of (2) and (3) is desired to gain semiquantitative information relative to irradiation-damage effects owing to hydrogen accumulation in the UN.

Progress Reported Previously

A capsule design for the irradiations was prepared. Each system will consist essentially of a 1-1/8-in.-OD aluminum shell containing two NaK-immersed UN specimens; each UN matrix is 0.30 in. in OD by approximately 0.75 in. long. Internal dosimeter wires will be present in the specimen zone of each capsule, and, in addition, removable dosimeter wires will be attached to the outer capsule surface. The latter will be removed periodically as the irradiations progress to obtain data for estimating specimen temperatures and burnup levels. Heat-transfer criteria indicate that the

maximum specimen-surface temperature (for 5 per cent enriched specimens in a flux of 1×10^{14} nv) will be approximately 750 F.

Initial data from a laboratory experiment involving the exposure of UN bodies to NaK at 500 F yielded no visible evidence of incompatibility after 1000 hr. A 900 F experiment was in progress.

Work This Period

Parts for the six capsules in the program were fabricated and plans for the first four irradiations were completed. The specimens in these four capsules will be fabricated by powder metallurgy. Each of the two specimens in an individual system will have the same enrichment. One specimen will be clad with approximately 15 mils of Type 304 stainless steel; the other will be unclad. The actual loading contemplated is as follows:

<u>Capsule</u>	<u>Uranium-235 Content in Specimens, per cent</u>	<u>Anticipated Burnup Level, per cent of total uranium atoms originally present</u>
1	0.72	0.33
2	0.72	0.33
3	0.30	0.14
4	0.30	0.14

It is likely that the specimens in the fifth and sixth capsules will also be fabricated by powder metallurgy. These will be enriched to 5 per cent.

After 1500 hr of operation, the 900 F UN-NaK compatibility heat was terminated and the system inspected. As was the case for the 1000-hr exposure at 500 F, no evidence existed of physical changes of the UN bodies as a result of the exposure. Spectrographic analysis of various residues remaining in the capsule gave no indication of NaK attack on the UN.

Plans for Future Work

The first four capsules of the program will be loaded about mid-April; all will be sent to the MTR by May 1. According to the present plan, the fifth and sixth capsules will be loaded in late April or early May if specimens become available.

MATERIALS DEVELOPMENT AND EVALUATION (HAPO)

F. R. Shober

Research and development is being conducted on materials to be used in conjunction with construction of the NPR (New Production Reactor) and operation of the PRTR (Plutonium Recycle Test Reactor) at the Hanford Atomic Products Operation. The creep properties of Zircaloy-2 at elevated temperatures are being studied in an effort to determine the feasibility and advisability of using Zircaloy-2 for process-tube material. Changes in thermal and electrical properties of UO_2 resulting from irradiation and fabrication techniques are important in relation to the operational characteristics of the PRTR. Also in support of the PRTR program studies to develop a metallic container material for the HAPO fused chloride-electrolytic fuel-recovery process are in progress. These three studies, although not directly related, contribute to the over-all program at HAPO.

Mechanical Properties of Zirconium Alloys

J. A. Van Echo

Objective

To obtain creep and creep-rupture data, including long-time creep (25,000 to 30,000 hr or longer), on annealed and cold-worked Zircaloy-2 sheet material and to evaluate the effect of stress and temperature cycle on the creep properties of this material.

Program Approach

In order to prevent excessive contamination of the specimens, all tests are made in vacuum of the order of 0.5μ of pressure. Standard Battelle creep machines and furnaces are used for making these tests. Temperatures of interest include 550, 650, and 750 F. Optical creep measurements are taken daily.

Progress Reported Previously

This is a long-term program. Twenty tests are now in progress for various test times ranging from a few to 24,000 hr. As data are accumulated, they are reported. Generally, there are only small changes in creep results from one month to the next. Therefore, except for incremental increases in creep deformation, most of the data have been reported previously in BMI-1489.

Work This Period

Seventeen of the tests listed in BMI-1489 were continued through March. One test (Specimen Zr-A-33) was previously reported as discontinued in January and two tests (Specimens ZR-A-18 and Zr-A-27) were discontinued in March. The latter two tests were on annealed Zircaloy-2 at 550 F. They were discontinued because the minimum creep rates were reported as essentially zero over the past several thousand hours. It was believed that no additional information could be gained by continuing these two tests.

Two long-time creep tests were started during March. These two tests are on specimens taken from an annealed section of a PRTR pressure tube and are being run at 550 F and 15,000 and 12,500 psi.

Plans for Future Work

The 20 creep machines will continue to be occupied with tests on annealed and cold-worked Zircaloy-2 sheet. These tests are summarized as follows:

- (1) Two creep tests on 15 per cent cold-worked sheet at 650 and 750 F
- (2) Ten creep tests on annealed sheet at 550, 650, and 750 F
- (3) Five creep tests on annealed sheet at 550 and 650 F
- (4) Three tests on annealed PRTR pressure tube at 550 F.

No additional work is planned other than continuing these 20 tests in progress during the month of April.

Thermal Conductivity and Electrical Conductivity of UO₂

H. W. Deem and C. F. Lucks

Objective

To measure the thermal and electrical conductivity of irradiated and unirradiated UO₂.

Program Approach

An apparatus based on a steady-state, absolute, thermal-conductivity method has been assembled for the thermal-conductivity measurements of the irradiated UO₂. Existing Battelle equipment is being used for the thermal- and electrical-conductivity measurements on unirradiated material.

Progress Reported Previously

The assembly of a thermal-conductivity apparatus, its calibration, and the measurements of four control UO₂ specimens have been summarized in BMI-1448 (Rev). Included in the report are data on the thermal conductivity of an irradiated UO₂ specimen with a burnup of 0.005 a/o. Also included in the summary are data on the electrical conductivity of the four control specimens.

Tentative thermal-conductivity data have been reported for an irradiated UO₂ specimen having a burnup of 0.016 a/o in BMI-1455.

Final thermal-conductivity data have been reported for a sintered UO₂ disk specimen, identified as Fabrication Specimen A, in BMI-1489. Interpolated values at 200, 800 and 1400 C are 0.067, 0.034 and 0.025 w/(cm)(C), respectively for 100 per cent theoretical density.

Techniques have been worked out for making thermal-conductivity measurements on vibrationally compacted powder UO₂ specimens.

Work This Period

The thermal-conductivity apparatus has been made ready again for measurements. Thermocouples have been repaired and at room temperature seem to be working properly.

The electrical-resistivity apparatus has been completed. First cycle measurements have been completed on a 1/4-in. -diameter by 3-in. -long sintered UO₂ specimen irradiated to a burnup of 0.005 a/o and designated by HAPO as Specimen 10. The data have not been reduced for reporting.

Specimen preparations for thermal-conductivity measurements on an unirradiated 3-in. -diameter UO₂ disk, Specimen D-L, are in progress.

Plans for Future Work

During the next work period measurements will begin on a 0.061 a/o burnup specimen similar to Specimen 10 and on the UO₂ disk, Specimen D-L. An additional

electrical-resistivity cycle will be run on Specimen 10. A single-crystal UO_2 specimen will also be prepared for measuring.

Materials of Construction for the
Fused Chloride-Electrolytic UO_2 Process

E. L. White, P. D. Miller, D. C. Drennen, C. H. Lund,
A. M. Hall, and W. K. Boyd

Objective

To develop a metallic container material for the HAPO fused chloride-electrolytic fuel-recovery process.

Program Approach

Several commercially available metals and alloys were screened as candidate materials of construction by using a bench-scale apparatus employing chlorine to produce uranyl chloride from uranium oxide. At present, a series of experimental iron- and nickel-base alloys, designed for this application on the basis of the results of the screening program, are being evaluated by weight changes during exposure to a 750 to 800 C equimolar solution of sodium and potassium chlorides containing U_3O_8 and sparged with chlorine.

Progress Reported Previously

Corrosion results were reported in BMI-1504 for 14 experimental alloys.

Work This Period

A second series of experimental-alloy compositions is being planned. This series will explore more completely the nickel-aluminum and nickel-silicon systems.

Several corrosion evaluations were made primarily to confirm results obtained earlier. Qualitative agreement of corrosion rates was found.

In addition to the experimental alloys, two additional materials were evaluated this month. It was found that the durability of Duranickel was only fair. An alloy 91 w/o copper and 9 w/o aluminum showed poor resistance.

Plans For Future Work

A second series of experimental-alloy compositions will be chosen and heats will be cast and evaluated.

COATED-PARTICLE FUEL MATERIALS (AEC-DRD)

R. W. Dayton and J. H. Oxley

This program is being carried out to determine the characteristics of various coated-particle fuels and to develop optimum materials for specific reactor applications. Various combinations of fissionable-core, protective-coating, and matrix-support materials are to be evaluated in terms of fission-product retention as a function of burnup and operating temperature.

There have been five tasks outlined within the scope of this development program. Tasks I, II, and III concern the optimization of the full capabilities of the three most promising coating materials, alumina, and pyrolytic carbon. Investigation of other possible coatings will be carried out during Task IV, while Task V will involve the evaluation of coated-particle materials produced commercially.

The immediate objective of Task I of this program is to evaluate fully a fuel material containing approximately 125- μ uranium dioxide particles coated with 40 μ of alumina. Limited evaluation of irradiated material of this type from the Sanderson & Porter program has been authorized and is in progress. In addition, preliminary experimental studies are being carried out on carbon-coated fissile materials. These studies are currently aimed at developing coating methods and studying the radiation behavior of the materials.

MATERIALS DEVELOPMENT PROGRAMUncoated Fuel-Particle Studies

G. W. Cunningham and S. J. Paprocki

Objective

To characterize and evaluate fuel particles of UO_2 , UC, and UC_2 for use in coating studies and in subsequent coated-fuel irradiation program.

Program Approach

Fuel materials are purchased as required from commercial vendors and are characterized with respect to enrichment, composition, particle size, porosity, shape factors (sphericity, roundness, and surface roughness), hardness, crushing strength, phase structure, grain size, fraction of particle agglomerates, surface activity ratio, xenon release ratio, and chemical impurities. As the relative importance of the various factors becomes known, emphasis will be increased or decreased on these factors.

Progress Reported Previously

This program was initiated in March.

Work This Period

The fuel material of primary concern is UO_2 , and at the present time emphasis is being placed upon those factors which may be considered as directly affecting the coating process. The most important factors are diameter, density, sphericity, roundness, surface roughness, hardness, crushing strength, and fraction of particle agglomerates. Suitable techniques are being evaluated and developed when necessary for measuring these properties.

Screen analyses are used to obtain approximate values for particle diameter, and metallographic measuring techniques are being used to obtain a more exact value for the average particle size and the size distribution. Both techniques are well established and require no development. Quantitative metallographic techniques are being developed for measurement of sphericity, roundness, surface roughness, and particle agglomeration. Sphericity is a measure of whether a particle is a spheroid. It is defined as the

ratio of the minor to major axis times the roundness. The roundness is defined as the ratio of the minimum radius of curvature at any section of the projected surface to the radius of curvature for a circumscribed circle. The values would range from unity for a perfect sphere to zero for particles with sharp corners. Values can be readily obtained by using a grid on a low-power binocular microscope. Since roundness is a gross effect and not concerned with slight surface irregularities, it is not necessary to measure the exact radius of curvature. In general, the particles will either contain sharp corners or will have a radius of curvature which can be estimated from the ratio of the minor to major axis. Particle agglomeration is also measured under the low-power binocular microscope (50X) and is reported as the number of agglomerates per million particles and is further broken down according to the number of particles in the agglomerates.

Hardness is measured by standard techniques on a Tukon hardness tester. Crushing strength is a measure of the load required to crush a single particle. Thus, the value is not only a function of the particle strength but of the particle size, and unless a large number of particles are evaluated the value may be subject to significant error. However, it appears that 5 to 10 particles represent a sufficiently large sample to provide a useful value for purposes of comparison.

Density and porosity measurements are extremely difficult to perform on high-density powders and no satisfactory inexpensive method has been developed. At the present time, a combination of xylene pycnometer measurements to determine density and petrographic measurements to determine porosity are being used with good agreement.

Plans for Future Work

Techniques will be investigated for measuring surface roughness and density. In addition, measurements of interest to the particle coating studies will be made on the materials of interest.

Alumina-Coating Studies

M. F. Browning, V. M. Secrest, and J. M. Blocher, Jr.

Objective

To establish conditions and equipment designs for coating fuel particles with Al_2O_3 of optimum quality.

Program Approach

Alumina is being deposited on UO_2 particles by hydrolysis of Al_2Cl_6 vapor in a fluidized bed. The coatings are being evaluated by microscopic examination, measurement of physical properties, alpha counting, and fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

A general summary of the experimental work on Al_2O_3 -coated particles prepared prior to this program was given in BMI-1471. Modifications of the fluidized-bed reactor design and water-vaporization system have improved process control and have extended the operating time to permit deposition of the desired coating thickness without interruption. The following specific types of coated fuel particles were prepared for in-pile evaluation:

- Dense Al_2O_3 on dense UO_2
- Dense Al_2O_3 on porous UO_2
- Dense Al_2O_3 over porous Al_2O_3 on porous UO_2 .

However, only the first type had satisfactory fission-gas retention after neutron activation.

Work This Period

Major effort this month has continued to be the preparation evaluation of satisfactory specimens of the types described above for in-pile.

Although the dense Al_2O_3 coatings on dense UO_2 particles were acceptable in the as-prepared condition, postirradiation fission-gas release after incorporation in a graphite matrix became excessive. An effort is being made to improve this situation by elimination of the agglomerated UO_2 particles which resulted in potentially weak coated particles. Agglomerates in the UO_2 were successfully eliminated by vigorous fluidization.

An evaluation of the products prepared at coating temperatures between 900 to 1150 C has indicated the temperature variable to be quite critical in relation to the type of coating deposited. At a coating temperature of 900 C a frosty translucent-to-opaque coating is obtained, while at a temperature of 1150 C a transparent coating is deposited. Coatings previously deposited at temperatures of 1000 C were of intermediate character. X-ray diffraction and electron-microscope data have indicated that the coatings may consist of alpha Al_2O_3 crystallites in an amorphous matrix. The degree of crystallization appears to increase with deposition temperature, as might be anticipated. Because of the apparent criticality of the coating temperature, an effort is being made to improve temperature control and measurement.

The effort to upgrade the sample of dense Al_2O_3 coating on porous UO_2 powder (Lot 717B) as described last month in BMI-1504 by a thermal cycle and oxidation treatment followed by leaching to remove poorly coated particles was not successful. The postirradiation xenon-133 release of the treated material was not improved. Since there is considerable doubt that the porosity of the UO_2 used in Lot 717B significantly differs from that used in Lot 715A (dense Al_2O_3 on dense UO_2) no further studies will be carried out with this material.

In an effort to determine the condition for preparing a satisfactory dense-over-porous Al_2O_3 coating, the deposition of porous coatings at higher temperature was evaluated. In addition, heat treatment of porous Al_2O_3 coatings prior to deposition of the dense Al_2O_3 coating was studied. A composite coating of a 1000 C deposit over a 700 C deposit cracked when heat treated at a temperature of 1260 C for 48 hr in helium, but to a lesser extent than did previous duplex coatings. A specimen was prepared with a dense Al_2O_3 coating deposited at a temperature of 1000 C over a porous coating deposited at a temperature of 700 C. The porous coating was heated for 48 hr at a temperature of 1260 C in helium prior to deposition of the dense coating. In a preliminary evaluation of this composite coating, no cracking resulted from the heat treatment at a temperature of 1260 C. This possible solution to the cracking difficulty will be investigated more thoroughly. If the initial stability results are substantiated and the inner layer retains significant porosity, a sample for in-pile evaluation will be prepared to determine the possible value of the porous inner layer.

Plans for Future Work

Evaluation of the Al_2O_3 -coated enriched UO_2 batches will be continued. Experimental work to further improve the uniformity and quality of the Al_2O_3 coating will be carried on concurrently. Temperature and coating rate are the variables of immediate interest.

Carbon-Coating Studies

A. C. Secrest, V. M. Secrest, M. F. Browning,
and J. M. Blocher, Jr.

Objective

To establish conditions for coating fuel particles with pyrolytic carbon of optimum quality.

Program Approach

Pyrolytic-carbon coatings are being applied by the decomposition of hydrocarbons in a tumbling-bed reactor at temperatures above 1500 C, as well as in a fluidized bed of particles at temperatures below 1500 C. The coatings are being evaluated by means of microscopic examination, measurement of physical properties, direct alpha counting, and by fission-gas release on postirradiation heating. Promising materials will be evaluated by in-pile tests at elevated temperatures.

Progress Reported Previously

In a previous program, pyrolytic-carbon coatings were applied to UC, UC₂, and UO₂ by the pyrolysis of hydrocarbons in a fluidized bed of the powders at temperatures up to 1500 C. Promising coatings have been obtained, but there was some indication of the mechanical failure of the coatings at temperatures above the coating temperature.

In the present program the fluidized-bed reactor design for coating temperatures under 1500 C was altered to eliminate the occurrence of poorly coated particles due to inadequate fluidization. Precoating the particles with a thin layer of carbon and transferring them to a clean reactor was initiated for the purpose of minimizing contamination of the coating with urania dust. Carbon-coated UO₂ particles have been prepared which exhibited a postirradiation xenon-133 release of only 1.6 ppm of the maximum theoretical value at 1300 C for 130 min.

Preliminary results have indicated that both deposition rate and coating temperature affect the occurrence of either an apparent laminar or columnar structure.

A second phase of the present program has been the use of a 1.5-in. -diameter tumbling-bed reactor for coating particles with carbon at temperatures between 1500 and 1750 C. The products from this reactor had coatings of nonuniform thickness, and a considerable fraction of the particles adhered to the walls of the barrel. This work has been deferred in favor of concentrating on the development of the carbon coatings deposited at temperatures below 1500 C.

Work This Period

Two lots of enriched UC₂ spheres, 150 to 250 μ , were coated at a temperature of 1400 C with pyrolytic carbon in a fluidized bed of the particles for in-pile evaluation. One lot (918A) had a 50 to 60 μ laminar carbon coating while the other lot (919A) had a 50 to 60 μ columnar carbon coating. These specimens have exhibited sufficiently low alpha activity to merit a postirradiation fission-gas-release evaluation. This is now in progress, and the results should be available early in April.

One lot (916A) of natural UC₂ spheres, 100 to 200 μ , was coated with 50 μ of columnar carbon at a temperature of 1400 C. This material had a postirradiation fission-gas-release at 1300 C for 110 min of 3.0 ppm of the maximum theoretical xenon-133.

A lot (905A) of depleted UO_2 coated with 36μ of laminar carbon at a temperature of 1400 C was subjected to an additional postirradiation fission-gas-release evaluation at a higher temperature than the 1300 C previously used. This specimen exhibited fission-gas release of only 0.2 ppm xenon-133 at 1500 C for 120 min.

Plans for Future Work

Evaluation of the carbon-coated enriched UC_2 specimens will be continued. Establishment of optimum conditions for depositing both dense and porous coatings at temperatures below 1500 C in a fluidized bed will also be continued. The effect of temperature and coating rate on coating structure will be the major interest.

Carbon-Matrix Studies

M. C. Brockway, D. J. Bowers,
and W. H. Duckworth

Objective

To prepare and characterize graphite-matrix bodies containing coated fuel particles.

Program Approach

For initial tests, bodies which are representative of current graphite technology are desired. Specimens are to be prepared with the minimum of development studies. The development work is to be carried out only to the extent needed to achieve reasonable physical properties, reproducibility, and fuel dispersion. Later, improved properties and fabrication techniques will be sought.

Progress Reported Previously

Processing conditions were established for unfueled specimens made from AGOT graphite flour and medium-hard coal-tar pitch. The conditions which yielded the highest density, 1.65 g per cm³, then were used in an investigation of methods to disperse Al₂O₃-clad depleted UO₂ particles in graphite.

Two general methods were investigated for dispersing fuel. In one procedure, the coated fuel particles were introduced into one composite batch of flour and pitch during the benzene-solvent mixing step; in the other specimen, quantities of fuel and premixed flour and pitch were dry mixed together just prior to molding each specimen.

Characterization data were reported in BMI-1504 for specimens containing Al₂O₃-clad depleted UO₂ particles dispersed by the first method outlined above. A series of specimens containing Al₂O₃-clad enriched UO₂ particles was prepared by the second of the dispersion methods.

Work This Period

The specimens with the depleted coated fuel, characterized in BMI-1504 were neutron activated. Upon subsequent heating to a temperature of 1370 C, the specimens had a xenon-133 fractional release of 62 ppm. Comparing this with a release of 25 ppm for the undispersed coated fuel, there appears to have been no significant increase in the release, indicating no fuel-coating damage during incorporation of the coated fuel into the graphite matrix. This coated fuel was dispersed at the solvent-mixing step in the fabrication of the fueled-graphite specimens.

Specimens prepared last month containing dense Al₂O₃-clad enriched UO₂, which were dispersed in specimen-quantity mixtures just prior to molding, also were neutron activated. Upon subsequent heating to a temperature of 1370 C, these specimens had a xenon-133 fractional release of 170 ppm. A comparison of this release to the 3-ppm value for undispersed fuel strongly suggests that coating damage occurred during incorporation of the coated fuel into the graphite matrix.

To determine whether the damage to the coated enriched fuel was due to the method of fuel dispersion or to the greater susceptibility of this particular fuel sample to damage, two additional groups of specimens were prepared. One group contained a sample of the original coated depleted fuel, and the other coated enriched fuel. Both groups were processed in the same manner and contained the same volume fuel loading. The method of dry mixing the coated fuel with the matrix powder was that which involves dispersing the coated fuel in specimen quantities prior to molding. This mixing method is preferred to the solvent-mixing single-batch method because it provides specimens that are precisely fueled and have close dimensional tolerances. To reduce the chance

of fuel-coating damage during molding, the molding pressure was lowered from 20,000 to 2,000 psi, and the resulting baked unfueled matrix density was 1.61 g per cm³. Baking temperature was the same as used previously, i. e. 1260 C.

The specimens containing both types of fuels were then neutron activated. Both types of dispersed coated fuels exhibited high xenon release ratios. The enriched sample had a release ratio of 200 ppm and the natural sample a value of 800 ppm. It, therefore, appears that dry mixing of coated fuel and premixed matrix powder in specimen quantities results in damage to the coated fuel.

Plans for Future Work

Further use of the solvent-mixing method of fuel dispersion will be explored. Modifications which will improve the precision of specimen-to-specimen fueling will be sought. This mixing method will be used instead of the method involving specimen quantities of fuel and matrix powder. Dispersion specimens will be prepared containing carbon-coated UC₂ fuel.

FUEL IRRADIATION PROGRAM

W. H. Goldthwaite, D. N. Sunderman, and R. F. Dickerson

Sweep Capsule SP-5

G. E. Raines, C. W. Townley, S. D. Beck, and R. J. Burian

Objective

To obtain preliminary fission-gas-release information on alumina-coated uranium dioxide particles in a graphite matrix.

Program Approach

Fission-gas-release data on a preliminary sample of alumina-coated UO_2 particles is to be determined by periodic sampling during irradiation and by a variety of post irradiation fission-gas-release experiments. These data are correlated considering various release mechanisms and phenomena. The postirradiation examination includes metallographic inspection, X-ray studies, and strength tests.

Progress Reported Previously

Two 1.5-in. -diameter graphite spheres fueled with about 10 w/o of highly enriched 127- μ -diameter UO_2 particles coated with about 40 μ of Al_2O_3 were included in an experiment which was terminated in November, 1960, after a burnup of about 6 a/o of the uranium-235 at a temperature level between 510 and 730 C. A summary of the experimental results from this capsule for the in-pile fission-gas-release behavior of the specimen containing alumina-coated particles (No. 471) and the graphite specimen fueled with uncoated UC particles (E8-7) was presented in BMI-1480. A correlation of the fission-product-release data from the coated-particle specimen with respect to the disintegration constants of the various species and a discussion of plausible release mechanisms were reported in BMI-1496. The spheres were undamaged after irradiation but exhibited greater contraction and loss of weight than was expected. One sphere was compression tested and the other sectioned for metallographic examination.

The compression-tested sphere was tested to an ultimate load of 4520 lb and a total deflection of 0.0725 in. The straight-line load-deflection curve had a slope of 69,000 lb per in.

Two specimens were sectioned from the other irradiated sphere and examined metallographically to reveal particle condition after irradiation. Cracks were observed in the alumina coatings of about 8 to 11 per cent of the particles. Almost all of the cracked particles were situated between the midradius and the surface of the sphere. Detailed results of these examinations are presented in BMI-1496 and BMI-1504.

Work This Period

The principal effort during this period has consisted of continuing the postirradiation studies. An unidentified phase was observed in the region between the UO_2 and the Al_2O_3 coating of several of the particles in the two metallographic specimens prepared from irradiated sphere No. 471. A similar material was observed in one or two of the UO_2 fuel particles in an unirradiated specimen. The nature of this phase will be investigated.

The sphere from which the metallographic specimens were obtained was also sectioned to obtain a specimen for particle separation and collection. The specimen was cut from a sphere radius and was approximately 3/16 in. square by 3/4 in. long. The particles on the cut surfaces were removed by electrolysis in chromic acid with the specimen as the anode. It is believed that the particles were virtually unaffected by the separation technique since the process caused the graphite to sluff off in almost colloidal-size particles. The alumina-coated fuel particles were separated from the specimen by increments along the radius by controlling the depth of immersion in the electrolyte.

Plans for Future Work

The separated particles from each radial increment will be divided into smaller samples and subjected to examinations, which will include heat-treatment and fission-gas-release studies, designed to differentiate the behavior of fuel particles with cracked coatings from that of those with apparently undamaged coatings. Acid-leach tests are planned to confirm the degree and distribution of the cracked coatings, and metallographic examinations and compression tests will be performed. A final correlation of the in-pile release data will be undertaken considering these factors. The Al_2O_3 coating will be studied by X-ray diffraction techniques to determine if any changes in the structure occurred as the result of fission-fragment attenuation. The nature of the unidentified phase observed in the irradiated and unirradiated fuel particles will be investigated and if possible identified.

Static Capsules CFP-1A, -1B, -1C, and -1D

R. J. Burian, N. E. Miller,
W. J. Zielenbach, and R. H. Barnes

Objective

To determine the effects of reactor radiation on alumina-coated uranium dioxide particles as a function of temperature of irradiation, flux, matrix support, and particle porosity.

Program Approach

The fission-product-retention properties of UO_2 fuel particles coated with Al_2O_3 may be improved by using various combinations of high- and low-density UO_2 and Al_2O_3 . It is planned to irradiate two types of fuel particles, as described below, both supported in a graphite matrix and also unsupported by any matrix material. The combinations of coated fuel contemplated for study in this set of experiments are:

- (1) Dense UO_2 particles coated with dense Al_2O_3
- (2) Low-density UO_2 particles coated first with low-density Al_2O_3 and then with dense Al_2O_3 .

Four static capsule experiments are planned for irradiation in core positions in the BRR. Each type of particle will be irradiated in each capsule experiment, both in a graphite matrix and in an unsupported condition. The specimens in the four capsules will be irradiated at temperatures of about 150 C and 985 C to burnups of approximately 2 and 6 a/o of the uranium. Evaluation of the specimen performance will be achieved by pre- and postirradiation examinations which will include measurement of physical dimensions and weights, visual examination, metallographic examinations, and strength tests. In addition, the quantity of fission gases released from each specimen will be determined. Dosimeter wires within the capsule will be analyzed to obtain flux data; and selected specimens will be analyzed for uranium burnup.

Progress Reported Previously

The capsule design has been completed and is described in BMI-1489 and BMI-1496. The three types of fuel particles have been prepared, and development of techniques for incorporating them in a graphite matrix was initiated.

Work This Period

Incorporation of the fuel particles in a graphite matrix is in progress and is described in this report under the heading "Carbon-Matrix Studies". Preparations have been made to perform the preirradiation examination of these specimens when they become available.

Plans for Future Work

When the specimens become available, the preirradiation examinations will be performed and the specimens encapsulated. Irradiation in the BRR will begin as soon thereafter as possible.

Sweep-Capsule CFP-S-2

N. E. Miller, G. E. Raines, T. S. Elleman, S. D. Beck, and R. J. Burian

Objective

To obtain preliminary fission-gas-release information on pyrolytic carbon-coated uranium dicarbide particles supported in a graphite matrix.

Program Approach

The fuel specimen is contained in a sweep-gas capsule, and the fission gas released by the specimen is continually removed from the capsule during the irradiation by a helium sweep gas. Periodic analysis of the sweep gas is made to determine the fission-gas-release characteristics of the fuel as a function of burnup.

Progress Reported Previously

The design of the capsule for this experiment was presented in BMI-1496.

Work This Period

A thermal mock-up of the capsule, in which the fuel specimen was replaced with a small electrically heated element, was operated. The temperature data from this experiment agreed well with those of the heat-transfer calculations and indicate that the capsule design should be useful over the temperature range of approximately 870 C to 1200 C with approximately 50 watts of fission heat and less than 1 kw of auxiliary electrical heat.

Plans for Future Work

Under the present schedule this capsule irradiation is anticipated to begin during May. The installation of the out-of-pile equipment at the BRR will begin during April.

Sweep Capsule CFP-S-3

N. E. Miller, G. E. Raines, T. S. Elleman, S. D. Beck, and R. J. Burian

Beam-Tube Loop Studies

J. B. Melehan and F. A. Rough

Objective

To modify the beam-tube facility so that coated-particle fuels can be irradiated at temperatures up to 2200 C under conditions in which the release of fission products can be studied continuously as a function of time, temperature, and burnup.

Program Approach

As a part of an investigation of irradiation effects on refractory fuels, an in-pile loop for the continuous study of fission-product gas release during irradiation has been developed. This apparatus consists essentially of a helium gas-purification system, a beam-tube furnace for operation in an 8-in. beam tube of the BRR, and a fission-product trapping system. The system operates by flowing purified helium over the specimen while it is heated by an induction furnace in the beam tube. Released fission gases are separated from the helium carrier gas outside the reactor for gamma-ray spectrometric analysis. Because of problems encountered in insulating the induction furnace leads, the high-temperature potential of the system has not been fully realized. This equipment is being modified to improve the insulation of the induction leads and coil and to meet the specific needs for continuous in-pile testing of coated UO_2 and UC_2 particles at temperatures up to 2200 C. It is planned that glass or Vycor insulation will be developed for the leads and coil, and the in-pile furnace shielding will be redesigned or modified to incorporate it. Additional problems associated with this development are the measurement of temperatures and the selection and design of refractory components in the hot zone.

Progress Reported Previously

Experiments were begun in the evaluation of induction-furnace design for electrical and thermal efficiency, durability, and material compatibility. The basic features of the present furnace design were outlined in BMI-1504. The induction furnace was operated intermittently for a total of 24 hr at 1650 C. The power limit of the 5-kw r-f power unit had not been exceeded, and still higher temperatures could have been attained. However, alumina and Mullite thermal insulators which enclose the induction susceptor suffered numerous longitudinal and transverse fractures as the result of large temperature gradients and thermal stresses.

Work This Period

It has already been mentioned that the principal problem encountered during in-pile operation of the r-f induction furnace has been the failure of induction coil and coil leads as the result of high-voltage electric arcing. Use of several rubber and plastic insulating materials has not been successful in solving this problem. Glass and vycor, although their brittleness is a drawback, appear to be two of the more promising candidates as electrical insulators. Their electrical properties are not significantly changed by fast-neutron and gamma irradiation until irradiation dosages are in the range of 10^{19} and 10^{20} nvt. Also, annealing of damage in glass can be expected at elevated temperatures. Consequently, a Vycor electrical insulating cover was fabricated to contain the induction coil. Similarly, the induction-coil leads are to be enclosed in Vycor tubing, the objective being to enclose in a continuous glass insulating cover that part of the induction coil and the coil leads which is located in the region of highest neutron and gamma-ray flux. An out-of-pile test of this modification of the coil and its insulation is presently being prepared, although a direct indication of its in-pile behavior will require operation under actual in-pile conditions.

It appears that the best possibility for solving the problem of high-temperature thermal insulation for the susceptor is to design segmented insulators so as to avoid the high thermal stresses which develop in massive single-piece insulators. Enclosure of the induction susceptor in small ceramic insulating pellets or loosely compacted insulating powder is an approach which merits some consideration. While alumina is an effective thermal insulator to 1650 C, higher temperature ceramics, such as zirconia, will be required for thermal insulation above 1650 C. Zirconia is not only more stable mechanically and chemically, but it retains its electrical insulating properties above 1900 C.

Plans for Future Work

Future work will continue in the solution of the problems of electrical and thermal efficiency of the induction furnace, electrical insulation of the induction coil to prevent failure by electrical arcing, and improvement of mechanical stability of ceramic thermal insulating components. These efforts will be made with the objective to extend the furnace capability to above 1650 C.

Particle-Separation Studies

R. A. Ewing and T. S. Elleman

Objective

To develop techniques for the separation of coated-fuel particles from graphite matrices for postirradiation examination.

Program Approach

Chemical methods for the separation of coated-fuel particles from graphite matrices are being investigated, including disintegration of the graphite matrix with boiling fuming nitric acid and electrolysis. Efficiency of separation is evaluated by visual observations. Estimates of the freedom from physical damage and extent of chemical damage are based primarily on the amount of uranium dissolved during the separation.

Progress Reported Previously

It has been shown that Al_2O_3 -coated UO_2 particles can be separated from a graphite matrix by boiling in fuming nitric acid or by application of an electrolytic disintegration technique. The electrolytic method is convenient to use but, like nitric acid disintegration, results in some dissolution of exposed UO_2 . In evaluating coated-particle systems, it would be desirable to recover exposed particles intact, and experiments have been carried out to determine if this can be accomplished with the electrolytic technique. The use of dilute sulfuric acid, chromic acid, or sodium chromate electrolytes resulted in lower uranium oxide dissolution than was observed with concentrated electrolytes but the quantity dissolved was still higher than desired. Since very low rates of dissolution are obtained with these dilute electrolytes on uncoated UO_2 particles, evidently some other factor accounts for the high uranium dissolution observed during actual matrix disintegrations.

Work This Period

A possible cause of uranium oxide dissolution during electrolysis is electrolytic attack upon the coated particles. This possibility was investigated by an electrolysis experiment using separated Al_2O_3 -coated UO_2 particles similar to those in graphite matrices. Dissolution of uranium was very low, comparable to the uranium dissolution obtained by leaching. It is concluded that the causes of the losses of uranium lie in the fabrication of the particle-containing matrix, rather than in the electrolysis.

Plans for Future Work

No additional work is planned.

Fission-Gas Analytical Studies

R. L. Ritzman and T. S. Elleman

Objective

To develop improved techniques for the analysis of fission-gas mixtures.

Program Approach

The measurement of fission-gas-release rates from in-pile fuel specimens requires accurate sampling of the offgas and a suitable method for analyzing the fission gases. Using the gamma-ray spectrum-analysis method, it is necessary to know the size of the gas sample collected, the detection efficiency of the gamma-ray spectrometer for the various fission-gas gamma rays, and the accuracy with which the gamma-ray spectrum can be resolved into contributions from individual fission gases. The present program is directed toward the improvement and expansion of current techniques for fission-gas analysis. The retention time of fission gases on charcoal traps will be measured to determine the maximum sampling times in collecting gas samples. Fission gases with gamma-ray photopeaks in the energy range from 80 to 230 keV will be

standardized in the gamma-ray spectrometer to check the presently used efficiency values for this energy range. New techniques for resolving complex gamma-ray spectra will be studied as possible replacements for present methods.

Progress Reported Previously

Previous progress reports have discussed the construction of new sampling traps for fission gases and the use of these in gathering helium-elution data for krypton and xenon from charcoal traps at liquid-nitrogen and dry-ice temperatures. Also, a 4-pi beta and gamma-ray standardized solution of iodine-133 provided known concentrations of xenon-133 and xenon-133m for determining the gamma-ray spectrometer detection efficiencies of these radionuclides on charcoal traps. Construction of additional equipment was reported for use in the experimental portion of the development of new techniques for resolving complex krypton and xenon fission-gas gamma-ray spectra.

Work This Period

A krypton and xenon fission-gas mixture, produced by irradiating uranium foil, was separated by gas chromatography and the fractions adsorbed on separate charcoal traps. The traps were scanned repeatedly for several days with a multichannel gamma-ray spectrometer. The spectrometer scans were divided into a number of energy increments. The contribution of each fission gas (in terms of counts per minute) to each energy interval has been determined in part at this time.

Plans for Future Work

The remaining energy interval contributions will be determined for each fission gas. Once the counting rate for each radioisotope has been measured for each energy range, the information can in principle be applied to resolving unknown fission-gas spectra into the counting-rate contributions from each fission gas. The accuracy of this technique will be studied.

SUPPORTING RESEARCH PROGRAM

Physics and Engineering Guidance

R. O. Wooton, H. M. Epstein, and J. W. Chastain

Objective

To investigate the importance of effects of coated fuel particles on the physics and engineering characteristics of reactor systems which may utilize these coated particles.

Program Approach

Reactor systems previously designed by other laboratories are selected and examined to determine the effects of substituting coated fuel particles for the given fuel. Parameters investigated are coating material, coating thickness, particle size, fuel and moderator loading, and reactor size. The calculation techniques are based on the simplest analysis available consistent with the program objectives. Other miscellaneous problems for which solutions will assist the total program are also considered.

Progress Reported Previously

A literature survey was completed to assist in evaluating the amount of fission-product release which can be tolerated in a system without interfering with the normal reactor maintenance. The effects of Al_2O_3 fuel-particle coatings on the reactivity of a graphite-moderated pebble-bed reactor (PBR) were reported.

Work This Period

A preliminary analysis was made to examine the nuclear-criticality problems involved in designing a chemical reactor of roughly cylindrical geometry which could be used to coat 125- μ -diameter fuel particles with Al_2O_3 , or carbon. The preliminary investigation indicated that an analysis based on simple thermal-neutron diffusion theory would probably not yield any trustworthy information. Plans were made to examine one or two cases by using the VALPROD machine code with a MUFT III input. The results of this study will tell how to best proceed with the rest of the investigation.

Plans for Future Work

A criticality analysis will be made to determine the maximum sized chemical reactor of cylindrical geometry which can be used to coat 125- μ -diameter fuel particles with Al_2O_3 , or carbon. Also, a study will be started to assemble information on weight percentage fuel loadings which will be required for reactors presently designed.

Fission-Gas-Diffusion Studies

R. H. Barnes, T. S. Elleman, and D. N. Sunderman

Objective

To determine diffusion coefficients in the absence of radiation for rare-gas fission products in alpha alumina, pyrolytic carbon, and other potential coating materials for nuclear fuel particles.

Program Approach

Small particles of the coating material of interest are dispersed in uranium dioxide powder and irradiated at low temperatures in the Battelle Research Reactor. These particles, which contain fission products that recoiled into them from the surrounding uranium dioxide powder during irradiation, are separated from the powder and heated at constant temperature in a flowing helium stream. The fission gases released from the particles during heating are collected from the helium on activated charcoal at liquid-nitrogen temperature and radioassayed. The results of the measurements are then used to determine fission-gas diffusion coefficients.

Progress Reported Previously

Diffusion coefficients for xenon-133 in Linde Company 1-mm-diameter alpha alumina spheres at temperatures ranging from 800 to 1500 C were reported in BMI-1504.

Work This Period

Measurements were made of xenon-133 release from a vapor-deposited coating of 60 w/o alpha alumina-40 w/o gamma alumina on Linde Company 1-mm-diameter alpha-alumina spheres at temperatures ranging from 900 to 1500 C. Releases ranging from about 1.5 to 75 per cent were observed for heating periods of 4 to 5 hr. The release data were erratic and cannot be interpreted in terms of diffusion coefficients. Since new alumina coatings with better characteristics have been prepared, the work on this material, which also consists of coated Linde Company 1-mm-diameter alpha-alumina spheres, is currently being evaluated and will be used for diffusion studies if the coating is similar to that which is to be used for actual fuel particles.

Pyrolytic carbon particles were prepared for diffusion studies. These particles, which are nearly spherical in shape with an average diameter of about 1 mm, were formed by vapor depositing pyrolytic carbon on 500- μ coke spheres. A sample of this material has been irradiated in contact with uranium dioxide powder and is ready for xenon-133 diffusion measurements.

Plans for Future Work

Next month, xenon-133 diffusion measurements will be made using the pyrolytic-carbon particles and uncoated Linde Company alpha-alumina spheres.

Further diffusion experiments are to be made using alpha-alumina spheres with a pure-phase alpha-alumina coating formed by vapor deposition. Other studies will be performed to measure krypton-85 diffusion from alpha-alumina and pyrolytic carbon, and to measure xenon-133 release during postirradiation heating from alpha-alumina and pyrolytic-carbon spheres irradiated in contact with uranium dioxide for about 8 weeks in a thermal-neutron flux of 1×10^{13} nv.

Radiation-Damage Studies

T. G. Knorr and C. M. Schwartz

Objective

To study fission-recoil damage in nonfissionable ceramic materials.

Program Approach

Decoration procedures are being investigated to allow observation of the fission tracks with optical and electron microscopical techniques. Alpha Al_2O_3 will ultimately be the material studied, but the alkali halides will be investigated initially because of ease of handling and the information already available.

Progress Reported Previously

NaCl, KBr, and LiF were chosen for the initial studies in the alkali halides. Fueling techniques were developed where UO_2 was suspended in a plastic solution which was then allowed to dry directly on the cleavage faces of the crystals. Surface damage is being studied by means of high-resolution direct carbon replicas (preshadowed with platinum) observed by transmission electron microscopy.

Work This Period

Various types of radiation damage have been observed in NaCl and KBr. In any one area of either crystal the damage is uniform and ranges in dimensions from about 600 A to more than 15,000 A. Regions of damage smaller than 600 A have been recorded but are not consistent and have not been characterized as yet. Long, thin, elevated regions are observed in both materials. These tracks correspond to a fission-particle path with a low angle of approach to the crystal surface. Similar but longer tracks have been observed in surfaces of UO_2 and other oxides.* Irregularly shaped

* Noggle, T. S., and Stiegler, J. O., "Electron Microscope Observations of Fission Fragment Tracks in Thin Films of UO_2 ", J. Appl. Phys., 13, 2199 (1960).

Bierlein, T. K., and Mastel, B., "Damage in UO_2 Films and Particles During Reactor Irradiation", J. Appl. Phys., 13, 2314 (1960); and HW-61656 (September 9, 1959).

Bierlein, T. K., and Mastel, B., "Fission Fragment Damage in Nonfissionable Thin Films", J. Appl. Phys., 13, 2315 (1960).

plateaus have also been seen in both crystals, similar but smaller in size than those reported elsewhere.* Pits have been observed in both NaCl and KBr in regions far removed from the plateaus but of similar structure and size. Another structure, which seems to be connected with the presence of the plastic holding the fuel, is present only in the KBr. The damage produces irregularly shaped hills of the order of a few thousand Angstroms which contain two types of structure; first, there is evidence of shorter elongated tracks due to low-angle fission particles, while, second, there appear fairly large craters within these hills which contain the various types of damage previously seen in KBr but in smaller sizes. Apparently the particles producing these tracks are of lower energy. Effects of the plastic have been seen on the NaCl surfaces which are considerably different in size and structure and are easily recognized. These regions show no consistency with the fueling, and are presently being discounted on this basis. No neutron damage has been seen.

Fuels of various enrichments are being used as well as different irradiation times to change damage doses. The plastic-fuel charges were predried on glass microscope slides before placement on the crystal faces for one irradiation. The results suggest that a further investigation into this technique would be profitable to characterize the effects of the plastic.

Optical microscopy has yielded no positive information, and is being temporarily abandoned because of the more favorable results obtained with the electron microscope.

Plans for Future Work

Replicas from a set of LiF irradiations will be studied. New fueling methods are to be tested to try to improve the fuel-to-surface contact. Optimum conditions would result in consistent observation of tracks at all angles to the surface. Vacuum evaporation of a few Angstroms of fuel directly on the surface seems most promising. Alpha Al_2O_3 crystals are prepared and will be irradiated and studied. Optical decoration of damage in AgCl crystals will also be attempted.

Fuel-Compatibility Studies

G. W. Cunningham, D. C. Carmichael, J. J. Ward, and S. J. Paprocki

Objective

To investigate reactions which may affect the maximum operating time or temperature of coated fuel particles.

Program Approach

The stability of Al_2O_3 -coated UO_2 particles in an oxidizing atmosphere will be studied by measuring weight changes and by determination of changes in oxygen content of the UO_2 in the temperature range of 1500 to 1800 C. Specimens will be examined by metallographic and X-ray diffraction techniques and the results compared to weight changes obtained on a recording microbalance. Initial studies on the Al_2O_3 -carbon reaction will involve the use of standard diffusion couples. Later studies will require the measurement of gas pressures. Both resin-bonded and pitch-bonded carbons as well as pyrolytic carbon will be reacted at temperatures of 1000 to 1800 C. Diffusion of uranium through pyrolytic carbon coatings on UC, UC_2 , and UO_2 particles will be investigated by annealing coated particles of fully enriched uranium compounds at temperatures of 1000 to 1800 C for various times. The distribution of uranium in the carbon coating will then be determined by a combination of metallographic, autoradiographic, and alpha-counting techniques.

* Billington, D. S., and Crawford, J. H., Radiation Damage in Solids, Princeton University Press (1961), pp 303-4; Unpublished work of Jahoda, Siegel, and Kernohan.

Progress Reported Previously

Particles of UO_2 with a 53- μ -thick coating of Al_2O_3 and an overcoating of 120- μ -thick pyrolytic carbon showed no indication of reaction after 4 hr at a temperature of 1800 C in vacuo. Fully enriched UC powder coated with 55- and 100- μ coatings of pyrolytic carbon was heated at a 1400 C temperature level for 2 hr in vacuo.

Work This Period

The reaction between UO_2 and carbon coatings to form UC or UC_2 is being investigated by heating the coated particles in vacuum. Particles of UO_2 with 38- μ -thick coatings of vapor-deposited carbon have been heated at a temperature of 1760 C for 2, 4, and 8 hr in graphite boats and for 2 hr in an alumina boat. For direct comparison, particles with 7- μ -thick coatings were also heat treated at a temperature of 1760 C for 2 hr in alumina and graphite boats. Preliminary metallographic examination has been made of all of these samples.

Both the samples and the graphite boat showed a slight gain in weight, whereas a loss in weight would be expected for reaction. The slight gain (5 mg) is probably due to volatilization of impurities on the heater tube or radiation shield. In the metallographic preparation, considerable pullout occurred and the extent, if any, of the reaction zone cannot be determined until sample preparation is completed. However, comparison of the samples coated with 38 μ of carbon run for 2, 4, and 8 hr at a temperature of 1760 C suggest that little or no reaction occurred. The UO_2 visible in the metallography specimens shows no reaction products.

The UO_2 particles with only a 7- μ -thick carbon coating which were run in a graphite boat were severely reacted after 2 hr at a temperature of 1760 C and only a few nonreacted particles were detected. X-ray diffraction of the reacted sample revealed UC, UO_2 , and a smaller amount of UC_2 . The sample run in the alumina boat was even more severely attacked, and X-ray diffraction indicated the presence of UO_2 , UC, UC_2 , and uranium metal. In this case, the reaction probably proceeded more rapidly due to attack of the carbon coatings by the Al_2O_3 boat.

An investigation of the reaction between Al_2O_3 and carbon is being conducted using massive couples of these materials and using UO_2 particles coated with Al_2O_3 and overcoated with pyrolytic carbon. Particles of this type have been heated in vacuum at a 1800 C temperature level for 8 hr, and visually no apparent change in the particles was noted. These samples will be examined metallographically and compared to samples heated for 2 or 4 hr which showed no reaction as reported previously.

Massive couples of high-purity Al_2O_3 in contact with reactor-grade graphite and pyrolytic carbon have been prepared and diffusion annealed under slight pressure at temperatures of 1200 and 1400 C for a 2-day period. Evaluation of these specimens is in progress.

To investigate the stability of Al_2O_3 -coated UO_2 particles in an oxidizing atmosphere, a sample was heated at a temperature of 1480 C in air, and the weight change was observed by means of a recording microbalance. In the equipment utilized, the sample was heated rapidly to temperature and showed a small but rapid weight loss followed by a weight gain of approximately 20 mg in a period of about 1 hr. Since diffusion of oxygen through alumina at this temperature would be very slow, it appears that the coatings were cracked during the rapid heating, thus permitting oxidation of the UO_2 particles. During a subsequent period of 20 hr, the sample showed a slow, steady weight loss of about 7 mg per hr. These particles are now being examined by metallography and X-ray diffraction. Also, the equipment utilized will be modified to permit a sample to be heated slowly for comparison with the rapidly heated sample.

Fully enriched UC powder was prepared and coated with 55- and 100- μ -thick coatings of pyrolytic carbon for use in studying the diffusion of uranium through the coating. A sample of each of these particles was heated at a temperature of 1400 C for 2 hr in vacuum and has been examined metallographically. There was no evidence of reaction whereas interdiffusion coefficients reported in BMI-1441 indicate that in 1 hr at a 1400 C temperature level the UC particles should have completely reacted with a graphite coating. The discrepancy apparently lies in a considerable difference in the diffusion coefficients for conventional graphite and pyrolytic carbon.

Plans for Future Work

Carbon-coated UO_2 specimens will be run for periods up to 12 hr at a temperature of 1760 C and 2 hr at a temperature of 2000 C. Carbon-overcoated Al_2O_3 -coated UO_2 particles will be run at a temperature of 1800 C for 24 hr. Carbon-coated UC will be heat treated at a 1600 C temperature level and evaluated to plan a series of diffusion anneals. Thermogravimetric equipment will be modified so that a low heating rate can be used to study the behavior of Al_2O_3 -coated UO_2 in air at a temperature of 1500 C.

Coolant-Compatibility Studies

J. F. Foster and A. Levy

Objective

To collect and appraise the information available in the technical literature on possible reactions between gas coolants and ceramic materials of interest for use in gas-cooled nuclear reactors, and to define experimental studies that should contribute missing data on the use of the most promising combinations.

Program Approach

The solid materials included in this study and listed below fall into three categories:

<u>Fuels</u>	<u>Coatings</u>		<u>Matrices</u>
Oxides, nitrides, or carbides of uranium, thorium, or plutonium	Al_2O_3 MgO ZrO_2	Carbon SiC ZrC	Carbon

The gas coolants to be considered are CO_2 , air, nitrogen, H_2O , CO, hydrogen, and helium. Since helium is the preferred coolant at present, it is included primarily for comparison with the other gases. Trace impurities that may be carried by the primary gas coolant are also considered, but the likely impurities are mostly those already listed as possible primary coolants.

To establish qualitatively the compatibility of the above coolants and ceramics the literature is being reviewed for pertinent thermodynamic, kinetic, and radiation data. Quantitative estimates of rates of reaction and evaporation of ceramic materials are carried out by applying available free-energy data to assumed corrosion reactions. With the aid of free-energy data, equilibrium partial pressures of volatile species are calculated; rates of evaporation are then calculated from the Knudsen expression. These calculations ignore any kinetic control of the rate of evaporation and in this sense yield estimated rates of volatilization which should never be lower than the experimental rates. As such, the estimates give maximum values and mean that higher temperature limits should be attainable in practice.

Progress Reported Previously

Thermodynamic and kinetic calculations were completed to give the corrosion rates of Al_2O_3 , and MgO in the presence of air, nitrogen, steam, and carbon dioxide. Preliminary interpretations were presented.

Work This Period

Thermodynamic calculations were made on the rates of volatilization of additional oxides, and the study of all oxides was almost completed this month. The results of the calculations to date are summarized in the tabulation below. These results are highly dependent upon the accuracy of available free-energy data. For purposes of selecting various gas coolant-ceramic coating systems, however, the results are probably reliable within 80 C. The volatilization rates are calculated from the Knudsen diffusion equation; since other kinetic factors, which would reduce volatilization rates, are not included in these calculations, the results are probably on the safe side for predicting the temperature limit at which corrosion problems may become important.

The study of systems in which graphite carbon is the solid in contact with the coolant has been carried far enough to indicate the possibility that none of the selected coolants may be compatible with graphite to the same extent as are some coolants with some oxides in the tabulation below.

Impurity or Coolant	Pressure, atm	Maximum Operating Temperatures ^(b) , C				
		Al_2O_3	MgO	$\text{Nb}_2\text{O}_5^{(b)}$	UO_2	ZrO_2
H_2O	0.001	1230	1230	--	730	>1230
H_2O	100	1130	800	--	530	1230
H_2	0.001	1030	630	--	>1230	1230
H_2	100	730	530	--	>1230	1030
CO_2	100	>1230	>1230	--	630	>1230
O_2	100	>1230	>1230	>1230	(c)	>1230
N_2	100	>1230	>1230	>1230	1230	>1230

(a) Temperature at which the loss of oxide via some volatile component yields an oxide loss rate of approximately 6μ per year.

(b) Cannot be used in reducing atmosphere.

(c) Easily oxidized to U_3O_8 .

Plans for Future Work

Major attention will be given to the probable effects of the various coolants on graphite and the carbides. Interpretations will be formulated, and parts of a topical report will be drafted.

Thermal-Conductivity Studies

H. W. Deem and C. F. Lucks

Objective

To select a method and to design and assemble an apparatus for making thermal-conductivity (thermal-diffusivity) measurements on unirradiated and irradiated specimens containing coated fuel particles.

Program Approach

The problem was approached through a literature survey of methods for measuring thermal conductivity at elevated temperatures.

Progress Reported Previously

The work has been directed toward the selection of a method suitable for measuring the thermal diffusivity of specimens of a size dictated by coated-particle size, encapsulation considerations, and irradiation conditions. A breadboard setup was planned in order to check out certain ideas for making the measurements. The breadboard setup is now ready for measurements on various sensing detectors.

Work This Period

The work during March was directed toward a study of total-radiation receivers that might be applicable to the present problem. Several of these receivers were studied.

The Golay infrared detector made by the Eppley Laboratory shows promise as a total-radiation detector. The Golay detector itself has a response time of from 2 to 30 milliseconds for 63 per cent of the eventual maximum. The usual amplification system for the detector would not be used as it has a response time of from 1 to 12 sec, which is too slow for the present use. It will be necessary to use a special amplifier and readout system if the Golay cell is used.

The use of the thermocouple bead as a total-radiation receiver continues to hold some promise. The bead can be made sufficiently small so that it will have a quick time response and might be applicable for the present purpose, especially if a way can be found to calibrate the bead for attenuation at different frequencies.

Another possibility is the thermister bolometer. One such bolometer is said to operate in the frequency range between 0.8 and possibly 50 cps. Band-pass filters can be used to reduce electrical noise in the amplifier above and below this frequency range.

S. B. Lang of the University of California, Berkley, described a technique for measuring extremely small temperature changes by means of a pyroelectric device. He described several such devices, a typical one being a small disk of polarized barium titanate. The particular disk described as 1 mm thick with a cross sectional area of 1 cm². Silver films to serve as electrodes were deposited on the flat surfaces of the disk and electrical leads were attached. If such a disk is momentarily short-circuited at room temperature and then its temperature is changed by 1 C, a potential of 20 v is developed across the disk. In practice an electrometer is used to measure the voltages. The reed in such an electrometer vibrates at 450 cps, and presumably voltages at frequencies up to perhaps one-fourth of this reed frequency might be measured using an oscilloscope for readout. Extremely high sensitivities are claimed for this type of a detector. Using the disk-shaped sensing element the sensitivity is said to be 10⁻⁶ C. Using a rod that is about 1 cm long and 0.09 cm² in cross section, the sensitivity is about 2 x 10⁻⁷ C. The vibrating-reed electrometer measures currents of the order of 10⁻¹⁷ amp. It is said that these detectors are not limited by Johnson noise, and the only limitation is thermal noises. Of course the disk and rod mentioned could not be used because of their large thermal masses. One of the chief problems in using such a detector will be that of reducing the thermal mass sufficiently, or including the effect of thermal mass in the calibration of the element.

Plans for Future Work

During the next work period an effort will be made to select one or two temperature sensors to be investigated. If possible these sensors will be checked for operation using the breadboard setup.

General plans for the conductivity apparatus itself are nearly completed. When the temperature sensor is selected various parts of the apparatus will be ordered. Basically it is planned to use a tungsten ribbon source to thermally irradiate the specimen. The optics will involve front-surface mirrors with the entire assembly in a vacuum envelope.

Evaluation of Commercial Products

H. S. Rosenberg, T. S. Elleman, and D. N. Sunderman

Objective

To provide information on the availability and quality of coated-fuel particles from commercial sources.

Program Approach

Interested manufacturers of coated particles who request information on the Coated-Particle Program are sent a standard form letter and summary of the evaluation procedures presently used for coated particles. Battelle is authorized to purchase 100-g lots of coated-fuel particles at a price of \$1 per g and to report the results of an evaluation of the material in this section of the report.

Progress Reported Previously

Requests for information on this program from 16 interested commercial firms have been received and answered.

Work This Period

Requests for information on this program from an additional three interested commercial firms were received and answered.

Plans for Future Work

No future work is contemplated until offers of commercial products are received.

PROBLEMS ASSOCIATED WITH THE RECOVERY OF
SPENT REACTOR FUEL ELEMENTS (ORNL)

R. A. Ewing and C. L. Peterson

Two problem areas associated with the recovery of spent reactor fuels are under study in support of the work on fuel reprocessing being carried on by the Chemical Technology Division at ORNL. Corrosion problems are common to all chemical reprocessing schemes yet suggested, and are under continuing investigation. Also being studied are the effects of fuel-element irradiation upon the chemical processes used in aqueous head-end treatments of spent reactor fuels.

The Fluoride-Volatility process, originally intended for fuel elements clad with zirconium or containing zirconium as a diluent, involves a hydrofluorination step during which the fuel elements are dissolved in a fused-salt melt by an HF sparge. This is followed by a fluorination with elemental fluorine which separates uranium as the volatile hexafluoride.

The corrosion of various process vessels and equipment removed from service in the ORNL Fluoride-Volatility pilot plant is being evaluated at BMI, chiefly by metallographic techniques.

Research at ORNL has indicated that oxide-type fuels might also be recovered by the Fluoride-Volatility process. Bench-scale hydrofluorinations are being made at BMI simulating the conditions anticipated with oxide-type fuels (containing UO_2 , BeO , ZrO_2 , or combinations thereof) in order that materials of construction may be evaluated. Of primary interest will be the behavior of the alloy INOR-8 (Hastelloy N or INCO 806). Bench-scale fluorinations will also be studied.

A number of aqueous head-end treatment processes have been devised for the several types of power reactor fuels of the present generation of power reactors. Two of these processes were developed for stainless steel-clad ThO_2 - UO_2 fuels of the type to be employed in the Consolidated Edison reactor at Indian Point, New York. Both are two-step processes, designed to remove the stainless steel cladding for separate disposal. One, the Darex-Thorex process, utilizes dilute aqua regia for decladding, while the Sulfex-Thorex process employs sulfuric acid for this operation. Both processes rely on fluoride-catalyzed nitric acid for dissolution of the declad fuel core.

A considerable fund of information is already available on the performance of both processes on unirradiated fuels, primarily derived from the extensive development work which has been conducted on each. However, comparable data for irradiated fuels are relatively meager, especially for fuels at high burnups. Since significant radiation effects may be encountered in processing such spent fuels, laboratory investigations are being conducted to identify and evaluate these effects.

Corrosion Studies of the Fluoride-Volatility Process

P. D. Miller, E. L. White, E. F. Stephan, and C. L. Peterson

Objective

To evaluate materials of construction for various components of the Fluoride-Volatility process.

Program Approach

Candidate metals and alloys are being subjected to hydrofluorination and fluorination conditions in bench-scale equipment. Also, components from the ORNL Fluoride-Volatility pilot plant are being evaluated by metallographic techniques.

Progress Reported Previously

The start of a combined hydrofluorination-corrosion study involving BeO dissolutions was described in BMI-1504.

Work This Period

The corrosion study mentioned above was completed after a total exposure time of about 300 hr for the INOR-8 specimens. Corrosion rates based on weight-change measurements and micrometer readings were of the order of 2 mils per month or lower. Little preferential attack was noted at liquid-vapor interface positions. One small area of accelerated attack was seen on a specimen located in the liquid directly above the gas sparge.

As was mentioned in the earlier report, severe corrosion was noted near the top of the container where hydrofluoric acid could condense. This corrosion was eliminated when the temperature in this area was increased.

It appeared that the dissolution rate of the BeO decreased during the later portions of the run. The condensate during this period was analyzed to be 96 per cent hydrofluoric acid, showing that less water was being evolved as a product of the reaction.

Nickel containers have been fabricated for the fluorinator corrosion studies. Auxiliary equipment is being assembled.

Plans for Future Work

The INOR-8 corrosion specimen will be examined metallographically.

Materials will be collected to conduct corrosion studies on INOR-8 during the dissolution of ZrO_2 with HF at 700 C.

Work on assembling the fluorination-corrosion equipment will be continued.

Study of the Effects of Irradiation on Cladding- and Core-Dissolution Processes

R. A. Ewing, H. B. Brugger, and D. N. Sunderman

Objective

To investigate the significance of radiation effects upon the chemical processes used in head-end treatments in the recovery of spent reactor fuels of the Consolidated Edison type.

Program Approach

Irradiated prototype fuel pins, in the 20,000-MWD/T burnup range, are being dissolved according to the standard Darex-Thorex and Sulfex-Thorex flowsheets. Principal radiation effects being evaluated are losses of fuel and fertile material to decladding solutions during dissolution of the stainless steel jackets, extent of core fracturing, and the ease of dissolution of the dejacketed cores.

Progress Reported Previously

Two B&W pin remains from postirradiation studies and two ORR pins, all having burnups in the 20,000-MWD/T range, were dissolved by the standard Sulfex-Thorex and Darex-Thorex processes. One B&W specimen and one ORR pin were dissolved by each system. Decladding of all four irradiated pins in 3-hr runs and dissolution of their cores in 5-hr runs were essentially complete. Density determinations of the two B&W cores ranged from 9.20 to 9.57 g per cm^3 . Density determinations of the two ORR cores ranged from 9.14 to 9.55 g per cm^3 .

Work This Period

Analyses are now in progress on the core solutions from the two B&W specimens and the two ORR pins dissolved in the February hot-cell campaign. Analyses of the cladding solution, essentially complete, confirm the superiority of Sulfex decladding with respect to uranium losses. In the absence of complete data on total thorium and uranium, approximate losses have been calculated on the basis of the known core weights, compositions, and the estimated uranium depletions. These data are summarized in Table M-1.

TABLE M-1. APPROXIMATE CUMULATIVE THORIUM AND URANIUM LOSSES DURING DECLADDING OF HIGH-BURNUP CONSOLIDATED EDISON FUEL PINS

Decladding Method	Pin	Approximate Burnup, MWD/T	Decladding Time, hr	Approximate Loss, per cent	
				Thorium	Uranium
Sulfex	ORR-16	20,000	1-1/2	0.022	0.069
			3	0.033	0.156
	B&W 303-6	20,000	1-1/2	0.024	0.061
			3	0.036	0.064
Darex	ORR-21	20,000	1	0.088	0.71
			3	0.20	2.2
	B&W 303-2	20,000	1	0.087	0.77
			3	0.16	1.96

The B&W species were sectioned remains from earlier postirradiation studies, so that the fuel was exposed to the decladding solution during the entire dissolution. Thorium losses for the ORR cores were higher by a factor of 6 to the Darex solution than to the Sulfex solution. Thorium losses by the B&W cores were higher by a factor of 4-1/2 to the Darex solution. Likewise, uranium losses to the Darex solution were 13 to 14 times higher than to the Sulfex solution. These results confirm the results obtained on previous B&W and ORR pins.

Plans for Future Work

Chemical analysis will be continued.

EVALUATION OF FUELED-GRAPHITE ELEMENTS FOR PEBBLE-BED REACTOR (S&P)

A. K. Smalley and H. Rosenberg

Objective

To develop and evaluate ceramic fuel materials for use in the Pebble-Bed Reactor.

Background

Materials have been under development in support of the Pebble-Bed Reactor program being conducted for the AEC by Sanderson & Porter. The retention of fission products within fuel particles in a ceramic matrix by the use of coatings on the particles has been a major objective of the work.

Program Approach

Alumina-coated UO_2 particles and pyrolytic carbon-coated UC_2 particles are being incorporated into graphite spheres as fuel elements for the Pebble-Bed Reactor. Laboratory evaluations of the coated particles before incorporating them into graphite include HNO_3 leaching, alpha-emission measurements, determination of oxidation resistance in the case of oxide particles, evaluation of thermal-cycling resistance, neutron activation, and metallographic examination.

Progress Reported Previously

Evaluation of the carbon-coated UC_2 particles received from Al_2O_3 and of those produced at Battelle were completed. Neutron activation of the Al_2O_3 -coated UO_2 particles received from NUMEC was completed and reported last month in BMI-1504.

Work This Period

Experimental fueled-graphite spheres received from three different vendors were evaluated. Whole-ball alpha-emission measurements were obtained on the as-received spheres and are reported in Table P-1. Three of the spheres from National Carbon Company had high counting rates. One sphere from each vendor was selected on the basis of the lowest alpha emission for neutron activation. The results are given in Table P-2. The specimens from 3M had a particularly low release rate.

The sphere received from Speer had a high release of Xenon-133. As shown in Table P-2, during 388 min at room temperature, with a sweep of 3 liters per hr of helium, xenon-133 release was 4.4×10^{-2} per cent of that present. The specimen was heated to 1600 F in 5 min and during 2 min at 1600 F the xenon-133 release was 5.4×10^{-2} per cent. The release was increasing at this point and the test was stopped.

Metallographic examination of the 3M and National Carbon specimens indicated that the particle coatings in the 3M specimens were free of uranium, while those on the National Carbon specimens exhibited hard, uranium-bearing phases distributed through the coating.

Plans for Future Work

Work reported here terminates this program.

TABLE P-1. SURFACE URANIUM CONTAMINATION OF FUELED-GRAPHITE SPHERES

Sphere	Vendor	Position Counted ^(a)	Counting Rate Normalized to Total Ball Surface, cpm	Surface Uranium Contamination	
				Mg of Surface Uranium	G of Surface Uranium/ G of Total Uranium
16762-86-PCN-6	3M	Area 1 (P)	9.1 ± 4.2	0.012	2.5 × 10 ⁻⁶
		Area 2 (E)	18.2 ± 4.2	0.024	5.0 × 10 ⁻⁶
16762-86-PCN-7 ^(b)	3M	Area 1 (P)	6.3 ± 4.2	0.0083	1.7 × 10 ⁻⁶
		Area 2 (E)	4.9 ± 3.5	0.0065	1.4 × 10 ⁻⁶
16762-86-PCN-8	3M	Area 1 (P)	2.8 ± 4.2	0.0037	7.8 × 10 ⁻⁷
		Area 2 (E)	11.2 ± 4.9	0.015	3.2 × 10 ⁻⁶
16762-86-PCN-9	3M	Area 1 (P)	8.4 ± 4.2	0.011	2.3 × 10 ⁻⁶
		Area 2 (E)	13.3 ± 4.9	0.018	3.0 × 10 ⁻⁶
FA-27(N)-1 ^(b)	NC	Area 1 (P)	15.4 ± 6.3	0.0203	4.27 × 10 ⁻⁶
		Area 2 (E)	18.2 ± 6.3	0.0240	5.05 × 10 ⁻⁶
FA-27(N)-2	NC	Area 1 (P)	474.0 ± 18.0	0.625	1.32 × 10 ⁻⁴
		Area 2 (E)	583.0 ± 20.0	0.769	1.62 × 10 ⁻⁴
FA-27(N)-3	NC	Area 1 (P)	621.0 ± 21.0	0.820	1.73 × 10 ⁻⁴
		Area 2 (E)	611.0 ± 21.0	0.806	1.69 × 10 ⁻⁴
FA-27(N)-4	NC	Area 1 (P)	561.0 ± 20.0	0.740	1.56 × 10 ⁻⁴
		Area 2 (E)	700.0 ± 21.0	0.924	1.94 × 10 ⁻⁴
FA-27(N)-5	NC	Area 1 (P)	35.7 ± 6.3	0.0470	9.90 × 10 ⁻⁶
		Area 2 (E)	29.4 ± 6.3	0.0388	8.16 × 10 ⁻⁶
16782-87-1	Spear	Area 1 (P)	2.8 ± 3.5	0.00370	8.36 × 10 ⁻⁷
		Area 2 (P)	11.2 ± 4.2	0.014	3.34 × 10 ⁻⁶
		Area 3 (E)	7.7 ± 4.2	0.0102	2.31 × 10 ⁻⁶
		Area 4 (E)	0.0 ± 2.8	--	--

(a) P = polar region
E = equatorial region.

(b) Selected for neutron activation.

TABLE P-2. NEUTRON ACTIVATION OF FUELED-GRAPHITE SPHERES

Sphere	Postirradiation Heat Treatment			Incremental Release, per cent of xenon-133
	Test Temperature, F	Time at Temperature, min	Time at Temperature, min	
16762-86-PCN-7 (3M)	1500	6	90	5.72×10^{-3}
	2000	4	90	1.78×10^{-3}
	2400	6	100	3.36×10^{-3}
16762-87-1 (Speer)	RT	--	388	4.40×10^{-2}
	1600	5	2	5.40×10^{-2}
FA-27(N)-1 (NC)	1500	6	60	2.4×10^{-2}
	2000	4	40	5.9×10^{-3}
	2400	6	55	1.7×10^{-2}

DEVELOPMENT OF FABRICATION PROCESSES FOR COLD BONDING OF ZIRCALOY-2 TO TYPE 410 STAINLESS STEEL (SRO)

S. W. Porembka

This program was established to develop techniques for joining pressure tubes of Zircaloy-2 and Type 410 stainless steel for the CANDU reactor program. Ultimately, it is desirable that the developed methods be suitable for performance in-pile in order to facilitate the replacement of such tubes. Also, since brittle compounds are formed in alloying these materials, it is desirable that the techniques selected be restricted to those which result in a minimum of diffusion. Consequently, bonding studies to date have concerned friction techniques, the effects of surface preparation in roll forming, and explosive joining. Each of these methods is characterized by limited diffusion since each is performed at ambient temperatures. In each approach, both the direct joining and transition joining of the materials is being investigated through studies of dissimilar metal joints and self-bonds. The program has recently been expanded to include a study of end capping Zircaloy-2 fuel sheaths by friction techniques.

Friction Bonding

S. J. Paprocki, E. S. Hodge, and H. D. Hanes

Objective

To study the joining of Type 410 stainless steel to Zircaloy-2 and the end capping of Zircaloy-2 fuel sheaths by friction techniques.

Program Approach

Joint design, pressure, rotational velocity, time, and surface preparation are being investigated in joining Zircaloy-2 to stainless steel by friction bonding. More recently these studies have been directed to joining tubular components. The same variables will be examined in the end-capping studies. Metallographic techniques are presently employed as the primary method of evaluation. Satisfactorily joined specimens are subsequently corrosion tested in 750 F steam and their mechanical properties determined. A burst test is being considered as a means for inspecting the integrity of end caps.

Progress Reported Previously

Upsetting of the Zircaloy into the bore of the stainless steel tubing was reported in BMI-1496. This problem was solved by the use of a mandrel as described in BMI-1504. Shearing in the Zircaloy after adhesion to the stainless steel was also reported.

Work This Period

Attempts were made to heal the tears developed in the Zircaloy-2 member during friction bonding by using a two-step bonding cycle. Initially, the rod-type specimens were preheated at a relatively low pressure and the pressure was then applied as the relative motion between the samples was stopped. There was no noticeable increase in bond integrity.

Plans For Future Work

In view of the extent of engineering development that would be required in the application of friction techniques to joining CANDU pressure tubes, the present effort will not include full-scale components. This study will continue to the point where conditions for bonding small-scale components are established. Major emphasis will be shifted to the end capping of Zircaloy-2 fuel sheaths.

Roll Forming

S. J. Paprocki, R. J. Carlson, and E. G. Smith, Jr.

Objective

To determine the effects of surface preparation and design on metallurgical bonding in roll forming joints between Type 410 stainless steel and Zircaloy-2.

Program Approach

Studies of surface preparation and rolling reductions were completed on flat-plate couples of stainless steel and Zircaloy. Presently the applicability of these results to the roll-forming technique is being considered. It is planned that variations of groove design in the present joint will also be included in this study.

Progress Reported Previously

Preliminary cold-rolling studies involving surface preparation and fabrication variables to achieve bonding between Type 410 stainless steel and Zircaloy-2 components have been completed. The results indicated that machined surfaces yield a greater degree of apparent bonding than those subjected to either belt abrading or commercial chemical etches. Cold reductions of 45 to 50 per cent were required to achieve any degree of bonding. The most successful surface preparation consisted of a 300- μ in. rms finish on the mating surfaces of both the stainless steel and Zircaloy-2 which were subsequently scrubbed with a MgO-NaCO₃ solution.

Work This Period

Samples of cold-rolled stainless steel-Zircaloy-2 couples have been heat treated to promote diffusion across the bonded interface. The amount and uniformity of the diffusion zone formed should be indicative of the relative degree of bonding in the samples. Heating the samples in vacuum at 1400 F and 1500 F each for 4 hr produced only a small amount of diffusion, and this was limited to the specimens that had taken the highest reductions.

Materials and equipment necessary for the roll-forming operation are being obtained. Zircaloy-2 tubing for these tests was obtained from CANDU. Material for the Type 410 stainless steel headers is on order as well as the tube rollers. A survey is being made on available power supplies that are capable of producing the speed and torque necessary to drive the tube rollers.

Plans For Future Work

All future efforts in roll forming will be directed toward achieving a metallurgically bonded joint using full-size Zircaloy-2 and stainless steel components.

The surface preparations that resulted in the most consistent bonding in the preliminary rolling study will be used on the surfaces of the joint components in the roll-forming operation. Emphasis will be placed on groove design to achieve the high local deformations that have been found necessary for bonding Zircaloy-2 to stainless steel.

Explosive Joining

S. J. Paprocki, R. J. Carlson, and C. C. Simons

Objective

To determine the feasibility of achieving mechanical and metallurgical bonding between Zircaloy-2 and Type 410 stainless steel by explosive techniques.

Program Approach

Explosive techniques are being employed to bond Zircaloy-2 to Type 410 stainless steel. Evaluation is based upon bond strength as determined by metallographic examination and mechanical testing.

Progress Reported Previously

Explosive-joining tests to date have included both flat-plate assemblies and butt-welding assemblies. Results are reported in BMI-1489, BMI-1496 and BMI-1504. Successful bonding of flat-plate assemblies were achieved. Possible ways to improve the technique are being evaluated.

Work This Period

Two additional flat-plate assemblies have been subjected to the explosive-welding operation. These specimens were similar to those described in previous flat-plate tests with the exception that the evacuation tubes were extended to the specimen on the interior of the assembly. With this modification it was anticipated that a more complete evacuation of the system could be achieved and the formation of intermetallics at the Zircaloy-stainless steel interfaces eliminated. In one of these assemblies a rubber buffer was placed between the explosive and the bonding assembly. Preliminary examination of the bonded specimens indicated less total deformation in the experiment utilizing the buffer. Sections of the samples are being prepared metallographically for further evaluation.

Plans For Future Work

Metallographic evaluation of the two explosive-welded flat-plate assemblies will be completed. Further tests employing small-scale tube components are also planned.

RADIATION-EFFECTS STUDY OF CANDIDATE
FUEL MATERIALS FOR THE MGCR (GA)

N. E. Miller, P. Steinback, J. E. Gates, and W. H. Goldthwaite

Objective

To determine the irradiation stability of selected ceramic-matrix fuels under conditions of temperature and burnup similar to those anticipated in the MGCR.

Background

Battelle has assisted General Atomic in the development and evaluation of core materials for the Maritime Gas-Cooled Reactor and the High-Temperature Gas Reactor. This work has included research and development of ceramic-matrix dispersion-type fuels and fuel-particle coatings and study of coolant-core material compatibility in an irradiation environment. At the present time, with the emphasis on the BeO-moderated helium-cooled concept for the MGCR, Battelle is conducting studies of the irradiation damage of BeO-matrix fuels up to relatively high burnups.

Program Approach

Clad fuel pellets are irradiated in capsules which are equipped with thermocouples and auxiliary electric heaters and designed to maintain specimen-surface temperatures in the 1500 to 1700 F range, primarily by fission heating. Target burnups range from 1 to approximately 50 per cent of the uranium-235 atoms originally present. Data concerning specimen burnup, swelling, fission-gas release, and microstructural changes are obtained in postirradiation hot-cell examinations.

Progress Reported Previously

Capsules GA-BRR-1 and BMI-31-1, -2, and -3. Each of these four capsules contained two specimens of the following three fuel compositions: 20 volume per cent UO₂ in BeO, 20 volume per cent uranium monocarbide in graphite, and 20 volume per cent uranium dicarbide in graphite; the uranium enrichment in each of the 31-series capsules was 30 per cent, whereas in the GA-BRR-1 capsule it was 93 per cent. The fuel pellets were canned in Type 316 stainless steel tubing under a helium atmosphere.

One of the four capsules, GA-BRR-1, was irradiated in the Battelle Research Reactor to specimen burnups ranging from 1.0 to about 1.7 a/o with specimen-surface temperatures ranging from 1275 to 1560 F. Postirradiation examinations at the Battelle Hot-Cell Facility indicated that all three types of specimens were in good physical condition.

The irradiation of the remaining three capsules at the Materials Testing Reactor was completed; the last capsule in the series, BMI-31-1, was discharged February 27, 1961. Estimated specimen burnups (in terms of percentage of the original uranium-235 atoms fissioned) are as follows: for Capsule BMI-31-1, 35 to 40 a/o; for Capsule BMI-31-2, 16 to 20 a/o; for Capsule BMI-31-3, 17 to 22 a/o.

Capsule BMI-36-1. This capsule is being prepared for the irradiation at the MTR of pellets of 30 volume per cent UO₂ (12 per cent enriched) in BeO encased in Hastelloy X. Target irradiation conditions include a specimen-surface temperature of 1700 F and an in-pile period of about 18 months at a specimen-effective peak flux of about 2×10^{13} nv. These exposure conditions will yield a peak burnup in the neighborhood of 50 a/o.

S-2

Work This Period

Capsule BMI-31-1. As was the case following the discharge of BMI-31-2 and BMI-31-3, BMI-31-1 was shipped to the GA hot cells for postirradiation examination.

Capsule BMI-36-1. The fueled pellets for this capsule were furnished to Battelle by GA during March. Assembly of various capsule members has begun.

Future Work

Capsule BMI-31-1. Further work in this capsule series is not contemplated.

Capsule BMI-36-1. According to the present schedule, this capsule system will be ready for shipment to the MTR during May.

RADIATION STUDIES OF SM-2 FUELS (ALCO)

J. H. Stang, J. F. Lagedrost, and W. H. Goldthwaite

Objective

The effects of irradiation on stainless steel-clad dispersions of UO_2 (nominally 26 w/o, highly enriched) and certain burnable poisons in Type 347 stainless steel are being investigated.

Background

This study is being conducted in assistance to Alco Products to develop satisfactory fuel materials for Army pressurized-water reactors. Reference and alternate SM-2-type specimens have been fabricated and are being irradiated in the ETR.

Program Approach

Capsules containing five to eight specimens each are being irradiated or have been irradiated in the MTR or the ETR. Desired experimental conditions include nominal specimen-surface temperatures of 600 F, and burnups of 37 and 70 per cent of the total uranium-235 atoms. The program includes three noninstrumented and seven instrumented (heaters and thermocouples) capsules; the former were designed for irradiation in MTR core-lattice positions, the latter for irradiation in ETR beryllium-reflector positions. This capsule group is identified as the BMI-32 series.

Progress Reported Previously

Irradiations of the first four capsules were completed early in 1960; irradiation histories and results of specimen analyses were reported in BMI-1442, BMI-1448, and BMI-1464.

The irradiation status of Capsules BMI-32-5 through BMI-32-10 in the ETR for the period up to December 31, 1960, was reported in BMI-1489 and BMI-1496. It was pointed out that peak-flux specimens in each capsule should reach the target burnup of 70 a/o (total, uranium-235 atoms) by mid-1961 with normal reactor operation. Efforts are being made to maintain the specimen-surface temperatures at 600 F as long as possible as burnup progresses by gradually increasing the auxiliary heating in the capsules.

Work This Period

Irradiations of Capsules BMI-32-5 through BMI-32-10 were continued. During the latter part of ETR Cycle 35, which ended on March 6, 1961, the heaters in four of the capsules and 8 of the 21 thermocouples considered operable were not functioning. In some cases, the malfunction could be attributed to circuit failures in the lead-tube terminal boxes or between these boxes and the instrument panels. Maintenance work during the current shutdown is directed toward eliminating some of the sources of trouble and restoring the circuits to operation.

Estimates have been made of the anticipated total burnup level of peak-flux specimens in each capsule after ETR Cycles 37, 38, and 39. These cycles are scheduled to end on May 29, July 10, and August 21, respectively. The estimates, shown in Table T-1, take into account variations in reactor flux which have been indicated by ETRC measurements and capsule thermocouple temperatures and changes in flux perturbation which accompany burnup.

TABLE T-1. ESTIMATED RANGE OF PEAK-FLUX SPECIMEN BURNUP IN BMI-32 SERIES CAPSULES UNDER IRRADIATION IN THE ETR

Capsule	Burnup, per cent of uranium-235 atoms originally present		
	End of Cycle 37(5-29-61)	End of Cycle 38(7-10-61)	End of Cycle 39(8-21-61)
BMI-32-5	60-65	70-75	75-80
-6	65-70	70-75	75-80
-7	65-70	72-77	77-82
-8	67-72	73-77	78-82
-9	85-90	>90	>90
-10	60-65	67-72	75-80

For these estimates, it has been assumed that the reactor will operate for 28 days at full power for each of Cycles 36 through 39. Also, it was assumed that the trend in reactor fluxes observed during the past eight cycles will not change appreciably. Fluxes during Cycle 35 are believed to have been as much as 25 per cent lower than they were when the first of these irradiations started.

The reductions in reactor fluxes have resulted in corresponding reductions of specimen fission-heat generation and burnup rates. (These flux changes were brought about by core-loading changes necessary to accommodate loop experiments being installed in the ETR.) Furthermore, it seems apparent from a consideration of planned core loadings for future cycles that even lower fluxes for the BMI-32 capsules will result. The possibility of transferring the capsules to higher flux positions in the ETR has been considered. However, some straightening and rebending of lead tubes probably will be necessary. This would be quite hazardous from the standpoints of lead-tube failure and loss of heater or thermocouple lead continuity.

Plans for Future Work

The ETR irradiations of BMI-32-5 through BMI-32-10 will be continued.

GAS-COOLED REACTOR PROGRAM (AGN)

D. L. Keller

The Army Reactor Branch (ARB) is developing small portable reactor systems for military applications. Aerojet-General Nucleonics (AGN) is the prime contractor under the Army Gas-Cooled Reactor Systems Program (AGCRSP) and is charged with the design, construction, and operation of the GCRE and ML-1. The object of the ML-1 program is to produce a prototype mobile nuclear plant suitable for military field use.

The programs reported in the following sections are in support of this ML-1 program and are concerned primarily with fuel-element development.

An Alternate Method of Fabricating Advanced ML-1 Fuels

S. J. Paprocki and D. L. Keller

Objective

To investigate the simultaneous densification and cladding of Al_2O_3 -coated UO_2 as a method of obtaining a high-density oxidation-resistant fuel material.

Program Approach

It has been demonstrated that the oxidation resistance of UO_2 powder can be significantly improved by protecting the powder with a thin coating of Al_2O_3 . In this program, the coated powder will be used as a starting material. Various methods of packing the coated powder into stainless steel tubes will be investigated. The steel tubes will be evacuated and under isostatic pressure collapsed around the ceramic core. The maximum Al_2O_3 diluent is restricted to about 40 volume per cent. The main concern is the retention of coating integrity during densification. This will be evaluated by nitric acid leach tests, oxidation tests, and metallographic studies.

Progress Reported Previously

Spherical UO_2 particles 150 and 50 μ in average diameter have been coated by a vapor-deposition method with from 5 to 10 μ of Al_2O_3 . Six cylindrical specimens were prepared from mixtures of these coated particles by hot pressing in mild steel tubes at 2300 F for 3 hr under an isostatic pressure of 10,000 psi. The tubes were removed from the core by pickling, and the cores were subjected to metallographic examination, leach tests, and oxidation tests. The most significant factors indicated by these preliminary results are:

- (1) The coated powder used in these studies is resistant to both nitric acid and air oxidation.
- (2) After hot isostatic pressing, the maximum density obtained was approximately 85 per cent of theoretical. Higher densities can likely be obtained by increasing the hot-pressing temperature or increasing the amount of "active" Al_2O_3 (that alumina added as a powder as opposed to that added as a vapor-deposited coating).
- (3) Metallographic examination of two specimens made before the cladding was removed revealed no cracks in the Al_2O_3 phase.
- (4) Dissolution tests in a nitric acid solution showed a varying degree of attack on four unclad specimens. Two of these specimens showed some resistance to air oxidation in that the specimens

were still intact after 2 hr at 1550 F. One specimen gained weight rapidly at 1200 F and had disintegrated to powder after 1 hr.

- (5) These results suggest that the Al_2O_3 is failing both when the cladding is removed and when the specimens are rapidly heated to temperature prior to oxidation testing.

Work This Period

No new work was performed during this period.

Plans For Future Work

An effort will be made to determine at exactly what stage of fabrication or testing the Al_2O_3 coatings are failing.

Additional specimens will be prepared and some of these will be tested as defected pins rather than as unclad cores. Also, some cores will be prepared in niobium tubing to permit an evaluation of a higher pressure-bonding temperature.

Encapsulation Studies

W. J. Zielenbach, J. F. Lagedrost, and J. H. Stang

Objective

To investigate the elevated-temperature radiation stability of Hastelloy X-clad compacts of highly enriched UO_2 - both alone and dispersed in a BeO matrix.

Program Approach

Two capsules, each containing six clad specimens based on highly enriched UO_2 , are being irradiated, one capsule at the Materials Testing Reactor and the other at the Battelle Research Reactor. The specimens in the MTR capsule consist of 70 and 80 w/o UO_2 dispersed in BeO; there is no diluent in the specimens being irradiated at the BRR. The peak burnup levels presently desired are 9 per cent of the original uranium-235 atoms for the former specimens and 3.5 per cent for the latter. The desired specimen cladding-surface temperature is 1725 F in both cases.

Progress Reported Previously

The irradiation history of these capsules was reported in BMI-1480, BMI-1489, BMI-1496, and BMI-1504. Performance and monitoring of the MTR capsule (BMI-38-1) has been hampered by failures of auxiliary heaters and thermocouples. Performance of the BRR capsule has been satisfactory.

Work This Period

Capsule BMI-38-1 completed its ninth in-pile cycle at the MTR on March 20. Estimates of current performance are being based on data collected previous to the failure of the capsule thermocouples and heaters. The peak specimen-surface temperature is approximately 1550 F and the peak flux specimens have accrued an estimated 10 per cent burnup of the original uranium-235 atoms.

Capsule AGN-III has completed its thirteenth cycle in the BRR; the estimated accumulated burnup of the peak flux specimen is 3 a/o. During this period, the peak cladding temperature averaged 1530 F with the addition of full auxiliary electrical heat (1000 w). This lower-than-normal temperature level has resulted from reactor operation at less than full power.

Plans For Future Work

The MTR capsule will be discharged about April 10 while the BRR capsule will be discharged about May 1. It is planned to examine the former capsule at the Phillips hot cell. The capsule irradiated at the BRR will be examined at the BMI Hot-Cell Facility.

CORROSION OF THORIUM AND URANIUM UNDER STORAGE CONDITIONS (LIVERMORE)

P. D. Miller

Thorium, uranium, and their alloys are being considered by the Lawrence Radiation Laboratories and by the Sandi Corporation for applications which may require exposure to a variety of atmospheric conditions. Two research programs, aimed at enlarging the information concerning the corrosion resistance of these metals are under way at Battelle.

In one study, procedures are being developed for applying corrosion-resistant electroplated coatings to thorium and to uranium.

A fairly extensive corrosion evaluation is also being conducted under a variety of conditions on bare and coated specimens of the above-mentioned metals and their alloys.

Electrodeposited Coatings for Thorium (SC)

J. G. Beach and C. L. Faust

Objective

To develop a coating for thorium that prevents corrosion and to evaluate techniques developed for thorium to the protection of uranium.

Program Approach

The various factors for producing sound protective electroplated coatings on thorium are being studied: (1) prefinishing, to remove surface contamination and defective basis metal; (2) activating, to provide maximum adhesion of the plated coating; (3) coating system, to provide the needed protection and/or surface properties; (4) post-plating treatments, to improve the properties of the composite system. Metallography, bond tests, and corrosion tests are being used to evaluate the coated thorium. A cursory study toward protecting uranium by using the techniques developed for thorium is to be made so as to define the factors that need further research.

Progress Reported Previously

Nickel (1 to 2 mils) plus chromium (0.035 mil) protected as-plated thorium samples (2 by 3 in.). Outgassing of the plated thorium by "slow" heating to 500 F in vacuum was necessary to prevent blistering during exposure to a corroding medium. Samples of coated and outgassed thorium are being exposed to conditions of the UCRL corrosion evaluation.

Work This Period

The various factors developed thus far for producing sound, protective, electroplated coatings on thorium (prefinishing, activating, nickel plating, chromium plating, and outgassing) are being investigated toward the protection of uranium. The quality of the protective coatings on uranium will be evaluated by exposure to water-saturated air at 200 F. Electropolishing to remove 3+ mils of the surface metal after machining is used to remove damaged surface metal caused by machining. The uranium is activated for adherent plating by using a procedure* which involves nitric acid pickling, anodic

*"Electroclad Aluminum on Uranium", J. Electrochem. Soc., 106 (8), 654-659 (August, 1959).

etching in phosphoric-hydrochloric acid solution, and again pickling in nitric acid prior to plating. Nickel plating is done to a thickness of 1 to 2 mils in a leveling-type semi-bright nickel bath. Heating of the nickel-plated uranium (vacuum outgassing at 500 F and/or 1000 F) is being investigated to improve the protection.

Electropolishing of thorium compared to chemical pickling of thorium was not an improved pretreatment for adhesive bonding. Break loads of samples in tensile shear were better for chemically pickled thorium (2000 versus 1500 psi), using an epoxy resin adhesive. Further study of electropolishing as a preparation for adhesive bonding does not appear merited.

Plans For Future Work

Evaluation of electroplated coatings on uranium will be continued. Comparison of various types of thorium metal as to the quality of fabricated sheet and as to plating characteristics is planned. The protection of thorium by a chromate conversion coating will be explored. An unexpected corrosion resistance of a "bare" thorium sample was observed in the UCRL corrosion series. The corrosion resistance is attributed to a conversion coating that had been applied prior to paint coating and, in this case, the paint coating had been stripped off to supply an additional bare thorium specimen.

Corrosion Evaluation (LRL)

E. F. Stephan, P. D. Miller, and F. W. Fink

Objective

To determine the durability of thorium, uranium, and related alloys under storage conditions.

Program Approach

Bare and coated coupons of the metals are exposed under simulated storage conditions and under accelerated laboratory conditions.

Progress Reported Previously

In a Phase I program, thorium, uranium and uranium-10 w/o molybdenum-alloy specimens were exposed to three accelerated laboratory conditions, as summarized in BMI-1469. The attack was localized and resulted in some pitting. The alloy was the most durable material studied. The uranium was the most severely attacked. BMI-1504 summarized the results of the Phase II study involving exposure of materials to 100 per cent relative humidity at 120 F for 2500 hr. A greater variety of specimen types was included in this program. Again, the uranium-10 w/o molybdenum alloy was the most durable material.

Work This Period

A topical report is being prepared, presenting a fairly detailed account of the results from the Phase I and II programs. Additional electroplated thorium specimens were introduced into the Phase III exposure areas.

Plans For Future Work

The topical report on the Phase I and II programs will be completed. The Phase III program, described in BMI-1473, will be continued.

GAS-PRESSURE BONDING OF BERYLLIUM-CLAD ELEMENTS (ORNL)

S. J. Paprocki, E. S. Hodge, and J. S. Perrin

Objective

To develop fabrication techniques for cladding UO_2 fuel cores with beryllium.

Background

The gas-pressure-bonding process is valuable as a fabrication technique because in this process the components undergo a minimum of deformation. The process is thus particularly suited for use with materials, such as beryllium, that have directional properties. In addition, the technique will enable cladding of brittle fuel core materials, such as uranium oxide, to be accomplished without producing fragmentation. Beryllium-clad UO_2 is of interest in ORNL's studies on potential materials for gas-cooled reactors, and the program reported here is in support of the work at ORNL.

Program Approach

The gas-pressure-bonding technique is being used to fabricate specimens. Evaluation is based upon bond and cladding properties after bonding as determined by metallographic examination and mechanical testing.

Progress Reported Previously

Studies of the effects of surface preparation and bonding parameters on the self-bonding of beryllium are being conducted. Self-bonding of beryllium structural components similar in design to those contained in compartmented fuel elements has been studied. Beryllium specimens containing UO_2 cores have been bonded at various parameters to determine the compatibility of the UO_2 fuel with beryllium. No reaction was observed between a pyrolytic carbon-coated UO_2 core which was bonded in contact with beryllium at 1650 F for 4 hr at 10,000 psi.

Work This Period

Two additional specimens containing beryllium in contact with coated UO_2 cores have been bonded and metallographically examined. One specimen contained a pyrolytic carbon-coated core and was bonded at 1750 F for 4 hr at 10,000 psi. Metallographic examination indicated that pyrolytic carbon is not a suitable coating material at these bonding parameters. The second specimen contained a chromium-overcoated iron-coated UO_2 core and was bonded at 1650 F for 4 hr at 10,000 psi. No reaction occurred between the beryllium and UO_2 core.

Additional specimens are being bonded to determine the effect of surface preparation prior to cleaning, assembling, and bonding on the self-bonds achieved during bonding. These include pickling in three different acid solutions, grit blasting, and rolling. The purpose of the latter two treatments is to increase the amount of cold work in the beryllium components, which tends to promote recrystallization and grain growth across the interface.

Specimens which contain beryllium plates in contact with various barrier materials have been bonded at 1650 F for 4 hr at 10,000 psi. Barrier materials being studied include mild steel, chromium-coated mild steel, stainless steel, MgO-coated stainless steel, thoria-coated stainless steel, molybdenum, nickel, niobium, tantalum, and tungsten. These are being evaluated by metallographic examination.

X-2

Plans for Future Work

Fabrication of specimens to study the self-bonding characteristics of beryllium and the compatibility of beryllium with spacer materials and UO_2 will continue.

RWD:CRT/all