3204p.

BMI-1886

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PROGRESS ON DEVELOPMENT OF FUELS AND
TECHNOLOGY FOR ADVANCED REACTORS
DURING JULY, 1969, THROUGH JUNE, 1970
ANNUAL REPORT

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Prepared for U. S. Atomic Energy Commission
Under Contract No. W-7405-eng-92

Report Date: July, 1970

P7580

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✓ URANIUM-PLUTONIUM NITRIDE FUELS FOR
LMFBR APPLICATIONS (AEC-DRDT)✓

W. M. Pardue, A. A. Bauer, P. Cybulskis,
and C. A. Alexander

These studies are being conducted for the AEC Division of Reactor Development and Technology to evaluate the potential of mixed UN-PuN fuels for fast breeder applications. The use of mixed-nitride fuels in liquid-metal fast breeder reactor (LMFBR) applications is considered desirable because of the high fissile density of the materials, the high thermal conductivity as compared to that of oxides, and the high breeding ratio attainable. Several studies of uranium nitride have been undertaken for thermal reactor applications and the UN in these programs has exhibited desirable irradiation-stability behavior, thus implying that the UN-PuN fuels may be satisfactory in this respect.

The program is divided into efforts involving the preparation of materials by gas-solid reactions, property determinations, irradiation studies, and supporting studies. Physical properties of importance for reactor design are being determined, including volatility tendencies, conductivity, expansivity, and chemical stability of the fuels in contact with potential LMFBR cladding materials. The resistance of nitride materials to both thermal and fast flux irradiations will be determined to evaluate the suitability of these fuels for applications involving long-term irradiations.

The evaluation of sodium bonding of the fuel to the cladding is another objective of the program. Nitride fuel grows at a rate of about 0.5 v/o per 10^{20} fissions per cm^3 . To minimize cladding deformation, this fuel growth can probably be accommodated more effectively in the annulus between the fuel and the cladding rather than in the porosity in the fuel. To minimize fuel temperatures, another desirable objective, this annulus must be filled with a good heat-transfer material. Effort is thus devoted to studies of sodium bonding and film formation in contact with nitride fuels and their claddings.

Thermodynamics and Transport Characteristics
of PuN and Alloys of UN-PuN

C. A. Alexander, J. S. Ogden, and W. M. Pardue

Objective of Program

To determine those properties pertaining to plutonium nitride-bearing fuel compositions which allow predictions of fuel stability, transport, and other factors needed to evaluate the behavior of the fuel under possible reactor environmental conditions of both transient and steady-state nature.

Approach being Used

Kinetics and thermodynamics of vaporization processes are being investigated by effusion-thermobalance and mass-spectrometric techniques. The existence of a homogeneity range is being examined under controlled atmosphere conditions. The Oelsen calorimeter is being utilized to experimentally determine heat capacities as a function of temperature and alloy composition.

The experimental apparatus used in the studies consists of a cold-wall furnace constructed of stainless steel which is evacuated through a pipe 10 cm in diameter, a diffusion pump with a capacity of 300 liters per sec, and a mechanical pump with a capacity of 7 liters per sec. The system is equipped with valves so that the furnace

chamber can be isolated from the pumps and backfilled with inert gas or any nonoxidizing gas to the desired pressure. The furnace assembly has a maximum leak rate of 10^{-9} std cm^3 . A Cahn recording microbalance is also incorporated into the vacuum chamber which houses the basic vaporization furnace. The electrobalance is used to continuously monitor the vaporization rates as a function of time and/or temperature.

The double-focusing mass spectrometer used for the surface-ionization studies on neodymium has a focal point about 8 cm on either side of the 25-cm radius of curvature in the electrostatic sector. The magnetic sector is asymmetric, and the distance from the image to the 60-deg, 20-cm sector is shorter than the object distance. The electromagnet incorporates a Rawson field meter in a feedback arrangement to supply short-term stability of about 1 ppm in the magnet power supply. A field sweep is also incorporated in such a manner that the sweep can be made in gaussses rather than in amperes. The design criteria of the machine are such that with 2500 v of accelerating potential, masses up to 1200 are resolvable in the magnetic sector. A resolution of about 2500 at mass 25 was selected as a reasonable compromise between resolution and transmission. The sensitivity is such that the individual events can be monitored. Data output is on a strip-chart recorder using a Cary Model 31 vibrating-reed electrometer and Faraday cage or an electron multiplier tube and appropriate amplifier.

Significant Accomplishments During Fiscal Year 1970

The behavior of neodymium in NdN as a measure of burnup revealed that there was neither a lack of thermal stability, nor the existence of a gaseous species, nor an anomaly in chemistry that can account for any lack of agreement when isotopic analyses of mixed-nitride fuels are compared with those obtained from mixed oxides.

Equations for mass transport and general thermochemical properties were obtained. It was also established that the presence of PuN in solution in UN decreases the uranium activity and thereby prevents the formation of a molten phase at temperatures to at least 2000 K. At both 1 and 0.1 atm of nitrogen no homogeneity range for PuN was detected over the temperature range from 1500 to 2140 C. Pressure-temperature-composition relationships for PuN and (U,Pu)N were also prepared.

The homogeneity ranges for PuN and $(\text{U}_{0.8}\text{Pu}_{0.2})\text{N}$ are very narrow. The nitrogen/metal ratio varies from 1 at 1 atm of nitrogen to 0.968 at 10^{-4} atm of nitrogen.

Summary of Research Carried Out

The usage of the concentration of Nd^{150} as a measure of burnup has been well established for irradiated mixed-oxide fuels; however, when the same technique is applied to mixed-nitride fuels the agreement between the isotopic determination and the burnup calculated from heat-transfer data and fission-gas release is not as close as with oxide fuels. Since little is known concerning the stability and chemistry of NdN, which is the assumed compound formed by the Nd^{150} , it was considered that an understanding of these properties might indicate the reason for the lack of accord when the isotopic technique was applied to burnup measurements of mixed nitrides.

A 3-g right circular cylinder of 99.99 percent pure neodymium metal was electro-discharge machined from a rod and placed within a Knudsen cell suspended from a thermobalance. Hydrogen was admitted to a pressure of 0.9 atm, and the temperature was slowly raised to 420 C. Hydriding was completed in about 30 min at that temperature, and the material reached a composition of $\text{NdH}_{2.70}$. Nitrogen at a pressure of 0.9 atm was introduced to the system and the temperature was slowly raised to 825 C. After about 70 min at that temperature the nitriding ceased; the composition, determined by weight change, was $\text{NdN}_{0.99}$. The apparatus was subsequently evacuated to 10^{-6} torr and a series of effusion experiments begun. At 1500 K the initial rate of weight loss indicated the dissociation pressure of nitrogen to be 1.3×10^{-6} atm; however, it was observed that this initial rate tended to decrease with time. When a composition near

$\text{NdN}_{0.94}$ was reached the evaporation rate was no longer time-dependent, and it appeared that congruent vaporization was occurring. This means the weight loss occurred as a result of the reaction $\text{NdN}_{0.94} = \text{Nd}_{(g)} + 0.47\text{N}_{2(g)}$. At 2000 K the partial pressure of neodymium was 9.8×10^{-5} atm; that of nitrogen was 2.1×10^{-5} atm. These quantities can be compared to the pressure of metal and nitrogen over the mixed fuel at a similar temperature. For $(\text{U}_{0.8}\text{Pu}_{0.2})\text{N}$ at 2000 K: $p_{\text{N}_2} = 1.1 \times 10^{-6}$ atm, $p_{\text{Pu}} = 6.7 \times 10^{-6}$ atm, and $P_{\text{U}} = 4.0 \times 10^{-8}$ atm; hence, pure neodymium nitride is somewhat less stable than the fuel, but these data represent the situation for unit activity. At a concentration of NdN anywhere less than 1 m/o it would be expected to exhibit quite good stability, and this concentration is the case in irradiated fuels.

In an effort to determine whether something in the chemistry of NdN is contributing to the discrepancy, some surface-ionization mass-spectrometric runs were performed. Initially, 1.000 g of Nd_2O_3 was dissolved in 1 liter of 8M HNO_3 . The isotopic distribution of neodymium was run and both Nd^+ and NdO^+ ions were observed. A solution of $\text{Nd}_2^{143}\text{O}_3^{16}$ was then made, and a known quantity of this solution was added to a portion of the oxide solution. On the basis of isotopic intensities and the dilution technique, it was established that about 9×10^{-9} g of neodymium would be sufficient for an isotopic analysis. No attempt was made to optimize the emission; therefore, a considerable improvement over this value may be possible.

The nitride charge was subsequently removed from the Knudsen cell and 0.313 g dissolved in 8M HNO_3 . A spike of the Nd^{143} solution was added and the isotopic analysis run. Within the precision of the mass-spectrometric method the results were identical with those of the oxide experiment; thus, the expected response of neodymium to mass-spectrometric isotopic analysis is the same regardless of its chemical form. It then must be concluded that neither lack of thermal stability, nor existence of gaseous species, nor differences in chemistry can account for any lack of accord when isotopic analyses of mixed-nitride fuel are compared with those obtained from mixed oxides.

Experiments establishing the existence of the phase limits of the PuN phase and the limits of the $\text{U}_{0.8}\text{Pu}_{0.2}\text{N}$ phase were carried out in selected partial pressures of nitrogen. On the basis of effusion and microbalance determinations it was established that the PuN phase exists at all nitrogen pressures greater than those listed in Table A-1 for the corresponding temperatures. If an attempt is made to lower the system pressure to one of these pressure values at its corresponding temperature, say 2×10^{-4} atm at 2400 K, a liquid phase would not be encountered. This is because the partial pressure of plutonium, as determined in previous work, would be 1.5×10^{-3} atm and the plutonium would sublime at this higher system pressure rather than exist as a liquid. Clearly, then, it is necessary to utilize a total system pressure in excess of the nitrogen partial pressure to suppress the volatilization of plutonium. This is a requirement because of the nature of the measurement. If plutonium is lost by vaporization, then it is not possible to determine the change in the plutonium/nitrogen ratio from the balance measurements. Gases containing nitrogen at pressures of 10^{-4} atm, 10^{-3} atm, 10^{-2} atm, 10^{-1} atm, and 1 atm were selected for use in establishing the entire homogeneity range for PuN. All of the above gases except for the last two are gaseous mixtures of nitrogen in argon, while the pressures of 10^{-1} atm and 1 atm are achieved with nitrogen alone.

In these experiments PuN powder contained in an open crucible suspended from the thermobalance was first outgassed at 1200 C. The constant weight in vacuum at 1200 C was selected as the starting point, and if a composition of $\text{PuN}_{1.00}$ is arbitrarily assigned to this composition all changes in composition can be referred to this point.

TABLE A-1. MINIMUM NITROGEN PRESSURE
FOR EXISTENCE OF PuN PHASE

Temperature, K	Nitrogen Pressure, atm
1400	8×10^{-13}
1600	4×10^{-11}
1800	7×10^{-9}
2000	4×10^{-7}
2200	1×10^{-5}
2400	2×10^{-4}

Temperatures from 1500 to 2140 C were investigated at 1 atm and at 0.1 atm, and after buoyancy corrections were made in the microbalance it was established that the charge gained no weight as a result of nitrogen uptake during these runs. Changes in the plutonium/nitrogen ratio in the third decimal place can be observed with this balance arrangement. It is concluded, then, that PuN (referred to $\text{PuN}_{1.00}$ in vacuum at 1200 C) does not exist at compositions richer in nitrogen than $\text{PuN}_{1.00}$. Of course it is possible that the partial molar free energy of nitrogen in the PuN phase rises so steeply that it is not possible to observe compositional changes when the pressure is changed only by one order of magnitude. Considering the sensitivity of the thermobalance, this is unlikely.

Additional experiments that were performed over the temperature range from 1720 to 1990 C indicated that the nitrogen/plutonium ratio varied from 0.991 at 1720 C to 0.968 at 1990 C at a constant nitrogen pressure of 1.0×10^{-4} atm. The experimental values are plotted in Figure A-1. On the basis of two-phase nitrogen pressures given in BMI-1872, it is calculated that the plutonium-PuN phase boundary at 1.0×10^{-4} atm occurs at 2067 C, or 2350 K. At a temperature of 2350 K, then, the PuN phase can exist at compositions to $\text{PuN}_{0.954}$. Some preliminary thermodynamic quantities have been calculated for the solution of nitrogen in PuN. The partial molal enthalpy $\bar{H} = -112,000$ cal per mole, while the partial molar entropy $\bar{S} = -38.5$ cal/(mole)(deg). The above values were calculated over the temperature range from 2000 to 2200 K. The integral enthalpy change across the homogeneity range is about 4 kcal per mole, which is sufficient to account for some of the differences in first-law and second-law thermodynamic investigations of PuN.

The data obtained to date indicate a moderately small homogeneity range for PuN, only slightly larger than that of UN. Similar experiments with $(\text{U}_{0.8}\text{Pu}_{0.2})\text{N}$ have not indicated any evidence for a homogeneity range in the mixed fuel. The very slight weight change that is observed at temperatures above 2000 C shows a constant rate of change with time. This is indicative of a zeroth-order reaction, and it is considered to be caused by volatilization of plutonium in a gaseous environment.

A series of determinations has been performed to determine the decomposition pressure of the sesquinitride when it is in equilibrium with $(\text{U}, \text{Pu})\text{N}$ fuels. Charges of between 1.5 and 2.0 g of $(\text{U}_{0.8}\text{Pu}_{0.2})\text{N}$ containing approximately 3 m/o U_2N_3 were employed in the determinations. The average particle size of the powder employed was 3μ ; hence, there was adequate surface for maintaining equilibrium conditions within the Knudsen cell. The cell employed was constructed of tungsten and contained a 0.1-cm-diameter orifice with a Clausing factor of 0.92. The cell was suspended from the Cahn electrobalance into the hot zone of a vacuum furnace. Results of the runs are shown in Figure A-2. The phenomenological interpretation of the results is that there is very little or no solubility of plutonium in U_2N_3 . This can be demonstrated thermodynamically in the following manner.

FIGURE A-1. PuN HOMOGENEITY RANGE AT 1.0×10^{-4} ATM OF NITROGEN

FIGURE A-2. DECOMPOSITION PRESSURES OF NITROGEN-RICH NITRIDE FUELS

Consider first that the standard states are defined as pure $UN_{1.5}$, pure UN, and N_2 gas all at the pressure and temperature of the individual measurement. For the decomposition of $UN_{1.5}$:

$$UN_{1.5} = UN + 0.25N_2 \quad (A-1)$$

$$\text{The equilibrium constant, } K = \frac{a(UN) (p^\circ N_2)^{0.25}}{a(UN_{1.5})} \quad (A-2)$$

or since UN and $UN_{1.5}$ are in their standard states, $a = 1$. Then,

$$K = (p^\circ N_2)^{0.25} \quad (A-3)$$

where p° has been used to refer to the pressure for the pure components for the fuel.

$$UN_{1.5} = UN_{\text{soln}} + 0.25N_2 \quad (A-4)$$

$$K = \frac{a(UN) (pN_2)^{0.25}}{a(UN_{1.5})} \quad (A-5)$$

Since we are referring to the same reaction, the K in Equation (A-3) is the same K as in Equation (A-5). Then

$$(p^\circ N_2)^{0.25} = \frac{a(UN) (pN_2)^{0.25}}{a(UN_{1.5})} \quad (A-6)$$

Previous vapor-pressure determinations have established that UN-PuN is essentially an ideal solution; hence, $a(UN_{\text{soln}}) = 0.8$, and if we assume as a first approximation that $a(UN_{1.5}) = 1$, then

$$\left(\frac{p^\circ N_2}{pN_2} \right)^{0.25} = 0.8 \quad (A-7)$$

or

$$\frac{p^\circ N_2}{pN_2} = 0.41 \quad (A-8)$$

This implies that the decomposition pressure over the sesquinitride in equilibrium with $(U_{0.8}Pu_{0.2})N$ would be 2.5 times higher than for the sesquinitride in equilibrium with UN, provided the activity of $UN_{1.5}$ was the same in both cases, i. e., no appreciable plutonium solubility in $UN_{1.5}$. Since this is the value measured, it certainly appears that the initial assumption of no plutonium solubility was warranted, and such is the case.

It is apparent that nitriding conditions are more severe for $UN_{1.5}-(U_{0.8}Pu_{0.2})N$ than for $UN_{1.5}$ -UN, at least based on equilibrium considerations; however, the pressures, although higher, are not of sufficient magnitude to influence any other properties in the system.

Plans for Future Work

Additional studies of the thermochemistry of mixed-nitride fuels will be undertaken.

(U, Pu)N Compatibility With Reactor Claddings and Coolants

J. M. Fackelmann, D. P. Moak, and W. H. Pfeifer

Objective of Program

To investigate the compatibility of UN-PuN fuels with potential cladding materials under simulated LMFBR conditions.

Approach Being Used

Studies will be primarily directed toward evaluation of the compatibility of $(U_{0.8}Pu_{0.2})N$ with Type 316 stainless steel in both sodium and helium environments. Studies in progress with Type 304 stainless steel claddings will be concluded. The effect of the presence of sesquinitride in hyperstoichiometric nitride fuels on compatibility with Type 316 stainless steel will be studied.

Fuel-cladding compatibility following in-pile irradiation will be evaluated. In addition, limited out-of-pile studies will be planned in support of the irradiation program.

Metallography, hardness measurements, electron-microprobe analyses, X-ray diffraction techniques, and analytical chemistry techniques will be used as required to identify reaction products and their rates of formation. Procedures for the preparation of compatibility specimens are outlined in BMI-1868.

Significant Accomplishments During Fiscal Year 1970

Powder mixtures of UN-10 v/o U_2N_3 , sodium bonded to Type 316 stainless steel, were found to react with the stainless steel to form diffusion zones rich in nitrogen. On the basis of extrapolation of results obtained in tests of up to 8000 hr in duration, the magnitude of this effect at 700 C is such that a diffusion zone of less than 1 mil in thickness will form in 10,000 hr. When dense UN-5 v/o U_2N_3 is substituted for the powder, the rate of cladding nitriding is significantly decreased. Contact bonding, as opposed to sodium bonding, also reduces nitriding effects at 700 and 800 C.

The low strain failure which had been observed in the cladding of mixed-nitride-fueled specimens was traced to the low ductility of the original as-received material rather than to excessive embrittlement during irradiation.

Summary of Research Carried Out

Heat treatment of UN_{1+x} -sodium-Type 316 stainless steel combinations has been performed in order to study compatibility behavior. The fuels and sodium were sealed in stainless steel capsules by Heliarc welding, and then heat treated for 8000 hr at 700 and 800 C under the static helium atmosphere of an outer Inconel capsule, also sealed by Heliarc welding. Two of these double capsules, containing powder mixtures of UN-10 v/o U_2N_3 , were heat treated for 8000 hr at 700 and 800 C, respectively. The U_2N_3 was heated for 1/2 hr at 500 C in vacuum to decrease its nitrogen content to a value near that at which the mononitride and sesquinitride are in equilibrium. A third capsule, containing 95 percent dense UN-5 v/o U_2N_3 as fuel, was heat treated for 8000 hr at 700 C. After heating, the double capsules were disassembled and examined by analytical and metallographic techniques.

The nitrogen profiles of these capsules, as determined by micro-Kjeldahl analyses of layers machined from the capsule wall at 10-mil increments, are presented in Figure A-3. The nitriding effects were also manifested in the microstructure by the

presence of a Cr_2N precipitate in the stainless steel adjacent to the cladding. Nitriding of the cladding proceeded at a higher rate with the powder fuel. The greater surface area of this fuel leads to a greater nitrogen flux, suggesting that the rate-limiting step in the nitriding process is the diffusion of nitrogen through UN.

The rate of cladding nitriding as a function of reciprocal temperature is plotted in Figure A-4 for UN-10 v/o U_2N_3 powder, sodium bonded to Type 316 stainless steel. The reaction rate in this plot is based on the metallographically measured thickness of the cladding zone containing the Cr_2N precipitate. The second curve is from previous studies, and provides a comparison with contact-bonded fuel elements. From Figure A-4, it is evident that the presence of sodium leads to a significant increase in nitriding rate. It should be noted, however, that past studies with dense, hyperstoichiometric fuels have indicated no significant difference in reaction rates for the two bonding conditions. The curves in Figure A-4 give a quantitative expression to the cladding nitriding kinetics for this fuel. Because of the high surface area and U_2N_3 content relative to actual LMFBR fuels, these curves probably represent upper limits of nitriding to be expected under these conditions. From Figure A-4, the calculated maximum nitride depth after 10,000 hr at 700 C is <1 mil.

In BMI-1868 it was suggested that the nitriding reaction,

reversed at low temperatures, which could result in the formation of U_2N_3 on the surface of UN fuels during cooling of compatibility specimens. In support of this belief it was noted that such surface zones were observed on UN particles in UN-10 v/o U_2N_3 powders after cooling from 800 to 1000 C, as shown in Figure A-5. In addition, chemical analysis of layers machined from the Type 316 stainless steel claddings showed decreasing surface nitrogen content with increasing heat-treating temperature. This effect was reversed in the body of the cladding, and the nitrogen content of the cladding in general increased with the heat-treating temperature as expected. It was suspected that the reverse reaction had occurred during cooling to deplete nitrogen from the stainless steel surface and to form U_2N_3 on the fuel. Two additional heat treatments were conducted in an attempt to reverse the reaction under controlled conditions. Previously nitrided pieces of Type 316 stainless steel were heated with UN under sodium for 72 hr at 500 and 600 C. No metallographic evidence of the reverse reaction was observed; neither depletion of Cr_2N from the surface of the nitrided steel nor U_2N_3 formation on the UN were seen. It therefore appears that the reverse of Reaction (A-9) does not occur at a significant rate at 500 or 600 C. At lower temperatures it is improbable that the rate would be sufficient to produce a visible reaction during the times necessary for the cooling of compatibility capsules. It is concluded that the observations noted above are not caused by the reverse of Reaction (A-9) during cooling.

An alternative explanation for the U_2N_3 observed as a surface phase on UN in capsules heated at 800 and 1000 C is that the UN reacts with nitrogen held in the sodium and capsule plenum during cooling. At temperature, the decomposing U_2N_3 establishes an equilibrium nitrogen activity in the capsule atmosphere. As the temperature decreases during cooling, this nitrogen activity is greater than that in equilibrium with $(\text{UN} + \text{U}_2\text{N}_3)$ at the lower temperature. Therefore, U_2N_3 forms to decrease the activity of nitrogen in the sodium. The apparent depletion of nitrogen from the cladding surface is not explainable at present.

In contrast to hyperstoichiometric UN fuels, (U,Pu)N-based fuels that contain sesquinitride and (U,Pu) O_2 as minor phases show no detectable reaction with Type 316 stainless steel. Several specimens containing mixed-nitride fuel but without (U,Pu) O_2 have been fabricated, along with all the necessary capsule components for heat treating.

FIGURE A-3. NITROGEN PENETRATION INTO TYPE 316 STAINLESS STEEL HEATED WITH UN- U_2N_3 FUELS

The base-line nitrogen content of the stainless steel is 0.053 0.001 w/o.

FIGURE A-4. THE RATE OF Cr_2N FORMATION IN TYPE 316 STAINLESS STEEL HEATED WITH UN-10 v/o U_2N_3 POWDER

These are to be tested in order to more fully define the role of the sesquinitride in (U, Pu)N fuels.

An effort in support of the irradiation task was undertaken when the Type 304L stainless steel claddings of specimens from the thermal irradiation capsules were found to have failed at what were considered relatively low strains (i. e. , approximately 0.5 percent). It was uncertain whether the low ductility was characteristic of the particular cladding material, which had been prepared from rod stock by drilling, as contrasted with normally used commercial seamless tubing, or whether the rod had embrittled during irradiation, possibly as a result of contact with the nitride fuel. Consequently, burst tests were performed on tubing produced by drilling and on commercial seamless tubing to compare their ductilities. The test specimens, as illustrated in Figure A-6, were tubes having the following dimensions:

Inside diameter	0.361 in.
Outside diameter, ends	0.501 in.
Outside diameter, reduced section	0.391 in.
Reduced-section length	1-1/8 in.

After the electron-beam welding of end caps on one end, the specimen was connected to a source of argon with stainless steel fittings. Internal argon pressure was then applied at approximately 1500 psi per min by means of oil pressure on a piston, with this isostatic pressure being measured and recorded as a function of time by means of a transducer-recorder apparatus. After the pressure required to burst the tubing was observed, a second test specimen was pressurized to about 90 percent of the burst pressure and the plastic deformation, as manifested by the expansion in diameter, was measured. Although the final diameter of burst specimens could not be measured directly because the specimens were blown apart, a rough measure was obtained by measuring the circumferential length of the pieces. Tests were conducted at room temperature and 650 C, with total time at 650 C usually less than 15 min.

Data obtained from these tests are summarized in Table A-2, and tested specimens are shown in Figures A-7 and A-8. The results show that the commercial tubing was much more ductile and weaker than the drilled rod at both temperatures. The maximum ductility observed for the machined-rod tubing was <0.5 percent at either temperature, whereas the diameter expansion of the commercially fabricated tubing was 11 to 22 percent at room temperature, and 3 to 4 percent at 650 C.

Although the rupture-strength values of the commercial tubing corresponded reasonably well with those expected on the basis of tensile tests for fully annealed tubing⁽¹⁾, the observed ductility values were considerably lower. This was not unexpected in view of the multiaxial stress state encountered in a tube-burst test. In the case of the drilled rod, the rupture strength and microstructure corresponded to those of a slightly cold-worked structure (≈ 10 percent), and ductility was essentially nil, compared to room-temperature tensile elongations of about 40 percent as reported in the literature for tubing in this condition⁽¹⁾.

On the basis of these limited tests, it was concluded that the drilled-rod cladding exhibits an unusually low ductility not characteristic of normal commercial tubing. Consequently, an unusual embrittling effect during irradiation need not be considered in explaining the low-strain failures encountered in the cladding of nitride-fueled specimens.

500X

9D209

FIGURE A-5. UN-10 v/o U_2N_3 POWDERS SODIUM BONDED TO TYPE 316 STAINLESS STEEL FOR 3000 HR AT 800 C

Stained areas are UN while the white areas are U_2N_3 . The secondary U_2N_3 formation on the surface of the particle in the center of the field was not seen after 700 C heat treatments and became a continuous film after heat treatments at 1000 C.

~1X

FIGURE A-6. TUBE-BURST TEST SPECIMEN

~1X

FIGURE A-7. APPEARANCE OF TYPE 304L STAINLESS STEEL DRILLED-ROD SPECIMENS AFTER "BURST TESTING" AT ROOM TEMPERATURE (SPECIMENS 199 AND 200) AND 650 C (SPECIMENS 202 AND 203)

~1X

FIGURE A-8. APPEARANCE OF TYPE 304L STAINLESS STEEL COMMERCIAL TUBE SPECIMENS AFTER "BURST TESTING" AT ROOM TEMPERATURE (SPECIMENS 193 AND 194) AND 650 C (SPECIMENS 196 AND 197)

TABLE A-2. RESULTS OF BURST TESTS ON 15-MIL-WALL, TYPE 304L STAINLESS STEEL TUBE SPECIMENS

Specimen	Test Temperature, C	Maximum Gas Pressure, psi	Maximum Hoop Stress, ksi	Diameter Swelling, percent
<u>Commercial Tubing</u>				
193	Room	5200	62.4 ^(a)	22 ^(b)
194	Room	4500	54.0	11
196	650	2500	30.0 ^(a)	4 ^(b)
197	650	2200	26.4	3
<u>Drilled Rod</u>				
199	Room	8800	105.6 ^(a)	0 ^(b)
200	Room	7800	93.6	0
202	650	4600	55.2 ^(a)	0 ^(b)
203	650	4100	49.2	0.3

(a) Value also represents rupture stress, since tube ruptured.

(b) Value obtained by measuring length of irregular ruptured segment along circumference.

Plans for Future Work

Oxide-free, hyperstoichiometric, mixed-nitride fuel pellets will be assembled in sodium-bonded stainless steel capsules and heat treated for various times at 700 C. In addition, irradiation specimens will be examined as they are received and prepared at the Battelle-Columbus Hot-Cell Facility. Other compatibility studies pertinent to LMFBR's will also be performed as considered necessary.

(1) Metals Handbook, American Society for Metals (1961).

Chemical Analysis of Irradiation-Capsule Sodium Bonds

P. L. Morris, O. L. Kruger, and W. M. Pardue

Objective of Program

To develop a reliable analytical method for the determination of nitrogen in the sodium used to bond (U, Pu)N irradiation specimens.

Approach Being Used

The approach planned for this program included conduct of a literature search to review various methods reported for analysis of nitrogen in sodium. A cursory review had indicated that very few methods have been developed and that the reliability and accuracy of these techniques are not well defined. After completion of the literature search, an evaluation of the various methods was to be made to determine their adaptability to plutonium glove-box and hot-cell operations. If methods in the literature did not appear to be adaptable, attempts were to be made to develop a suitable analytical technique. Once a choice had been made, effort on the program was to be directed to the following:

- (1) Procurement or synthesis of standard sodium samples and their subsequent analysis for nitrogen to determine the accuracy and reliability of the method
- (2) Determination of the total nitrogen level in the high-purity sodium used to bond (U, Pu)N irradiation fuel specimens

- (3) Analysis of a sodium sample for nitrogen content before and after bonding with (U,Pu)N specimens in a fuel pin
- (4) Transfer of the technique to the Battelle-Columbus Hot-Cell Facility to evaluate the effect of irradiation on nitrogen pickup or transfer by the sodium.

Significant Accomplishments During Fiscal Year 1970

During this fiscal year, studies were conducted to evaluate a modified micro-Kjeldahl method for determining nitrogen in sodium. Sodium samples were dissolved quickly and safely in an ethyl alcohol-water mixture. A combined reagent and procedural blank was accurately determined. Standard solutions were analyzed and nearly 100 percent nitrogen recoveries were noted. Analysis of high-purity sodium samples "spiked" with nitrogen in the approximate range of 10 to 1000 ppm indicated that only "nitride" nitrogen could be accurately determined by this method. The simplicity of the method and apparatus indicate that the procedure can be readily adapted to hot-cell operations.

Summary of Research Carried Out

The work conducted during the fiscal year was directed toward development of a reliable analytical method for the determination of nitrogen in the sodium used to bond (U,Pu)N to metallic specimens claddings. Few reliable methods existed for this purpose and development of a good analytical technique was warranted to expand the understanding of possible (U,Pu)N fuel-cladding-sodium chemical reactions. Such an analysis would shed light on possible transfer mechanisms of materials in the system and permit a more complete understanding of fuel behavior during irradiation. Adaptability of the method to plutonium glove boxes and hot-cell operations was a prime consideration.

The initial effort of the program was directed toward a literature search of the reported methods of nitrogen analysis in sodium and a critical review of these methods. A modified Kjeldahl technique was chosen for further evaluation⁽¹⁾. This method is simple, does not require an ultrahigh-purity atmosphere, and reportedly assures 100 percent recovery of the "nitride" nitrogen in a sodium sample. Briefly, a sodium sample is decomposed to NaOH in an ethyl alcohol-water mixture under an argon purge, whereupon the nitrogen present reacts with the NaOH to form ammonia. Subsequently, the ammonia is collected by steam distillation and determined titrimetrically with standard acid. After selection of this method, a modified micro-Kjeldahl apparatus was designed and fabricated. The apparatus was constructed in such a manner as to allow dissolution of sodium samples under an argon purge to prevent any possibility of fire or explosion from the hydrogen evolved.

The initial experimental work was directed toward determining the combined reagent and procedural blank. This was necessary as the blank level establishes the lower limit of detection of the method. Ten blank runs, involving no sodium metal, were made and subsequently analyzed for nitrogen content. The average blank value of the ten determinations was 1.820×10^{-5} g of nitrogen. This value corresponds to 9.1 ± 2.5 ppm nitrogen in a 2-g sodium sample, which is the normal weight of sodium taken for analysis. This indicated that the minimum detection limit of the method is about 10 ppm nitrogen. Subsequently, nine samples of high-purity sodium (99.99 percent) were analyzed for nitrogen to determine the residual nitrogen content. These analyses were performed as a base line because the high-purity samples were used later to establish the accuracy of the method. Data from these analyses indicated an average nitrogen level of 16.3 ± 1.8 ppm. By subtraction, it can be seen that the average residual nitrogen content of the sodium was about 7.2 ± 3.1 ppm.

The next phase of experimental work was concentrated on establishing the reliability of the method. Solution standards containing 0.1 and 1.0 mg of nitrogen, respectively, were analyzed to determine the relative recoveries that could be expected. These data

indicated that at the 0.1-mg level the average nitrogen recovery of three standards was 97 percent and at the 1.0-mg level the average nitrogen recovery of six standards was 97.3 percent, suggesting that nearly 100 percent nitrogen recoveries can be expected employing this method.

At this point, high-purity sodium samples were "spiked" with "nitride" nitrogen in the form of aqueous NH_4Cl . The samples were then analyzed. Data from this experiment, summarized in Table A-3, indicated that "nitride" nitrogen can be completely and accurately recovered from sodium samples in the ranges studied.

TABLE A-3. RECOVERY OF NITROGEN FROM HIGH-PURITY SODIUM SAMPLES

Amount of Nitrogen Added ^(a) , ppm	Amount of Nitrogen Recovered ^(b) , ppm
1213	1202 ± 5
123	126 ± 4
11.3	10.5 ± 3.3

(a) Nitrogen added in form of NH_4Cl solution.

(b) Values corrected for blank and yield and indicating standard deviation.

The studies completed during Fiscal Year 1970 indicated that sodium can be analyzed for nitrogen content with a reasonable degree of confidence. It is emphasized that at this time the method is applicable only to "nitride" nitrogen. Sodium samples were dissolved rapidly without fire or explosions employing a modified micro-Kjeldahl apparatus. The method is sufficiently developed for transferral to the Battelle-Columbus Hot-Cell Facility to evaluate the effect of irradiation on nitrogen pickup or transfer by the sodium.

Plans for Future Work

No future work is planned on this program.

- (1) Ward, E. H., et al., "The Titrimetric Determination of Traces of Nitride Nitrogen in Sodium Metal", LA-2892 (April, 1963).

Diffusion in Nitrides

J. W. Droege and C. A. Alexander

Objective of Program

To investigate the self-diffusion behavior in nitride fuels of interest to LMFBR applications.

Approach Being Used

Initially the self-diffusion behavior of nitrogen in UN will be determined in the temperature range from 1360 to 1800 C and the nitrogen pressure range of 0.5 to 10 torr. An isotopic tracer technique using N^{15} is employed. The sample is sectioned electrolytically and the nitrogen evolved is analyzed with a mass spectrometer. The primary purpose of the program is to develop techniques that will then be usable to determine diffusion characteristics of (U, Pu)N materials.

Significant Accomplishments During Fiscal Year 1970

An electrolytic sectioning technique has been developed to give suitable results on sections as thin as 0.5μ . This makes possible the determination of diffusion coefficients of the order of 10^{-14} cm^2 per sec in reasonable annealing times. Diffusion anneals of UN under several conditions of temperature and pressure have yielded diffusion coefficients considerably lower than those previously obtained by others.

Summary of Research Carried Out

Diffusion anneals were carried out in a small furnace containing a cylindrical tungsten-mesh heater surrounded by molybdenum shields. The specimen was suspended inside the heater by a thermocouple of tungsten-3 w/o rhenium versus tungsten-26 w/o rhenium. The junction was formed by twisting the 5-mil wires together, and was located inside a hole passing through the specimen. Temperatures were controlled manually with an autotransformer. Gas was circulated through the furnace by a Toepler pump. The gas passed from the pump through a cold trap and over a bed of hot CaH_2 . In this way any oxygen leak was converted to hydrogen before passing over the annealing specimen.

The specimens used in this work were arc-melted UN of fairly large grain size. Figure A-9 shows Specimen 69-1 prior to experimentation. The small-grained material along the curved edge was removed and not used in the experiment. Examination showed that slight quantities of uranium and U_2N_3 were present. Each specimen was therefore equilibrated with nitrogen at 1800 C for at least 24 hr before the N_2^{15} was introduced. Each annealing period was preceded by a period at least 2-1/2 times as long using N_2^{14} at the same temperature and pressure.

FIGURE A-9. UN SPECIMEN 69-1 BEFORE EXPERIMENTATION

Note that the large grains are separated by wide cracks.

The range of homogeneity of UN is not well known with respect to concentration, although the equilibrium nitrogen pressures corresponding to the limits of this composition range are known. The effect of composition on the diffusion coefficient may be studied by carrying out the equilibrium and diffusion anneal at a series of pressures. This has been done at one temperature by Holt and Almassy⁽¹⁾. To extend the treatment

to other temperatures requires more information about concentrations than is now available, but one may estimate the pressure-temperature relationship at constant composition (isopleth) and so approximate the condition of constant composition in expressing the effect of temperature on the diffusion coefficient.

After conditions of pressure, temperature, and composition were established by suitable equilibration periods in N_2^{14} , the gas was replaced with N_2^{15} , which was then circulated during the annealing period. The gas was replaced with fresh N_2^{15} at intervals depending on the temperature and pressure, so that the N_2^{15} composition of the gas did not drop too far by exchange at the UN surface. The gas composition did fluctuate somewhat, however, especially at low pressure. At the end of the annealing period the temperature and pressure were dropped quickly, and the specimen was removed and mounted for electrolysis.

Anodic dissolution of UN, with an inert electrode as the cathode, produces a solution of uranium and gaseous nitrogen, requiring about 6 Faradays per gfw of UN. With some electrolytes, and generally at low current densities, electrolytic etching takes place, attacking the specimen preferentially at the grain boundaries. With suitable choice of electrolyte and current density electrolytic polishing, which tends toward a smooth surface, can be achieved. This condition is required for the present application. Even under polishing conditions the grain boundaries are attacked with some slight degree of preference. A suitable polishing solution was found to consist of 8 parts isopropyl alcohol, 5 parts ethylene glycol, and 5 parts H_3PO_4 (86 percent). A current density of about 80 ma per cm^2 was used. The specimen was mounted in a fixture using RTV silicone sealant to hold it in place and to define the area to be exposed to the electrolyte. The thoroughly outgassed electrolyte was admitted to the evacuated cell. The electrolysis was conducted at a constant current for a measured time. The gas produced, including nitrogen from the anode and hydrogen from the cathode, was pumped through a cold trap and into a sample bulb with a Toepler pump. These samples, consisting of about 100 cm^3 torr of gas, were analyzed with a mass spectrometer for mass peaks 28, 29, and 30. The thickness of each "cut" was determined from the exposed area, the density of UN, and the number of coulombs passed in the electrolysis, together with the previously established relationship between coulombs and mass of UN dissolved.

The concentration, C , of N_2^{15} was plotted on probability paper and extrapolated to zero penetration to find the concentration at the surface, C_0 . Since the useful depth of penetration was of the order of 10 μ or less, the surface could be considered planar, even if the specimen had some curved surfaces. The applicable diffusion equation is

$$C/C_0 = \operatorname{erfc} \frac{x}{2\sqrt{Dt}} \quad , \quad (A-10)$$

where x is distance from the surface (cm), t is time (sec), and D is the diffusion coefficient. From tables of the error function, $x/2\sqrt{Dt}$ was evaluated for each value of C . This quantity was plotted as a function of x . Figure A-10 shows an example. D was obtained from the slope. The results follow the straight line well for the first few μ . At greater distances the deviations are larger. At these greater distances from the surface the concentrations of mass 29 and mass 30 remain higher than expected. It is suggested that this is probably due to penetration of the cracks between grains by the electrolyte, releasing N^{15} which had reached far into the specimen through cracks and grain-boundary diffusion.

Some of the results are summarized in Table A-4. By estimating pressure-temperature relationships within the homogeneity range of UN it is presumed that Specimens 69-2 and 66d were at the same composition while Specimens 69-1 and 66c contained less nitrogen. The first two results are somewhat in doubt because of the possibility of evaporation from the surface. Specimen 66c was annealed with an overpressure of argon to prevent this. Preliminary experiments without this precaution

showed evidence of evaporative loss of UN. There are also some indications of oxygen contamination, including a dull surface appearance in some cases and the release of some oxygen during the electrolysis. Figure A-11 shows a comparison of the results with those obtained by Sturiale and DeCrescente⁽²⁾ and by Holt and Almassy⁽¹⁾. The results lie well below those found previously. No explanation for the discrepancy is offered, especially since the latter authors used a similar technique, though the analytical method was much different. The two results at 1800 C show the effect of nitrogen pressure, and therefore of composition, on the diffusion coefficient. The effect is similar to that reported by Holt and Almassy at higher pressures and temperature.

TABLE A-4. ANNEALING CONDITIONS AND RESULT OF NITROGEN SELF-DIFFUSION IN UN

Specimen	Anneal Temperature, C	Anneal Time, hr	Nitrogen Pressure, torr	Diffusion Coefficient, cm ² per sec
66c	1800	12.4	0.5(a)	7.4×10^{-12}
69-1	1800	2	7	1.0×10^{-11}
69-2	1360	48	0.5	1.1×10^{-13}
66d	1580	48	10	3.7×10^{-12}

(a) Plus 9.5 torr argon.

Plans for Future Work

Modifications required for removal of the apparatus to the Battelle-Columbus Plutonium Laboratory will be made. These include principally some modifications of the techniques for handling and mounting the specimen. Work will then proceed with UN-PuN specimens.

(1) Holt, J. B., and Almassy, M. Y., J. Am. Ceram. Soc., 52, 631-635 (1969).

(2) Sturiale, T. J., and DeCrescente, M. A., "Self Diffusion of Nitrogen in Uranium Mononitride", PWAC-477 (September, 1965).

Bonding of Nitride Fuels and Cladding Materials by Sodium

R. B. Clark, O. L. Kruger, and W. M. Pardue

Objective of Program

To ascertain the influence of temperature, time, and gaseous atmosphere on the ability of liquid sodium to wet nitride fuel materials and various cladding materials of interest to LMFBR applications.

Approach Being Used

Initially, sessile-drop experiments will be performed over the temperature range from 100 to 700 C for sodium on Type 316 stainless steel and vanadium alloys and for sodium on high-density (U, Pu)N and possibly (U, Pu)O₂. These experiments will be performed in a glove box designed to contain an atmosphere of high-purity helium needed for sodium investigations. The atmosphere of the glove box will be monitored continuously for oxygen and water-vapor content. Contact angles between the liquid sodium and the fuel or cladding will be measured at selected temperatures and for various combinations of time at temperature. The wetting of the nitrides and claddings by the sodium will be

FIGURE A-10. N_2^{15} PENETRATION INTO UN SPECIMEN 69-2 ANNEALED AT 1360 C FOR 48 HR

FIGURE A-11. SELF-DIFFUSION COEFFICIENT FOR NITROGEN IN UN

recorded by photographic techniques for each of the various experimental conditions. Measurements are to be made during both heating and cooling cycles.

Significant Accomplishments During Fiscal Year 1970

Initial wetting studies of sodium on various fuel and cladding materials indicated that discrepancies in behavior can be expected when certain conditions are not properly controlled. In subsequent measurements it was found that sodium wets all of the materials studied between 350 and 500 C since it forms contact angles of less than 90 deg in this temperature range. Generally metals and alloys were wetted by sodium at lower temperatures than were ceramics. The contact angle-temperature relationship was non-reversible for all the materials studied since no dewetting was observed under any cooling conditions. No reactions or intergranular attack by the sodium was seen in the materials studied except for the (U,Pu)O₂, where spalling occurred at temperatures near 450 C. This work has shown that disintegration of the mixed-oxide surface by sodium takes place at low temperatures. Further studies on mixed oxides are recommended in support of the fast-reactor fuels program. It was also found that different lots of sodium do not have the same wetting characteristics on solid surfaces, particularly at low temperatures. This behavior has been attributed to the low solubility limits of various impurities such as oxygen in the sodium at the lower temperatures. At higher temperatures the solubility limits of sodium increase in such a manner that larger amounts of impurities are taken into solution, and the resulting data on various lots of sodium tend to fall closer together. Extrapolation of the contact angle-temperature curves, beyond the point where sodium volatilization occurs, to a contact angle of zero indicates that complete wetting would occur between about 600 and 650 C for all of the materials studied. This result, when analyzed in terms of the maximum size of bubble that can be formed at these temperatures, indicates that heat transfer and fission-gas release in sodium-bonded fuels is not a particularly acute problem.

Summary of Research Carried Out

Sodium wetting tests were performed to ascertain the influence of temperature, time, and gaseous atmosphere on the ability of liquid sodium to wet nitride fuel and various cladding materials of interest to the LMFBR program. The technique that seemed to lend itself most readily for measuring the contact angles of sodium on various materials was the sessile-drop method; further, these data are considered to be necessary in order to fully understand the parameters required for optimum bonding of fuel pins.

The fuel and cladding materials that were studied are shown in Table A-5. The wetting of sodium on an oxide, (U,Pu)O₂, was studied to determine the effect of an oxide layer which might exist on (U,Pu)N fuel. Three batches of sodium (designated Sodium 1, 2, and 3 in Table A-5) were investigated since early work showed that the results obtained from the initial experiments using various sources of sodium were slightly different. The same experimental technique was used throughout this work so as not to introduce another variable which would have to be factored into the analysis. Time and total effort considerations prevented a complete chemical analysis of the different lots of sodium; this is needed to obtain a better interpretation of the difference in the data.

Because of the high reactivity of the sodium, all wetting tests were performed in a glove box containing a high-purity helium atmosphere. Figure A-12 is a schematic drawing of the experimental setup. Initial experiments performed in a glove box indicated that the atmosphere contained some impurities which formed a white film on exposed sodium over a period of several minutes at the elevated testing temperatures. Various experimental procedures were employed to reduce the contamination problem, including high gas purge rates, but none were totally effective. To circumvent the contamination problem, a 6-in.-long stainless steel extension was adapted to the sodium extruder to permit direct loading of the sodium onto the substrate which was placed within the quartz tube of the sessile-drop apparatus. This tube was purged rapidly with ultrahigh-purity helium

FIGURE A-12. SCHEMATIC DIAGRAM OF THE SESSILE-DROP APPARATUS

so there was no possibility for the sodium to become contaminated during the transfer. When handled in this manner, the sodium appeared to stay very clean for the entire experimental run.

TABLE A-5. MATERIALS USED IN SODIUM WETTING STUDY

Fuel Materials
UN, hot pressed, 96 percent of theoretical density
$(U_{0.8}Pu_{0.2})N$, hot pressed, 95 percent of theoretical density
$(U_{0.92}Pu_{0.08})O_2$ sintered, 95 percent of theoretical density
Cladding Materials
Type 304 stainless steel
Type 304L stainless steel
Type 316 stainless steel
Vanadium-15 w/o titanium-7.5 w/o chromium
Vanadium-15 w/o chromium-5 w/o titanium
Sodium
Sodium 1, Battelle distilled
Sodium 2, Battelle distilled
Sodium 3, Gallard-Schlesinger Chemical Company

Cleanliness of the apparatus was imperative and all parts of the apparatus that came in contact with sodium were thoroughly cleaned and outgassed before any tests were made. Initial wetting tests showed that the state of the solid substrate surfaces greatly affected the wetting behavior. It was found that thorough cleaning and high-temperature vacuum outgassing gave the best wetting results. Each substrate surface was also polished in the high-purity helium atmosphere of the glove box with 600-grit paper immediately before loading into the sessile-drop apparatus to insure an initial oxide-free surface.

Isothermal tests of contact angle as a function of time at constant temperature were made in a helium atmosphere to determine the time necessary for equilibrium to be achieved. These tests showed that the drop assumed an equilibrium shape within 5 min at constant temperature since no change in contact angle was observed during longer periods of time. The only exception to this was in the very high temperature range, where volatilization of the sodium caused a change in drop size.

The isothermal results suggested that because of the comparatively small effect of time on the contact angle, except at high temperatures, true contact angle-temperature measurements could be made with sufficient accuracy by increasing the furnace temperature and holding the specimen for 5 min at constant temperature before measuring. A series of silhouette photographs was taken to give good contrast for contact-angle determinations.

A total of 33 sodium wetting experiments were performed. As a result of some of the early contamination problems only about 12 of these generated data acceptable for final reporting. Figures A-13 through A-16 are typical silhouette photographs taken for some of the materials studied. The wetting behavior of sodium on $(U, Pu)N$ is shown in Figure A-13. The sodium begins to visibly melt at 114 C but the sessile drop does not form until a temperature of about 210 C is reached. The drop does not change shape appreciably until a temperature of about 400 C is reached, where the contact angle begins

FIGURE A-13. CHANGE IN THE SODIUM/(U, Pu)N CONTACT ANGLE AT VARIOUS TEMPERATURES

FIGURE A-14. CHANGE IN THE SODIUM/UN CONTACT ANGLE AT VARIOUS TEMPERATURES

FIGURE A-15. CHANGE IN THE SODIUM/TYPE 316 STAINLESS STEEL CONTACT ANGLE AT VARIOUS TEMPERATURES

FIGURE A-16. CHANGE IN THE SODIUM/VANADIUM-15 w/o CHROMIUM-5 w/o TITANIUM CONTACT ANGLE AT VARIOUS TEMPERATURES

to sharply decrease. Wetting characteristics of sodium on UN (see Figure A-14) follow the same pattern as on (U, Pu)N; i. e., the sodium begins to wet the UN ($\theta < 90$ deg) at 455 C. Data for a typical run of sodium on Type 316 stainless steel can be seen in Figure A-15. The last photograph in this figure was taken after cooling to room temperature to determine whether dewetting occurred. The observed contact angle had decreased, probably because of an increase in temperature before cooling had started. Under no conditions was dewetting observed for the materials studied. Photographs showing the wetting behavior of sodium on vanadium-15 w/o chromium-5 w/o titanium are given in Figure A-16. The same type of contact angle-temperature relationship as was seen for the other materials is apparent.

The wetting behavior of the three batches of sodium mentioned previously is compared in Figures A-17 and A-18 for the UN and (U, Pu)N materials, respectively. The lower curve in Figure A-17 was generated using Sodium 1, the middle curve with Sodium 2, and the upper curve with the commercially purchased sodium. The curves are similar in shape but the data for the last two batches of sodium are displaced up and to the right. The curve for Sodium 2 in Figure A-18 is also displaced up and to the right, which is consistent with what was seen in Figure A-17. These results indicate that the differences among the curves are the result of actual differences in sodium and not in the experimental technique, since the relative positions of the curves with respect to the three types of sodium were consistent with all other substrate materials studied. Since volatilization was known to be occurring at the higher temperatures, Run 29 in Figure A-18 was purposely held at approximately 530 C for a period of about 30 min, after which the contact angle had decreased to zero. This behavior was obviously not due to equilibrium wetting but to volatilization of the sodium, since spreading was not simultaneously occurring.

Taylor and Ford⁽¹⁾ of Harwell did a study in 1955 of contact-angle relationships for sodium on various solids. Their results for stainless steel are compared to the Battelle data in Figure A-19. The relative position of the curves as a function of sodium is the same as was shown previously for UN and (U, Pu)N. This trend of behavior among different sodium batches appears to be much greater at the lower temperatures, where the curves are displaced the furthest with respect to one another. In the higher temperature regions, the contact angles tend to fall closer together, which suggests that the disparity among various batches of sodium would not cause a great deal of concern under reactor operating temperatures. Taylor and Ford show that the contact angle for sodium on stainless steel does not go below 60 deg, which by analogy with the data presented here can be attributed to contamination of the sodium. This type of behavior is typical of results first obtained at Battelle when the atmosphere was not purified to the extent later found to be required for this work.

Curves showing the wetting characteristics of Sodium 2 on vanadium-15 w/o titanium-7.5 w/o chromium and vanadium-15 w/o chromium-5 w/o titanium are given in Figure A-20. Both alloys appear to be completely wetted by the sodium between 540 and 660 C. There appears to be some indication that titanium would be wetted by sodium at a somewhat lower temperature than chromium; however, further work would be necessary to show this more conclusively. The wetting behavior of Types 304 and 316 stainless steel, UN, and (U, Pu)N is very similar to that of the vanadium alloys, as can be seen in Figure A-21. It appears that Types 304 and 316 stainless steel behave in a nearly identical manner, whereas the UN appears to be wetted more readily by sodium than does the mixed nitride, (U, Pu)N. In general, it was found that the metals are more readily wetted than the ceramic fuel materials.

The wetting of sodium on (U, Pu)O₂ was carried out to determine the effects of an oxide surface layer which might form on (U, Pu)N fuel. Because of the potentially high interfacial tension between sodium and most oxide materials, it was expected that mixed-oxide fuel would not be as readily wetted by the sodium. However, the two curves shown in Figure A-22 do not verify this; an entirely unexpected result was obtained. In both

FIGURE A-17. TEMPERATURE EFFECTS OF VARIOUS BATCHES OF SODIUM ON THE WETTING OF UN

FIGURE A-18. TEMPERATURE EFFECTS OF VARIOUS BATCHES OF SODIUM ON THE WETTING OF (U, Pu)N

FIGURE A-19. TEMPERATURE EFFECTS OF VARIOUS BATCHES OF SODIUM ON THE WETTING OF TYPE 304 STAINLESS STEEL

FIGURE A-20. TEMPERATURE EFFECTS OF SODIUM 2 WETTING ON VANADIUM ALLOYS

FIGURE A-21. TEMPERATURE EFFECTS OF SODIUM 2 WETTING ON VARIOUS FUEL AND CLADDING MATERIALS

FIGURE A-22. TEMPERATURE EFFECTS OF SODIUM 2 WETTING ON $(U,Pu)O_2$

cases an examination of the oxide-sodium interface after cooling indicated that a reaction or disintegration of the oxide surface had occurred. This reaction explains the differences between the two curves. Microstructures showing a typical, as-prepared interface and an interface examined after testing are given in Figure A-23. As can be seen, there is a general erosion and spalling of the surface after contact with sodium at these low temperatures. These results indicate that mixed-oxide fast-reactor fuels which are operated at considerably higher temperatures than investigated here might react spontaneously if they come into contact with the sodium coolant.

In general, for all of the materials studied, the contact angle-temperature curves can be divided into three areas. One occurs below the knee where the spreading of sodium becomes very sensitive to a change in temperature. Another occurs above the knee at lower temperatures where the slope of the curve is much lower. The third area is where the curve again changes slope and the contact angle is approaching the abscissa of the plot. In this region the contact-angle measurement is affected somewhat by volatilization of the sodium.

The following conclusions have been made regarding this work:

- (1) Sodium wets all of the materials studied between 350 and 500 C since it forms contact angles of less than 90 deg between these temperatures.
- (2) Sodium tends to wet metals and alloys at lower corresponding temperatures than ceramics.
- (3) The manner in which the solid surface of a fuel or cladding material is prepared before sodium loading can affect the manner in which sodium will wet its surface.
- (4) The contact angle-temperature relationship is nonreversible, since no dewetting was observed under any cooling conditions.
- (5) No reactions or intergranular attack by the sodium appear to occur in the materials studied except for the mixed oxide. Further studies of the wetting characteristics of mixed-oxide fuels are recommended in support of the fast-reactor fuels program.
- (6) Different lots of sodium do not wet solid surfaces the same particularly at low temperatures. This may be the result of the low solubility limits of various impurities such as oxygen in the sodium at the lower temperatures. At higher temperatures, where the solubility limits of sodium increase, the differences in wetting characteristics of the various batches of sodium were reduced significantly.
- (7) Extrapolation of the contact angle-temperature curves to $\theta = 0$ indicates that complete wetting of all the materials would occur between about 600 and 650 C.

Plans for Future Work

No further work is planned on this program.

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- (1) Taylor, J. W., and Ford, S. D., "Solid Metal-Liquid Metal Interaction Studies, Part II: Contact Angle Relationships for Sodium Angle of Solids", AERE-M/R-1729 (November 19, 1955).

Fabrication and Cladding ($U_{0.8}Pu_{0.2}$)N Thermal Irradiation Specimens

W. T. Langendorfer, M. D. Houston, E. O. Speidel, and O. L. Kruger

Objective of Program

To fabricate ($U_{0.8}Pu_{0.2}$)N pellets by powder metallurgical techniques and to clad them for use in irradiation experiments designed to evaluate fuel performance under thermal flux conditions.

250X

As Polished

PL7843

a. Typical As-Prepared $(U, Pu)O_2$ Substrate

250X

As Polished

PL7844

b. Inner Area of $(U, Pu)O_2$ Substrate That Was in Contact With SodiumFIGURE A-23. MICROSTRUCTURE OF $(U, Pu)O_2$ SUBSTRATE BEFORE AND AFTER REACTION WITH SODIUM

Approach Being Used

Sintering techniques are being used to fabricate ($U_{0.8}Pu_{0.2}$)N pellets of controlled density, size, and microstructure. Design variables in the thermal irradiation capsules include fuel density, bonding medium, and total diametral fuel-cladding gap.

Powders of the nominal fuel composition are ball milled to reduce the particle size, cold pressed in hardened steel dies, and sintered to pellets that are within ± 2.0 percent of the design densities. The final tolerances on diametral measurements for encapsulation are achieved by centerless grinding after sintering, if required. The pellets are right circular cylinders nominally 0.225 in. in diameter and are stacked to provide fuel specimens 0.75 in. long. Control pellets are characterized with respect to density, microstructure, chemistry, and purity.

The fuel pellets are encapsulated in Type 316 stainless steel cladding with sodium or helium as a heat-transfer medium in the annulus between fuel and cladding. The stainless steel cladding is first machined to dimensional specifications and an end plug is welded into one end of each tube prior to the fuel loading operation. All components are introduced into a glove box which contains a high-purity helium atmosphere.

For helium-bonded specimens, the components are assembled in the purified glove-box atmosphere and sealed by Heliarc welding the second end plug into position. Subsequent decontamination and inspection operations are performed as outlined previously in BMI-1809. The procedure for sodium bonding is similar except that the glove-box atmosphere is analyzed for oxygen content before introducing the sodium. The fuel cladding containing the mixed-nitride pellets is then heated to 300 C and a predetermined quantity of sodium is melted to fill the gap around the fuel specimens. An internal spacer is introduced and the second end plug is welded into position. Massive chill blocks are utilized during the welding procedure to prevent sodium boiling. To assure a continuous sodium bond, each fuel pin is heated to a temperature in excess of the melting point of sodium and centrifuged. The fuel pins are inspected by radiographic and eddy-current techniques to determine the weld penetration, the position and condition of the fuel pellets, and the integrity of the sodium bond.

Significant Accomplishments During Fiscal Year 1970

The fabrication, machining, and cladding of ($U_{0.8}Pu_{0.2}$)N pellets for Capsule BMI-50-12 and the characterization of control pellets were completed.

Summary of Research Carried Out

Work was devoted primarily to the fabrication of solid-solution ($U_{0.8}Pu_{0.2}$)N pellets for irradiation testing. Design variables are summarized in Table A-6. Dimensional, density, and encapsulation data for Capsule BMI-50-12 are summarized in Table A-7.

All fuel specimens were fabricated from nominal ($U_{0.8}Pu_{0.2}$)N powders synthesized using a hydride-nitride cycle on arc-cast uranium-20 w/o plutonium buttons. The as-synthesized powders were wet ball milled for 32 hr in a uranium-lined mill containing tungsten carbide-cobalt balls and nanograde hexane as the milling media. The ball-milled powders were vacuum dried to remove the hexane, and then pressed into pellet form in hardened steel dies using a pressure of 10 or 15 tsi with zinc stearate as a die lubricant. Each cold-pressed pellet had a length-to-diameter ratio of approximately 1 and a density approximately 50 percent of theoretical.

Low-density pellets were pressed at 10 tsi and sintered at 1700 C under 1 atm of nitrogen for a total of 6 hr. High-density pellets were pressed at 15 tsi and sintered for a total of 10 hr at 2100 C under 1 atm of nitrogen. Typical microstructures of the low-density and high-density pellets are shown in Figures A-24 and A-25, respectively.

TABLE A-6. DESIGN VARIABLES FOR EACH ($U_{0.8}Pu_{0.2}$)N FUEL CAPSULE BMI-50-12

Fuel Specimen	Density, percent of theoretical	Bonding Medium	Radial Gap Width, mils
1	95 ± 2(a)	Helium	2.5 ^{+1.0} -0.0
2	85 ± 2	Helium	2.5 ^{+1.0} -0.0
3	95 ± 2	Sodium	10.0 ± 0.5
4	85 ± 2	Sodium	10.0 ± 0.5
5	95 ± 2	Sodium	20.0 ± 0.5
6	95 ± 2	Sodium	20.0 ± 0.5

(a) Pellets will contain a 0.060-in. annular void to determine the effect of this type of porosity on accommodating fuel swelling during irradiation.

TABLE A-7. DATA FOR ($U_{0.8}Pu_{0.2}$)N IRRADIATION PELLETS PREPARED FOR CAPSULE BMI-50-12

Specimen	Pellet	Outside Diameter, in.	Pellet Density, percent of theoretical	Bonding Medium	Radial Gap Width, mils
1	A-29-11	0.2250(a)	93.0	Helium	2.5 ^{+1.0} -0.0
	A-29-12	0.2250(a)	94.0		
	A-29-43	0.2250(a)	94.9		
	A-29-44	0.2250(a)	95.4		
2	A-29-53	0.2250	84.4	Helium	2.5 ^{+1.0} -0.0
	A-29-56	0.2250	84.1		
	A-29-64	0.2250	84.4		
	A-29-65	0.2250	84.3		
3	A-29-18	0.2249	94.8	Sodium	10.0 ± 0.5
	A-29-25	0.2249	94.6		
	A-29-26	0.2249	94.2		
	A-29-27	0.2249	94.8		
4	A-29-66	0.2250	84.3	Sodium	10.0 ± 0.5
	A-29-68	0.2250	84.5		
	A-29-69	0.2250	84.4		
	A-29-67	0.2249	84.5		
5	A-29-29	0.2249	94.7	Sodium	20.0 ± 0.5
	A-29-32	0.2249	94.7		
	A-29-36	0.2249	94.1		
	A-29-49	0.2249	93.6		
6	A-29-14	0.2248	94.7	Sodium	20.0 ± 0.5
	A-29-31	0.2248	94.1		
	A-29-42	0.2248	94.5		
	A-29-46	0.2248	95.0		

(a) Pellet contains a 0.060-in. -diameter annular void.

500X

PL7430

a. As Polished

500X

PL7431

b. Etched

FIGURE A-24. MICROSTRUCTURE OF LOW-DENSITY $(U_{0.8}Pu_{0.2})N$ PELLET
Pellet A-29-73 is shown.

500X

PL7408

a. As Polished

500X

PL7414

b. Etched

FIGURE A-25. TYPICAL MICROSTRUCTURES FOR HIGH-DENSITY $(U_{0.8}Pu_{0.2})N$ PELLETS
Pellet A-29-19 is shown.

Characterization of the $(U_{0.8}Pu_{0.2})N$ densified pellets involved density measurements, with representative metallography and chemical analyses performed on control pellets. Analyses for control pellets are shown in Tables A-8 and A-9.

Type 316L stainless steel was selected as the cladding for these specimens and the chemical analysis of this material is shown in Table A-10. The fuel and cladding details for each specimen in Capsule BMI-50-12 are given in Table A-11. The loading and welding operations were carried out according to procedures described above. Subsequent qualifying tests, radiographs, and leak checks, indicated that each fuel specimen was acceptable for irradiation.

Plans for Future Work

Preparation of specimens for one capsule, BMI-50-13, is anticipated for Fiscal Year 1971.

Thermal Irradiation Studies of Nitride Fuels

E. O. Fromm, N. E. Miller, and J. W. Ray

Objective of Program

To irradiate UN-PuN fuels to burnups of 150,000 MWD/T at fission-heat-generation rates and temperatures typical of sodium-cooled fast-reactor operations.

Approach Being Used

A series of irradiation-capsule experiments involving $(U_{0.8}Pu_{0.2})N$ specimens is planned for exposure in the high-flux facilities of the ETR or MTR. The guiding criteria include:

- (1) Specimen temperature levels up to 700 C at the cladding surface
- (2) Fuel diameters of about 0.225 in.
- (3) Fission heat generation up to 40 kw per ft of fuel pin
- (4) Fuel burnup levels of 40,000 to 150,000 MWD/T.

The program includes design, construction, and loading of capsules, monitoring of the irradiation, and interpretation of the conditions of irradiation.

Significant Accomplishments During Fiscal Year 1970

The irradiation of Capsules BMI-50-6, BMI-50-8, and BMI-50-10 was continued. One new capsule, designated BMI-50-11, was assembled and sent to the ETR for irradiation beginning in December, 1969 (Cycle 105). This capsule was designed to operate at fission rates of 40 kw per ft with cladding surface temperatures of nominally 700 C. It was irradiated for two ETR cycles and discharged at the end of May, 1970, along with Capsule BMI-50-6 and returned to Battelle-Columbus for postirradiation examination. All of the components for Capsule BMI-50-12 were fabricated and, with insertion of fuel specimens, assembly was begun. This capsule is of the same design as Capsule BMI-50-11.

Summary of Research Carried Out

Each of the BMI-50-series irradiation capsules has contained six $(U_{0.8}Pu_{0.2})N$ fuel specimens (in pellet form) 0.225 in. in OD by 0.75 in. long contained in 0.015-in.-thick claddings of Type 304L stainless steel and having either a helium bond, a 10-mil sodium bond, or a 20-mil sodium bond between the fuel and cladding. Two clad specimens with the same type of bond are immersed in sodium within a thick-walled can. Three such cans are seal welded end to end to form a three-compartmented specimen container. Holes for six sheathed thermocouples are drilled longitudinally in the thick wall to accommodate thermocouples for monitoring specimen temperatures. One thermocouple junction is located adjacent to the midsection of each fuel specimen. Heat transfer

TABLE A-8. RESULTS OF VARIOUS ANALYSES OF (U_{0.8}Pu_{0.2})N FUEL PELLETS^(a)

Pellet	Nominal Density, percent of theoretical	Vacuum-Fusion Results, ppm		Nitrogen Content, w/o	Carbon Content, ppm	Wet-Chemical Results, w/o	
		Oxygen	Hydrogen			Uranium	Plutonium
A-29-4	95	292 ± 6	1 ± 1	NA	NA	NA	NA
A-29-5	95	90 ± 4	7 ± 1	5.35	NA	NA	NA
A-29-7	95	NA	NA	NA	200	NA	NA
A-29-8	95	NA	NA	NA	200	NA	NA
A-29-34	95	NA	NA	NA	NA	79.8	20.2
A-29-37	95	NA	NA	5.35	NA	79.4	20.6
A-29-51	85	176 ± 6	1 ± 1	5.45	NA	NA	NA
A-29-52	85	41 ± 2	26 ± 1	NA	NA	NA	NA
A-29-54	85	NA	NA	NA	110	NA	NA
A-29-55	85	NA	NA	NA	140	NA	NA
A-29-61	85	NA	NA	NA	NA	79.4	20.6
A-29-62	85	NA	NA	NA	NA	79.5	20.5
A-29-74	85	NA	NA	5.48	NA	NA	NA

(a) NA = not analyzed.

TABLE A-9. RESULTS OF EMISSION SPECTROGRAPHIC ANALYSES ON (U_{0.8}Pu_{0.2})N FUEL PELLETS

Pellet	Nominal Density, percent of theoretical	Impurity Content, ppm																
		Cu	Ag	Na	Mo	Sn	Ni	Mg	Mn	Pb	B	Al	Bi	Ca	Cr	Fe	Zn	W
A-29-37	95	<3	<0.3	<10	<5	<10	<10	<10	5	3	<0.3	30	<3	5	<10	<10	1	3
A-29-61	85	<3	<0.3	5	<1	<10	<5	<10	3	3	<0.3	30	<3	<1	<1	<10	1	3

TABLE A-10. CHEMICAL ANALYSIS OF TYPE 316L STAINLESS STEEL
CLADDING FOR THERMAL FUEL ENCAPSULATION

Element	Chemical Analysis, w/o	AISI Specifications, w/o
Aluminum	0.007	-
Boron	<0.001	-
Carbon	0.02	0.03
Chromium	17.0	17.00
Cobalt	0.04	-
Copper	0.07	-
Magnesium	<0.001	-
Manganese	1.69	2.00
Molybdenum	2.80	2.50
Nickel	13.60	12.00
Phosphorus	0.023	-
Silicon	0.47	1.00
Sulfur	0.012	-
Titanium	<0.001	-
Zirconium	0.001	-

TABLE A-11. SUMMARY OF FUEL AND CLADDING DATA FOR CAPSULE BMI-50-12

Fuel Specimen	Cladding Dimensions, in.			Fuel Average Dimensions ^(a) , in.		Total Fuel Weight, g	Average Density ^(a) , percent of theoretical	Bonding Medium
	OD	ID	Length	Diameter	Length			
D-2	0.260	0.230	1.770	0.225	0.700 ^(b)	5.9222	94.3	Helium
D-4	0.260	0.230	1.748	0.225	0.776	5.8768	84.3	Helium
E-1	0.275	0.245	1.780	0.225	0.752	6.5780	94.6	Sodium
E-2	0.275	0.245	1.770	0.225	0.747	5.8497	84.4	Sodium
H-2	0.295	0.265	1.775	0.225	0.747	6.5029	94.3	Sodium
H-3	0.295	0.265	1.766	0.225	0.751	6.5679	94.6	Sodium

(a) Total of four ($U_{0.8}Pu_{0.2}$)N pellets for each fuel specimen.

(b) Pellets have a 0.060-in. -diameter annular hole.

from the inner specimen container is through, successively, a 25-mil sodium annulus, a Hastelloy-X heater shell, another 25-mil sodium annulus, an "A" Nickel finned shell, and an "A" Nickel outer shell. The circumferential fins, which are only in the region adjacent to each fuel pin, make an interference fit with the outer shell. Since the heat-transfer path to the outer capsule wall is essentially fixed, the fuel temperature is governed by the heating rate of the fuel. In Capsules BMI-50-4 through BMI-50-10, the specimen container was "A" Nickel, which gave a temperature of about 600 C at the surface of the cladding of the helium-bonded specimen for a fuel-pin power rating of 30 kw per ft. In Capsule BMI-50-11, niobium-1 w/o zirconium was utilized for the specimen container to allow the heating rate to be increased to 40 kw per ft with a temperature of about 700 C on the surface of the cladding of the helium-bonded specimen. Electric heaters brazed into grooves in the heater shell keep the sodium molten in the various annuli during reactor startup. In addition, temperature-control capability up to 45 C is provided by the heaters. For added temperature control, the capsule is moved to different flux-level positions.

Capsule BMI-50-6 was the first of the BMI-50 series to be irradiated in the ETR and was inserted into a position (P-12-SW) with a neutron flux less than that required for design conditions in order to check out the operation at full reactor power. Subsequently, this capsule was moved to higher flux positions to obtain higher burnup rates and fuel temperatures, as indicated in Table A-12. The capsule was discharged in April, 1970 (at the end of ETR Cycle 106), with estimated fuel burnups of 102,000 to 157,000 MWD/T as shown in Table A-12. The variations in calculated burnups can be attributed in part to uncertainties introduced in estimates of burnup rates for some of the specimens because of thermocouple failures, which made it necessary to extrapolate temperatures (and burnups) for associated specimens from thermocouple data for adjacent specimens. The capsule was returned to Battelle-Columbus in May, 1970, for postirradiation examination.

Capsule BMI-50-8 was inserted in the aluminum reflector region of the ETR (P-9-SW) in July, 1968 (Cycle 97), where design irradiation conditions were achieved. However, as the irradiation progressed the fission rate continually declined, and the capsule was relocated into a higher flux position (E-5-NE) in July, 1969. Temperatures in the new position reflected the increase in flux level. The thermocouple opposite Specimen A-26 failed during January, 1970 (Cycle 106A), and the surface temperature for this specimen was extrapolated from thermocouple data from the adjacent specimen, A-25. In general, the specimen temperatures were below desired levels during Cycle 106 (January through April, 1970) and it was decided to relocate the capsule to a higher flux position for the next ETR cycle. Consequently, the capsule was moved into a new position (F-4) for Cycle 107. Preliminary temperatures during initial startup for the cycle in the new position indicated heating rates of about 35 kw per ft. Temperature data for Cycles 102 through 106B are given in Table A-13.

Capsule BMI-50-10 began irradiation in the corner of the reactor core (E-5-NW) during May, 1969 (ETR Cycle 102). The performance during the first cycle of irradiation was excellent, with the specimens operating in the range of 660 to 730 C with heating rates from 27 to 31 kw per ft. However, temperatures during the following cycles were much lower than for the first cycle because of the shielding effect of a new experiment in the companion facility, E-5-NE. It was not possible to relocate the capsule to a higher flux facility until April, 1970 (at the end of Cycle 106). At this time the capsule was moved into Position E-5-NE. Preliminary temperature data during startup for Cycle 107 (June, 1970) indicated heating rates of about 35 kw per ft. Specimen temperatures and burnup data for Cycles 102 through 106B are given in Table A-14.

Capsule BMI-50-11 was assembled and sent to the ETR for irradiation in a beryllium-reflector position (M-4) beginning in December, 1969 (Cycle 105). Temperatures during the cycle indicated fuel-pin power ratings of nominally 50 kw per ft with

TABLE A-12. SPECIMEN TEMPERATURES FOR CAPSULE BMI-50-6

ETR Cycle	Starting Date	Type of Temperature Data	Temperatures for Indicated Specimens ^(a) , °C					
			A-4	A-3 ^(b)	B-2 ^(c)	B-1	C-2 ^(d)	C-1 ^(e)
93 ^(f)	November 1967	Average	480	475	470	460	465	385
		Maximum	530	545	530	490	480	415
94 ^(g)	January 1968	Average	465	530	550	550	575	495
		Maximum	505	560	585	570	605	520
95	April	Average	535	605	630	600	670	565
		Maximum	600	630	650	630	680	570
96	May	Average	460	535	530	500	540	440
		Maximum	495	550	570	525	570	465
97	July	Average	465	530	555	510	560	455
		Maximum	485	555	580	535	575	475
98	September	Average	430	495	525	485	520	420
		Maximum	455	525	550	490	545	445
99	October	Average	410	475	490	440	480	390
		Maximum	440	505	505	460	505	405
100	January 1969	Average	440	510	540	475	540	440
		Maximum	470	535	565	495	465	460
101	February	Average	405	475	495	445	485	395
		Maximum	445	510	520	465	515	415
102 ^(h)	May	Average	605	680	670	545	565	460
		Maximum	680	735	720	560	590	485
103 ⁽ⁱ⁾	July	Average	650	725	720	620	650	550
		Maximum	705	790	780	715	675	570
104	September	Average	645	715	715	655	640	535
		Maximum	695	770	770	705	675	575
105	December	Average	600	670	670	625	655	555
		Maximum	630	705	705	650	680	575
106A ^(j)	January 1970	Average	630	705	705	665	700	640
		Maximum	725	800	800	720	750	645
106B ^(j)	March	Average	560	630	630	590	625	520
		Maximum	625	710	710	620	650	545
Total Accumulated Burnup								
10 ²⁰ Fissions per cm ³			39.7	47.7	48.6	43.1	46.3	35.0
10 ³ MWD/T ^(k)			130	132	157	123	131	102

- (a) Temperatures reported are extrapolated from thermocouple data and are on:
 Inner cladding surface for Specimens A-4 and A-3 (helium bond)
 Fuel surface of Specimens B-2 and B-1 (10-mil sodium bond)
 Fuel surface of Specimens C-2 and C-1 (20-mil sodium bond)
- (b) Thermocouple opposite Specimen A-3 failed at the end of Cycle 96. Temperatures for follow-on cycles were estimated using data from adjacent Specimen A-4.
- (c) Thermocouple opposite Specimen B-2 failed at the end of Cycle 98. Temperatures for follow-on cycles were estimated using data from adjacent Specimen B-1.
- (d) The thermocouple for Specimen C-2 failed on startup for Cycle 105. The temperatures for Specimens C-1 and C-2 for Cycles 105 and 106A and B were obtained using data from Specimen B-1.
- (e) Thermocouple opposite Specimen C-1 failed during Cycle 95. Temperatures for Cycles 95 through 104 were estimated using data from adjacent Specimen C-2.
- (f) Capsule initially inserted into Position P-12-SW for ETR Cycle 93.
- (g) Capsule transferred to Position P-10-SW for Cycles 94 through 101.
- (h) Capsule transferred from Position P-10-SW to beryllium-reflector Position 0-5 for Cycle 102.
- (i) Capsule transferred from Position 0-5 to Position 0-6 for Cycle 103.
- (j) Cycle 106A accumulated 5724 MWD, the length of a normal ETR cycle. Cycle 106B was approximately the same length.
- (k) Based on 200 Mev per fission.

TABLE A-13. SPECIMEN TEMPERATURES FOR CAPSULE BMI-50-8 DURING FISCAL YEAR 1970

ETR Cycle	Starting Date	Type of Temperature Data	Temperatures for Indicated Specimens ^(a) , C					
			A-25	A-26 ^(b)	B-25	B-28	C-25	C-26
102	May, 1969	Average	560	590	640	625	640	660
		Maximum	605	630	670	650	665	685
103 ^(c)	July	Average	595	620	780	700	755	880
		Maximum	690	715	825	760	800	940
104	September	Average	595	640	770	700	765	890
		Maximum	685	725	795	750	795	945
105	December	Average	515	560	670	595	665	795
		Maximum	600	635	715	640	685	800
106A ^(d)	January, 1970	Average	515	560	615	550	620	745
		Maximum	615	665	650	595	630	760
106B ^(d)	March	Average	505	550	615	540	605	735
		Maximum	590	630	640	570	620	755
Total Burnup Accumulated to Date:								
10 ²⁰ Fissions per Cm ³			31.5	34.6	42.6	38.4	42.0	47.9
10 ³ MWD/T ^(e)			105	99	139	110	121	136

(a) Temperatures reported are extrapolated from thermocouple data and are on:

Inner cladding surface for Specimens A-25 and A-26 (helium bond).

Fuel surface of Specimens B-25 and B-28 (10-mil sodium bond).

Fuel surface of Specimens C-25 and C-26 (20-mil sodium bond).

(b) Thermocouple opposite Specimen A-26 failed during Cycle 106A. Temperatures for Cycles 106A and 106B were estimated using data from adjacent Specimen A-25.

(c) Capsule transferred from Position P-9-SW to Position E-5-NE.

(d) Cycle 106A accumulated 5724 MWD, the length of a normal ETR cycle. Cycle 106B was approximately the same length.

(e) Based on 200 Mev per fission.

TABLE A-14. SPECIMEN TEMPERATURES FOR CAPSULE BMI-50-10 IN ETR FACILITY E-5-NW

ETR Cycle	Starting Date	Type of Temperature Data	Temperatures for Indicated Specimens ^(a) , C					
			A-41	A-51	B-41	B-42	C-41	C-42
102	May, 1969	Average	675	660	680	720	730	735
		Maximum	740	720	710	735	745	770
103	July	Average	555	545	555	570	605	635
		Maximum	640	595	580	595	625	660
104	September	Average	535	540	550	575	600	680
		Maximum	630	595	580	630	650	725
105	December	Average	470	490	500	525	570	590
		Maximum	520	530	515	540	580	660
106A ^(b)	January, 1970	Average	475	485	480	505	515	625
		Maximum	535	500	510	515	580	650
106B ^(b)	March	Average	475	490	485	510	545	615
		Maximum	525	525	520	520	570	645
Total Burnup Accumulated to Date:								
10 ²⁰ Fissions per Cm ³			15.6	15.7	15.9	16.9	17.8	20.1
10 ³ MWD/T(c)			59.5	53.3	61.6	53.6	60.0	70.4

(a) Temperatures reported are extrapolated from thermocouple data and are on:

Inner cladding surface for Specimens A-41 and A-51 (helium bond).

Fuel surface of Specimens B-41 and B-42 (10-mil sodium bond).

Fuel surface of Specimens C-41 and C-42 (20-mil sodium bond).

(b) Cycle 106A accumulated 5724 MWD, the length of a normal ETR cycle. Cycle 106B was approximately the same length.

(c) Based on 200 Mev per fission.

periodic fluctuating thermocouple output. As a result, the capsule was moved to a lower flux position (F-4) for the next cycle of irradiation. Nominal pin power ratings of 40 to 45 kw per ft were achieved in the new location. The capsule was discharged in May, 1970 (end of Cycle 106), and was returned to Battelle-Columbus for postirradiation examination. The temperature and burnup data for the two cycles of irradiation are given in Table A-15. Temperatures exceeded design goals for the experiment, with the fuel surface temperatures exceeding the atmospheric sodium boiling point in each specimen for the first cycle of irradiation.

The capsule components for Capsule BMI-50-12 were fabricated and assembly, with insertion of fuel specimens, was begun. The design of this capsule is identical to that of Capsule BMI-50-11.

TABLE A-15. TEMPERATURE HISTORY FOR CAPSULE BMI-50-11

ETR Cycle	Starting Date	Type of Temperature Data	Temperatures for Indicated Specimens ^(a) , C					
			A-43	A-44	B-43	B-54	C-43	C-44
105 ^(b)	December, 1969	Average	900	930	930	970	1055	1010
		Maximum	930	970	980	1015	1105	1035
106A ^(c, d)	January, 1970	Average	815	870	845	925	915	860
		Maximum	845	925	875	965	950	880
106B ^(d)	March	Average	780	835	830	875	900	855
		Maximum	850	895	865	955	960	925
Total Accumulated Burnup:								
10 ²⁰ Fissions per Cm ³			17.0	18.2	15.9	19.2	20.1	18.9
10 ³ MWD/T ^(e)			55.7	52.5	58.5	55.0	57.5	54.5

(a) Temperatures reported are extrapolated from thermocouple data and are on:

Inner cladding surface for Specimens A-43 and A-44 (helium bond).

Fuel surface of Specimens B-43 and B-54 (10-mil sodium bond).

Fuel surface of Specimens C-43 and C-44 (20-mil sodium bond).

(b) Capsule was initially inserted into Position M-4 for Cycle 105.

(c) Capsule was transferred from Position M-4 to Position F-4 for Cycle 106A.

(d) Cycle 106A accumulated 5724 MWD, the length of a normal ETR cycle. Cycle 106B was approximately the same length.

(e) Based on 200 Mev per fission.

Plans for Future Work

The irradiation of Capsules BMI-50-8 and BMI-50-10 will be continued to the end of Cycle 109, tentatively scheduled for August, 1970, at which time they will be discharged from the reactor and returned to Battelle-Columbus for postirradiation examination. Capsule BMI-50-12 will be assembled and sent to the ETR for irradiation. The irradiation of this capsule is tentatively scheduled to begin in September, 1970. It is anticipated that the target burnup of 100,000 MWD/T will be reached in approximately one calendar year.

Postirradiation Evaluation of (U_{0.8}Pu_{0.2})N Thermal
Irradiation Specimens

J. B. Brown, Jr., E. O. Fromm, V. W. Storhok, and V. G. Scotti

Objective of Program

To evaluate the stability of mixed-nitride fuels irradiated in a thermal-neutron flux at cladding temperatures up to 700 C, heat generation rates up to 40 kw per ft, and burnups up to 100,000 MWD/T.

Approach Being Used

Specimens consisting of helium-bonded and sodium-bonded (U_{0.8}Pu_{0.2})N pellets clad in 15-mil-thick Type 304L stainless steel will be examined following irradiation in the ETR or MTR. The postirradiation examination planned involves (1) measurement of fission-gas release from clad specimens to the capsule and from fuel to the cladding, (2) visual inspection of the cladding for defects or evidence of chemical reaction, (3) measurement of the dimensions and density of the specimens, and (4) metallographic examination of the cladding and fuel to determine cladding-fuel compatibility and changes in the microstructures of cladding and fuel. When appropriate, additional investigation techniques such as gamma scanning, autoradiography, X-ray diffraction analyses, electron-microprobe analyses, and fuel-burnup analyses will be used.

Significant Accomplishments During Fiscal Year 1970

During the year, the examination of fuel specimens from Capsules BMI-50-5, BMI-50-7, and BMI-50-9, plus some detailed examinations of claddings from Capsule BMI-50-4 were completed. Capsules BMI-50-6 and BMI-50-11 were discharged from the ETR late in the year and examinations were initiated.

In Capsules BMI-50-5, BMI-50-7, and BMI-50-9, burnups ranged from 11.0×10^{20} to 34.5×10^{20} fissions per cm³, and heat generation rates ranged from 13.5 to 29.2 kw per ft. Fuel swelling rates in these capsules were uniformly low at less than 0.5 per cent per 10^{20} fissions per cm³. None of the sodium-bonded specimens exhibited any cladding strain and the remaining sodium-filled annuli were large enough to accommodate considerably more fuel swelling. All the helium-bonded specimens experienced cladding strain and several had failed claddings. Fission-gas release from the high-density fuel was very low, being on the order expected from recoil from the free surfaces. The somewhat higher gas release from low-density specimens was attributed to the greater exposed surface area associated with the high degree of surface-connected porosity.

Consistent with the low swelling and low gas release was the absence of optically resolvable fission-gas bubbles in the fuel.

No evidence of interaction between the cladding and fuel or fission products was found by metallographic examination or electron-microprobe analysis. Electron-microprobe analysis also showed that no segregation of uranium and plutonium had occurred. Micro-Kjeldahl analysis indicated that the cladding did not pick up significant quantities of nitrogen during irradiation.

In Capsules BMI-50-6 and BMI-50-11, the irradiation conditions were more severe in terms of burnup (35×10^{20} to 48.6×10^{20} fissions per cm³ in Capsule BMI-50-6) or heat generation rates (41.6 to 49.0 kw per ft in Capsule BMI-50-11).

Preliminary results of the examination of fuel specimens from Capsule BMI-50-6 indicated large cladding strain in the helium-bonded specimens, a slight amount of cladding strain in the 10-mil sodium-bonded specimens, and no cladding strain in the 20-mil sodium-bonded specimens. Claddings on both helium-bonded specimens and one 10-mil sodium-bonded specimen were ruptured.

In Capsule BMI-50-11, all specimens experienced some cladding strain. However, only one 10-mil sodium-bonded specimen had a visibly failed cladding. This failure was oriented in a circumferential direction and did not appear to be caused by the hoop stresses generally associated with swelling fuel.

Summary of Research Carried Out

During the year, the postirradiation examinations of fuel specimens in Capsules BMI-50-5, BMI-50-7, and BMI-50-9 were completed and the examinations of fuel specimens from Capsules BMI-50-6 and BMI-50-11 were initiated. In addition, some detailed studies of claddings from specimens irradiated in Capsule BMI-50-4 were conducted.

The specimens in Capsule BMI-50-5 were irradiated to burnups of 23.8×10^{20} to 34.5×10^{20} fissions per cm^3 (69,000 to 105,000 MWD/T) at average linear heat generation rates of 13.5 to 19.6 kw per ft. In Capsule BMI-50-7, the fuel attained burnups of 28.5×10^{20} to 32.1×10^{20} fissions per cm^3 (85,000 to 97,000 MWD/T) at heat generation rates of 25.7 to 29.2 kw per ft. Capsule BMI-50-9 was discharged from the MTR when it shut down. Because of the low fission burnup, 11×10^{20} to 12.5×10^{20} fissions per cm^3 (34,000 to 41,000 MWD/T) the examination of specimens from Capsule BMI-50-9 was not extensive.

Specimen descriptions, irradiation conditions, and the results of postirradiation examinations for these capsules are summarized in Tables A-16 through A-18.

Macrophotographs and neutron radiographs of the as-recovered specimens from Capsule BMI-50-9 are shown in Figures A-26 and A-27. Macrophotographs and neutron radiographs of specimens from Capsules BMI-50-5 and BMI-50-7 were presented in BMI-1868.

Note from the tables that all of the sodium-bonded specimens from the capsules were recovered intact with no cladding strain. Four out of the six helium-bonded specimens from these capsules, on the other hand, had visibly failed claddings. These cladding failures were more common at the higher burnups and occurred at cladding strains as low as 0.5 percent. At the lower burnups experienced in Capsule BMI-50-9, the cladding strain in the helium-bonded specimens was low and no cladding failures were noted.

Volumetric and diametral swelling data obtained from specimens irradiated in Capsules BMI-50-4, BMI-50-5, BMI-50-7, and BMI-50-9 are plotted as a function of burnup in Figures A-28 and A-29. Note that volumetric swelling is uniformly low at less than 0.5 percent per 10^{20} fissions per cm^3 . Diametral swelling is likewise low. At burnups of $\sim 30 \times 10^{20}$ fissions per cm^3 , the remaining annular gaps in the 10-mil sodium-bonded specimens are sufficiently large to accommodate considerably more burnup before the fuel would contact the cladding. The swelling data give no consistent indication that swelling behavior is improved by decreasing fuel density. Evidently under the conditions of the irradiations, the fuel, even at the centerline, is sufficiently strong relative to the cladding or cooler outer regions of fuel to preclude fuel swelling into preexisting porosity.

Fission-gas release in all the specimens was low. Release from the high-density fuel was generally on the order expected from recoil at free surfaces. Somewhat higher gas release was measured when the fuel was extensively cracked, as with Specimen C-7 from Capsule BMI-50-5. This specimen also showed evidence of contamination. Release from low-density specimens was consistently higher. The low-density specimens presumably have a high degree of surface-connected porosity which accounts for their higher release rates.

Consistent with the low swelling rates and low gas release was the absence of optically resolvable fission-gas bubbles in the fuel. As shown in Figure A-30, the microstructures of irradiated and unirradiated fuel from Capsule BMI-50-7 are virtually identical.

4X

P1946-7

Specimen A-31

4X

P1944-5

Specimen A-32

4X

P1942-3

Specimen B-31

FIGURE A-26. IRRADIATED SPECIMENS FROM CAPSULE BMI-50-9

4X

P1940-1

Specimen B-32

4X

P1938-9

Specimen C-27

4X

P1936-7

Specimen C-31

FIGURE A-26. (CONTINUED)

Specimen
A-31
(helium bond)

Specimen
A-32
(helium bond)

Specimen
B-31
(sodium bond)

Specimen
B-32
(sodium bond)

Specimen
C-27
(sodium bond)

Specimen
C-31
(sodium bond)

0 Deg

120 Deg

240 Deg

FIGURE A-27. NEUTRON RADIOGRAPHS OF SPECIMENS FROM CAPSULE BMI-50-9

TABLE A-16. IRRADIATION CONDITIONS AND SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-5

	Specimen Designation					
	A-6	A-7	B-5	B-6	C-7	C-8
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	95.1	80.8	80.2	95.6	97.2	92.3
Smear	~95.1 ^(a)	~80.8 ^(a)	67.7	80.7	70.1	66.6
Burnup						
10^{20} Fissions per Cm^3	23.8	24.8	30.9	28.9	34.5	31.9
10^3 MWD/T	69	84	105	82	97	95
Fuel-Surface Temperature ^(b) , C						
Average	395	405	470	445	500	475
Maximum	465	460	535	505	630	630
Average Linear Heat Rating, kw per ft	13.5	14.1	17.6	16.3	19.6	18.1
Cladding Diameter Increase ^(c) , percent	2.2	0.5	0	0	0	0
Fuel Diameter Increase ^(d) , percent	2.5	0.6	1.2	2.6	5.7	2.9
Fuel Length Increase ^(e) , percent	3.4	3.5	4.4	5.4	5.2	2.9
Fuel Volume Increase						
Percent	8.4	4.7	6.8	10.6	16.6	8.7
Percent per 10^{20} Fissions per Cm^3	0.37	0.17	0.22	0.37	0.48	0.27
Fission-Gas Release, percent	--	--	13.5	0.8	4.8	--
Cladding Condition	Failed	Failed	Sound	Sound	Sound	Sound

(a) Sweat fit of cladding to fuel.

(b) Fuel temperature for helium-bonded specimens calculated assuming zero gap during irradiation. Average temperature is time average based on entire irradiation. Maximum value is that occurring during last half of irradiation.

(c) Micrometer measurements.

(d) Fuel diameter changes for helium-bonded specimens calculated assuming fuel-cladding contact; remaining diameter changes measured from neutron radiographs.

(e) Measured from neutron radiographs.

TABLE A-17. IRRADIATION CONDITIONS AND SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-7

	Specimen Designation					
	A-22	A-24	B-22	B-24	C-21	C-22
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	93.8	82.6	95.5	82.5	97.5	96.0
Smear	~93.8 ^(a)	~82.6 ^(a)	80.4	70.2	69.2	70.2
Burnup						
10^{20} Fissions per Cm^3	29.2	28.5	30.7	29.1	30.1	32.1
10^3 MWD/T	85	94	89	97	86	91
Fuel-Surface Temperature ^(b) , C						
Average	605	595	635	610	625	660
Maximum	650	660	650	650	735	765
Average Linear Heat Rating, kw per ft	26.3	25.7	28.0	26.3	27.3	29.2
Cladding Diameter Increase ^(c) , percent	1.6	1.1	0	0	0	0
Fuel Diameter Increase ^(d) , percent	1.8	1.3	2.3	3.9	3.3	2.9
Fuel Length Increase ^(e) , percent	3.2	4.1	4.2	3.7	5.6	2.1
Fuel Volume Increase						
Percent	6.8	6.7	8.8	11.5	12.2	7.9
Percent per 10^{20} Fissions per Cm^3	0.23	0.24	0.29	0.40	0.41	0.25
Fission-Gas Release, percent	--	--	0.4	9.2	--	0.4
Cladding Condition	Failed	Failed	Sound	Sound	Sound	Sound

(a) Sweat fit of cladding to fuel.

(b) Fuel temperature for helium-bonded specimens calculated assuming zero gap during irradiation. Average temperature is time average based on entire irradiation. Maximum value is that occurring during last half of irradiation.

(c) Micrometer measurements.

(d) Fuel diameter changes for helium-bonded specimens calculated assuming fuel-cladding contact; remaining diameter changes measured from neutron radiographs.

(e) Measured from neutron radiographs.

TABLE A-18. SWELLING DATA FOR FUEL SPECIMENS IRRADIATED IN CAPSULE BMI-50-9

	Specimen Designation					
	A-31	A-32	B-31	B-32	C-27	C-31
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Pellet Density, percent of theoretical	82.6	94.2	94.6	82.4	94.2	94.4
Burnup ^(a)						
10^{20} Fissions per Cm^3	11.0	11.8	12.5	12.4	12.0	11.4
10^3 MWD/T	36.0	34.2	37.8	41.0	34.5	34.0
Cladding Diameter Increase, percent	0.24	0.55	0	0	0	0
Fuel Diameter Increase, percent	0.27 ^(b)	0.63 ^(b)	0.49 ^(c)	0.49 ^(c)	0.13 ^(c)	2.0 ^(c)
Fuel Length Increase ^(c) , percent	0.0	0.0	2.0	1.9	1.4	1.1
Fuel Volume Increase						
Percent	0.54	1.3	3.0	2.9	1.7	5.1
Percent per 10^{20} Fissions per Cm^3	0.05	0.11	0.24	0.23	0.14	0.44
Cladding Condition	Sound	Sound	Sound	Sound	Sound	Sound

(a) Estimated from irradiation temperatures.

(b) Estimated from cladding diameter changes, assuming initial cladding-fuel contact.

(c) Measured from neutron radiographs.

FIGURE A-28. VOLUME CHANGE OF (U, Pu)N AS A FUNCTION OF BURNUP

FIGURE A-29. DIAMETER INCREASE OF (U, Pu)N AS A FUNCTION OF BURNUP

500X

Etched

PL4039

a. Preirradiation

500X

Etched

HC22714

b. Postirradiation

FIGURE A-30. MICROSTRUCTURES OF SPECIMEN B-22 FROM CAPSULE BMI-50-7 BEFORE AND AFTER IRRADIATION

Note the absence of fission-gas porosity in the irradiated structure.

An apparent second-phase material was noted in the fuel of several specimens from Capsule BMI-50-5, Figure A-31. This structure was only observed after etching. X-ray diffraction analysis of a sample with a large quantity (about 20 v/o) of this apparent phase yielded only characteristic (U,Pu)N peaks and a lattice parameter of 4.89 Å. The lattice parameter of unirradiated UN-20 w/o PuN is 4.90 Å. Thus, the phase is essentially (U,Pu)N in structure. It appears likely that the structure observed metallographically is a solid solution of (U,Pu)N with oxygen [(U,Pu)ON] or carbon [(U,Pu)CN] with somewhat different etching characteristics than the (U,Pu)N. Uranium-plutonium carbonitrides are known to exist, while uranium-plutonium oxynitrides would be analogous to uranium-plutonium oxycarbides.

To check the possibility of uranium-plutonium segregation during irradiation, the radial distribution of uranium and plutonium in two specimens, A-22 and A-24 (irradiated to burnups of about 29×10^{20} fissions per cm^3 at linear heat ratings of about 26 kw per ft), was determined by electron-microprobe analysis. The results for both specimens are typified by the X-ray intensity data for Specimen A-24 shown in Figure A-32. Note that no attempt was made to standardize the X-ray data to obtain absolute quantitative data. These data indicate that the distribution of uranium was fairly uniform across the radius while the plutonium concentration was low near the fuel perimeter and increased upon approaching the fuel center. The relative depletion of plutonium near the fuel surface is expected and is the result of the higher plutonium burnup near the fuel surface as compared to the fuel center because of the self-shielding of thermal neutrons by the fuel. Alpha and beta-gamma autoradiographs also indicated high burnup and plutonium depletion toward the fuel surface. On the basis of these results, it is concluded that no segregation of uranium and plutonium occurred during irradiation.

The claddings from several specimens have been subjected to additional study to check the possibilities of degradation by reaction with fuel or fission products or embrittlement by pickup of nitrogen from the fuel. As reported in BMI-1868, the irradiated Type 304 stainless steel claddings had sensitized microstructures characterized by carbide precipitates at grain boundaries and along deformation bands. In addition, several specimens had isolated regions near the cladding inner diameter that were susceptible to particularly heavy etched attack, Figure A-33. To check the possibility of fission-product or fuel reaction in these areas, samples of cladding from helium-bonded Specimen A-2 (18.2×10^{20} fissions per cm^3 at 25.4 kw per ft) and sodium-bonded Specimen B-3 (20.4×10^{20} fissions per cm^3 at 28.1 kw per ft) from Capsule BMI-50-4 were subjected to electron-microprobe analysis for cesium, cerium, zirconium, molybdenum, uranium, and plutonium in addition to the stainless steel constituents, iron, chromium, and nickel. The analyses revealed that no fission products, uranium, or plutonium were present. A slightly greater than normal concentration of chromium, probably in the form of iron-chromium carbide, was found in the grain boundaries.

A micro-Kjeldahl analysis of cladding from Specimen A-22 (Capsule BMI-50-7) showed the concentration of nitrogen in the cladding to be 360 ± 30 ppm, as opposed to 330 ± 30 ppm in an unirradiated control sample. Thus, no significant nitriding of the cladding occurred during irradiation.

On the basis of these results, it was concluded that the anomalous cladding microstructure was due to gross carbide precipitation at the grain boundaries, with the probability that the carbon was introduced as a surface contaminant during fabrication.

In Capsules BMI-50-6 and BMI-50-11 the irradiation conditions were more severe in terms of burnup and heat generation rate, respectively. Fuel specimens in Capsule BMI-50-6 were irradiated to burnups of 35.0×10^{20} to 48.6×10^{20} fissions per cm^3 (102,000 to 157,000 MWD/T). Heat generation rates of 29.5 to 39.7 kw per ft were attained in this capsule late in the irradiation.

Capsule BMI-50-11 was designed for the irradiation of specimens at high heat generation rates, nominally 40 kw per ft. Fuel specimens were irradiated to burnups

500X

FIGURE A-31. SECOND-PHASE MATERIAL FOUND IN THE FUEL OF THREE OUT OF FOUR SPECIMENS EXAMINED FROM CAPSULE BMI-50-5

Specimen A-6 is shown here.

FIGURE A-32. X-RAY INTENSITIES OF URANIUM AND PLUTONIUM AND URANIUM:PLUTONIUM RATIOS ACROSS THE RADIUS OF IRRADIATED (U, Pu)N SPECIMEN A-24 FROM CAPSULE BMI-50-7

of 15.9×10^{20} to 20.1×10^{20} fissions per cm^3 (52,500 to 58,500 MWD/T) at average heat generation rates of 41.6 to 49.0 kw per ft, with peak rates exceeding 50 kw per ft. Under these conditions, fuel-surface temperatures up to 1105 C and centerline temperatures in excess of 1600 C were realized.

500X

HC21909

FIGURE A-33. TYPE 304 STAINLESS STEEL CLADDING SODIUM BONDED TO HIGH-DENSITY (U, Pu)N SPECIMEN B-3 OF CAPSULE BMI-50-4

Note the fine grains near the ID. Electron-beam microprobe analysis indicated that heavily etched grain boundaries are rich in chromium, probably in the form of chromium carbide.

The specimen descriptions, irradiation conditions, and results of preliminary postirradiation examinations of Capsules BMI-50-6 and BMI-50-11 are summarized in Tables A-19 and A-20. Macroviews of the irradiated specimens from these two capsules are shown in Figures A-34 and A-35.

In Capsule BMI-50-6 (high-burnup experiment) both of the helium-bonded specimens experienced cladding strains of more than 4 percent. Expectedly, both claddings were ruptured. Specimen B-1 (10-mil sodium bond) also had a ruptured cladding; in the region of the rupture, the cladding had deformed 1.9 percent. Average cladding strain over the fueled region of this specimen was 0.6 percent. The other three specimens in the capsule were apparently sound and had experienced little or no cladding strain. Except for the helium-bonded specimens, no fuel-pellet swelling data are presently available for these specimens. On the basis of cladding diameter increases, fuel swelling in the helium-bonded specimens was on the same order as was exhibited by fuel from earlier capsule experiments. The cause of the cladding deformities and rupture in 10-mil sodium-bonded Specimen B-1 is uncertain and will be the subject of further investigations.

All the fuel specimens from Capsule BMI-50-11 (high heat-generation-rate experiment) exhibited some cladding deformation. However, only Specimen B-54 (10-mil sodium bond) had a visible cladding defect. The presence of fission gas in all the capsule compartments suggests that microdefects exist in the claddings of a number of these

TABLE A-19. IRRADIATION CONDITIONS AND PRELIMINARY SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-6 (IRRADIATION CONDITIONS BASED ON THERMOCOUPLE DATA)

	Specimen Designation					
	A-4	A-3	B-2	B-1	C-2	C-1
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	83.8	95.5	83.9	95.5	96.5	94.4
Smear	83.8(a)	95.5(a)	73.3	83.0	70.0	68.0
Burnup						
10^{20} Fissions per Cm^3	39.7	47.7	48.6	43.1	46.3	35.0
10^3 MWD/T(b)	130	132	157	123	131	102
Fuel-Surface Temperature(c), C						
Average	615	685	685	615	635	545
Maximum	725	800	800	720	750	645
Maximum Temperature at Fuel Center(c), C	1130	1205	1260	1070	1115	950
Linear Heat Generation, kw per ft						
Average(d)	22.6	26.2	26.5	23.4	25.0	19.5
Maximum	34.8	39.7	39.7	34.1	36.2	29.5
Cladding Diameter Increase, percent						
Maximum	5.8	5.2	0.3	1.9	0	0
Average	4.1	4.4	0.15	0.6	0	0
Fuel Diameter Increase, percent						
Maximum	6.6(e)	5.9(e)	(f)	(f)	(f)	(f)
Average	4.7(e)	5.0(e)	(f)	(f)	(f)	(f)
Cladding Condition	Failed	Failed	Sound	Failed	Sound	Sound

(a) Slip fit of fuel into cladding - essentially no clearance.

(b) Based on 200 Mev per fission.

(c) Capsule was relocated into higher flux positions for last six irradiation cycles. Irradiation temperatures are for these cycles.

(d) Time-weighted average for entire irradiation.

(e) Based on cladding deformation and assuming no clearance between fuel and cladding.

(f) Data not yet available.

TABLE A-20. IRRADIATION CONDITIONS AND PRELIMINARY SWELLING DATA FOR FUEL SPECIMENS IN CAPSULE BMI-50-11 (IRRADIATION CONDITIONS BASED ON THERMOCOUPLE DATA)

	Specimen Designation					
	A-43	A-44	B-43	B-54	C-43	C-44
Bond						
Type	Helium	Helium	Sodium	Sodium	Sodium	Sodium
Cold Gap, mils	<1	<1	10	10	20	20
Density, percent of theoretical						
Pellet	82.7	94.7	82.7	94.4	94.3	94.3
Smear	82.7(a)	94.7(a)	70.2	80.6	69.0	68.8
Burnup						
10^{20} Fissions per Cm^3	17.0	18.2	15.9	19.2	20.1	18.9
10^3 MWD/T(b)	55.7	52.5	58.5	55.0	57.5	54.5
Average Irradiation Temperature(c), C						
Fuel Surface	830	880	870	920	960	910
Fuel Center	1240	1310	1295	1375	1440	1360
Average Linear Heat Generation(c), kw per ft	41.6	44.5	44.0	46.5	49.0	46.4
Cladding Diameter Increase, percent						
Maximum	1.4	2.2	0.5	2.8	2.0	0.8
Average	0.8	1.7	0.2	1.2	1.0	0.2
Fuel Diameter Increase, percent						
Maximum	1.6(d)	2.4(d)	(e)	(e)	(e)	(e)
Average	0.9(d)	1.8(d)	(e)	(e)	(e)	(e)
Cladding Condition(f)	Sound	Sound	Sound	Failed	Sound	Sound

(a) Slip fit of fuel into cladding - essentially no clearance.

(b) Based on 200 Mev per fission.

(c) Time-weighted averages.

(d) Based on cladding deformation and assuming no clearance between fuel and cladding.

(e) Data not yet available.

(f) Based on macroappearance. The presence of fission gas in all the capsule compartments suggests that pinhole leaks exist in several of the specimens.

4X 2178-9
Specimen A-4
(Helium Bond)

4X 2180-1
Specimen A-3
(Helium Bond)

4X 2184-5
Specimen B-2
(10-Mil Sodium Bond)

FIGURE A-34. IRRADIATED SPECIMENS FROM CAPSULE BMI-50-6

Note that the claddings are ruptured on both helium-bonded specimens and on 10-mil sodium-bonded Specimen B-1.

4X

2186-7

Specimen B-1
(10-Mil Sodium Bond)

4X

2190-1

Specimen C-2
(20-Mil Sodium Bond)

4X

2188-9

Specimen C-1
(20-Mil Sodium Bond)

FIGURE A-34. (CONTINUED)

4X

2168-9

Specimen A-43
(Helium Bond)

4X

2166-7

Specimen A-44
(Helium Bond)

4X

2170-1

Specimen B-43
(10-Mil Sodium Bond)

FIGURE A-35. IRRADIATED SPECIMENS FROM CAPSULE BMI-50-11

Note that the only specimen with a visibly failed cladding is 10-mil sodium-bonded Specimen B-54.

4X

2172-3

Specimen B-54
(10-Mil Sodium Bond)

4X

2163-4

Specimen C-43
(20-Mil Sodium Bond)

4X

2162-2

Specimen C-44
(20-Mil Sodium Bond)

FIGURE A-35. (CONTINUED)

specimens. However, the failure of the cladding on Specimen B-54 is unusual in that it is oriented circumferentially and apparently is not related to hoop stresses in the cladding. This specimen is bowed, suggesting that it has been subjected to a bending moment. The cause of this failure will be sought in future detailed examinations.

It is interesting to note that the diametral swelling rates of the helium-bonded specimens from Capsule BMI-50-11 are approximately the same as were observed for helium-bonded specimens irradiated at lower heat generation rates. It is also noteworthy that the helium-bonded specimens experienced relatively large cladding deformations without rupturing their cladding.

Plans for Future Work

The postirradiation examination of fuel specimens from Capsules BMI-50-6 and BMI-50-11 will continue, with emphasis on determining the cause(s) of the two observed cladding failures in sodium-bonded specimens. Additionally, more detailed examinations of the sound specimens will be performed to establish the effects of high burnup and high heat generation rate on fuel swelling, fission-gas release, and fuel-cladding compatibility.

Preparation of EBR-II Fuel Pins

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Objective of Program

To fabricate solid-solution ($U_{0.8}Pu_{0.2}$)N pellets by powder metallurgical techniques and clad them in well characterized Type 316 stainless steel for irradiation in the EBR-II.

Approach Being Used

Sintering techniques are being employed to fabricate ($U_{0.8}Pu_{0.2}$)N pellets of controlled density, size, and microstructure for EBR-II fuel-pin irradiation experiments. Pellets for the BMI-3 series capsules were prepared in Fiscal Year 1970. Design variables included fuel density (88.5 and 95 percent of theoretical), bonding medium (sodium or helium), and total diametral fuel-cladding gap (0.0025 to 0.0100 in.). Each fuel pin contains 10 in. of fuel.

In the general procedure to be used in preparation of EBR-II fuel pins, nominal ($U_{0.8}Pu_{0.2}$)N powder containing fully enriched uranium will be ball milled to reduce the particle size, cold pressed in hardened steel dies, sintered, and centerless ground, if necessary, to produce fuel pellets that are within ± 2.0 percent of the design densities and 0.0005 in. of the required diameters. The fuel pellets will be right circular cylinders having a length-to-diameter ratio of approximately one. Pellets for two of the BMI-3 series capsule loadings have a nominal 0.070-in. -diameter annular hole. Control pellets from each powder batch will be characterized with respect to chemistry, microstructure, density, and impurity levels.

Fuel pins consisting of (U,Pu)N pellets either helium or sodium bonded to tubular cladding will be assembled in a high-purity helium or argon atmosphere [to permit post-irradiation measurement of helium generated by the (n,α) reactions] and sealed by TIG welding. Type 316 stainless steel tubes which have been well characterized for defects and dimensional tolerances will be utilized. After plutonium decontamination, each sealed fuel pin will be helium leak checked, visually examined, and radiographed to verify the position of the fuel pellets and the overall quality of the welds. Sodium-bonded fuel pins are heated to a temperature in excess of the melting point of sodium and centrifuged; an eddy-current inspection is performed to assure a continuous, void-free sodium bond.

Significant Accomplishments During Fiscal Year 1970

The fabrication of pellets for seven fuel pins, each containing approximately 10 in. of fuel, was completed. The pellets were fabricated by cold pressing and sintering nominal $(U_{0.8}Pu_{0.2})N$ powders prepared by a hydride-nitride cycle on arc-cast uranium-20 w/o plutonium buttons.

Seven fuel pins each composed of $(U,Pu)N$ pellets stacked within a tube of Type 316 stainless steel cladding, were loaded in an atmosphere of high-purity helium. In three pins, the bonding medium between the fuel and cladding was helium while sodium bonding was utilized in the other four pins. After welding, each of the pins was tested in accordance with the program requirements and found acceptable for secondary encapsulation.

Summary of Research Carried Out

The fabrication of both high-density and low-density $(U_{0.8}Pu_{0.2})N$ pellets for fuel pins to be inserted in the EBR-II was completed. A summary of dimensional and density data is given in Table A-21 along with fuel-pin design data.

TABLE A-21. DATA FOR $(U_{0.8}Pu_{0.2})N$ FUEL PINS IN THE BMI-3 SERIES SCHEDULED FOR EBR-II IRRADIATION

Capsule	Pellet Quantity	Total Pellet Length, in.	Average Diameter of Fuel ^(a) , in.		Average Density of Fuel ^(b) , percent of theoretical	Fuel-Pin Design Data	
			ID	OD		Bonding Medium	Radial Gap Width, mil
BMI-3-2	44	9.8592	--	0.2745	88.4	Sodium	5.0 ^{+0.0} -1.0
BMI-3-3	42	10.0623	--	0.2744	91.3	Sodium	5.0 ^{+0.0} -1.0
BMI-3-4	50	9.9935	--	0.2707	93.9	Sodium	7.0 ^{+0.0} -1.0
BMI-3-5	41	10.0302	--	0.2848	90.4	Sodium	5.0 ^{+0.0} -1.0
BMI-3-6	48	10.1077	0.069 ^(c)	0.2701	94.7	Helium	2.5 ^{+0.0} -1.0
BMI-3-7	35	9.6670	--	0.2698	88.5	Helium	2.5 ^{+0.0} -1.0
BMI-3-8	46	9.9254	0.069 ^(c)	0.2699	89.8	Helium	2.5 ^{+0.0} -1.0

(a) ID given for annular pellets.

(b) Based on a theoretical density of 14.17 g per cm³.

(c) Except for $U_{natural}N$ end pellets, which will be solid and 88.5 percent of theoretical density.

Nominal $(U_{0.8}Pu_{0.2})N$ powders containing fully enriched uranium were prepared using a hydride-nitride cycle on arc-cast uranium-20 w/o plutonium buttons. Seven powder batches (A-30, A-32, A-33, A-34, A-35, A-36, and A-37) were synthesized to fabricate the seven pins. The as-synthesized powder was wet ball milled for 32 hr in a uranium-lined ball mill containing tungsten carbide-cobalt balls and nanograde hexane. After milling, the powder was vacuum dried at room temperature, and then pressed into pellet form in hardened steel dies using zinc stearate as a die lubricant. High-density pellets were sintered at 2100 C for approximately 10 hr under 1 atm of nitrogen while pellets for low-density pins were sintered at 1975 C for 6 hr under 1 atm of nitrogen. Annular pellets were prepared for Capsules BMI-3-6 and BMI-3-8.

Table A-22 shows the powder batch associated with its fuel pin along with the corresponding die diameter and the pressure used in cold pressing. All pellets were centerless ground to meet the dimensional requirements.

TABLE A-22. FABRICATION DATA FOR BMI-3-SERIES FUEL PELLETS

Capsule	Powder Batch	Die Diameter, in.	Pressing Pressure, tsi
BMI-3-2	A-36	0.344	15
BMI-3-3	A-35	0.344	15
BMI-3-4	A-30	0.338	15
BMI-3-5	A-35 and A-37	0.344	15
BMI-3-6	A-32 and A-33	0.338 ^(a)	15
BMI-3-7	A-34	0.338	15
BMI-3-8	A-32 and A-33	0.338 ^(a)	15

(a) Die contained a 0.088-in. -diameter center mandrel.

Characterization of the densified ($U_{0.8}Pu_{0.2}$)N EBR-II pellets involved density measurements, metallographic examination, and chemical analyses. Although chemical analyses were not conducted on pellets from every powder batch, control pellets were placed in storage for future analyses if necessary. Selected pellets from each batch with the exception of Batch A-37 were examined metallographically. With the exception of Batch A-36 all microstructures were single-phased. The microstructure of the pellet from Batch A-36 showed a second phase believed to be higher nitride. Typical microstructures for high-density and low-density EBR-II pellets are shown in Figure A-36, while typical chemical analyses are given in Table A-23.

TABLE A-23. TYPICAL CHEMICAL ANALYSES ON ($U_{0.8}Pu_{0.2}$)N PELLETS

Pellet Identification	Carbon, w/o	Nitrogen, w/o	Uranium, w/o	Plutonium, w/o
A-33-70	0.05	5.48	--	--
A-34-16	0.03	5.49	--	--
A-30-28	--	--	75.6	19.1
A-32-1	--	--	76.6	18.8

Seven fuel pins, each composed of (U,Pu)N pellets stacked within a tube of Type 316 stainless steel cladding, were loaded in an atmosphere of high-purity helium. The bonding medium between the fuel and cladding was helium for three of the pins, while sodium bonding was utilized in the other four pins. The cladding OD of all pins in this irradiation series (BMI-3) was held constant at 0.315 in. The cladding wall thickness and the sodium annulus were varied as part of the irradiation study. The end plugs were welded in. Only seven usable pins were generated because of welding problems associated with the eighth pin. Each of the pins was tested in accordance with the program requirements and found acceptable for secondary encapsulation.

500X

Etched

PL7921

a. Low-Density Pellet A-33-22

500X

Etched

PL7929

b. High-Density Pellet A-33-66

FIGURE A-36. TYPICAL MICROSTRUCTURES OF LOW - AND HIGH-DENSITY EBR-II PELLETS

Plans for Future Work

Pellets will be fabricated for a 19-pin fuel bundle and the pins loaded in Fiscal Year 1971.

EBR-II Irradiation-Capsule Assembly and Safety Analysis Preparation

E. O. Fromm, N. E. Miller, and J. W. Ray

Objective of Program

To design and fabricate irradiation capsules, encapsulate clad (U,Pu)N fuel pins, perform required nondestructive examinations, prepare the Data Package for the experiments, ship the completed capsules to the EBR-II, and perform irradiation surveillance for experiments in the EBR-II.

Approach Being Used

Standard EBR-II irradiation capsules are being used for this program. Both the capsule components and some cladding material which are of Type 304L stainless steel will be procured from ANL. Additional cladding material of Type 316L stainless steel will be procured commercially. The fuel-pin cladding will be sodium bonded to the outer capsule by centrifuging at 1000 F. Sodium levels and bond soundness will be verified by X-radiography and eddy-current testing.

Significant Accomplishments During Fiscal Year 1970

Seven EBR-II capsules designated BMI-2-1, -2-2, -2-3, -2-5, -2-6, -2-7, and -2-8, designed to operate at fuel-pin power ratings up to 40 kw per ft were completed and sent to the EBR-II for irradiation. The Quality Assurance Description document associated with these experiments was prepared and forwarded to the EBR-II site for review. The eddy-current equipment used for determination of bond soundness was modified to improve the stability; the sensitivity of the modified equipment was determined by the use of dummy capsules fabricated from material having properties which closely approximated the electrical properties of the components in the actual irradiation capsules.

Three EBR-II capsules in the third series (C-BMI-3-6, -3-7, and -3-8) designed to operate at fuel-pin power ratings of 30 to 40 kw per ft were assembled and nondestructively tested. The Experiment Description and Safety Analysis documents associated with the BMI-3 series experiments were prepared. The Quality Assurance Description document for the three completed capsules was also prepared.

Capsule BMI-1-3 was inserted in the EBR-II in Subassembly X-070 for Run 39B.

Summary of Research Carried Out

Eight EBR-II capsules containing (U,Pu)N fuel pins in pellet form were originally included in the BMI-2 series irradiation experiments. However, one capsule (BMI-2-4) did not meet the BCL quality assurance and was not included in the BMI-2 experiments. Therefore, there are only seven capsules instead of the eight originally planned. The design details and desired irradiation conditions for the seven capsules, designated BMI-2-1, -2-2, -2-3, -2-5, -2-6, -2-7, and -2-8, are given in Table A-24. All of the capsules contain a nominal 10-in. active fuel region within claddings of Type 316L stainless steel with either 0.010-, 0.015-, or 0.020-in.-thick walls. Argon is used as the cover gas in the inner clad assembly so that the amounts of helium and hydrogen generated by the (n, α) and (n,p) reactions and their release from the fuel can be determined after the irradiation. The clad elements are immersed in sodium within an outer capsule of Type 304L stainless steel which is 0.375 in. in OD with a 0.020-in. wall. The fuel cladding and end-cap materials were obtained commercially, whereas the outer capsules and end fittings were obtained from ANL and are standard EBR-II components.

TABLE A-24. DESCRIPTION OF THE BMI-2 SERIES OF $(U_{0.8}Pu_{0.2})N$ IRRADIATIONS IN THE EBR-II

Capsule	Fuel-Pin Cladding Thickness ^(a) , in.	Fuel-Cladding Radial Sodium Bond Thickness, in.	Fuel Diameter ^(b) , in.	Fuel Smear Density ^(c) , percent	Specific Power ^(d) , kw per ft	Target Burnup ^(e) , a/o
BMI-2-1	0.020	0.010	0.255	81.7	30.7-33.8	6
BMI-2-2	0.020	0.010	0.255	81.7	30.7-33.8	9
BMI-2-3	0.020	0.010	0.255	81.7	30.7-33.8	12
BMI-2-5	0.015	0.015	0.255	76.1	30.7-33.8	12
BMI-2-6	0.010	0.010	0.275	82.6	35.7-39.3	6
BMI-2-7	0.010	0.010	0.275	82.6	35.7-39.3	12
BMI-2-8	0.010	0.010	0.275	82.6	35.7-39.3	9

(a) All cladding is Type 316L stainless steel, 0.315 in. in OD.

(b) All capsules will contain a nominal 10-in. length of fuel axially centered within the EBR-II core.

(c) Based on a fuel-pellet density of 95 percent of theoretical; the theoretical density of $(U_{0.8}Pu_{0.2})N$ is 14.3 g per cm³.

(d) Range of specific powers in core Row 4 of the EBR-II operating at 50 Mw; for the specified fuel this corresponds to 9.54×10^{13} to 10.50×10^{13} fissions/(cm³) (sec).

(e) For $(U_{0.8}Pu_{0.2})N$ of 95 percent of theoretical density, a heavy atom burnup of 1 percent equals 3.27×10^{20} fissions per cm³.

Sodium bonding of the inner clad assemblies for the BMI-2 series was accomplished by centrifuging, and verification of the soundness of the bond was made by eddy-current techniques.

The seven sodium-bonded fuel pins were loaded into the outer capsules using a procedure similar to that used for the BMI-1-series capsules. Essentially, the encapsulation is done inside a dry box previously evacuated and backfilled with purified helium. The clad assembly is preferentially heated from the top down to melt the sodium so as not to disturb the sodium bond or strain the cladding. The hot assembly is then lowered into the capsule which contains approximately 5 g of molten sodium. The sodium in both annuli is cooled from the bottom up, the top end cap is Heliarc welded to the capsule, and the welds are nondestructively tested for soundness and leak tightness.

The sodium bond between the cladding and the outer capsule was also accomplished by centrifuging, and verification of the soundness of the bonds was made by eddy-current techniques.

The eddy-current equipment used to evaluate the integrity of the sodium bonds of both the inner and outer sodium annuli was modified to improve stability. As before, the sensitivity of the modified equipment was evaluated by means of dummy capsules fabricated from material having properties which closely approximated the electrical properties of the components of the actual clad fuel pins and irradiation capsules. Various known void configurations were simulated as a basis for the determination of the eddy-current equipment sensitivity. For the inner clad assembly it was possible to mask out the effects of pellet interfaces and offset pellets and still detect a void as small as 1/16 in. in diameter through the 0.010-in. -thick annulus. The minimum detectable void in the outer sodium annulus was of comparable size. No voids of this magnitude were detected in either annulus of the seven capsules in the BMI-2 series.

Work was initiated on the third set of EBR-II irradiation experiments, designated the BMI-3 series. The planned fabrication of eight capsules was reduced to seven when

welding difficulties were encountered late in the year with fuel-pin fabrication. The seven capsules in this group of (U, Pu)N fuel pins emphasize specific fuel-pin power ratings in the range of 30 to 40 kw per ft and burnups to 100,000 MWD/T. The design details for each capsule are summarized in Table A-25. Essentially, each capsule contains a nominal 10-in. active fuel length of (U, Pu)N fuel pellets within a 0.315-in. -OD Type 316 stainless steel cladding obtained from a commercial vendor. Four of the capsules contain a 0.005- or a 0.007-in. -thick radial sodium bond between fuel and cladding, while the other three have a 0.0025-in. -thick radial helium bond between the fuel and cladding. The fuel-pin dimensions and target burnups were established on the basis of the fuel swelling observed in the thermal flux irradiation of mixed-nitride fuels conducted by Battelle-Columbus. Thus, a particular objective of these experiments is to provide a quantitative comparison between the solid-fission-product swelling rates occurring during fast-flux irradiation with those observed in a predominately thermal-flux environment. Additionally, an optimum sodium-gap size is being sought. The helium-bonded pins were designed to compare the effectiveness of centralized and distributed porosity in accommodating fuel swelling and to observe the effect of cladding restraint on fuel swelling.

The Approval in Principle for BMI-3 series was prepared and forwarded to the AEC for review; approval was granted for the proposed irradiations. The Experiment Description and Hazards Analysis was prepared and is being reviewed prior to forwarding to the EBR-II Project.

Characterization and fabrication of the outer capsule components for all seven capsules was completed. Assembly of the first three capsules (C-BMI-3-6, -3-7, and -3-8) was completed. Essentially, the procedure for encapsulation into the outer capsule was identical to that used for the BMI-2 series. The sodium bond between the cladding and outer capsule was accomplished by centrifuging, and verification of the soundness of the bond was made by eddy-current techniques. No voids 1/16 in. in diameter or larger through the 0.010-in. -thick sodium annulus were detected. The Quality Assurance Description for the three capsules was prepared. Assembly of the remaining four capsules was begun.

Capsule BMI-1-3 was loaded into Position 12 of MK-A-19 Subassembly X-070, and the subassembly was inserted into Position 3E1 of the reactor grid for EBR-II Run 39B. This is the first mixed-nitride capsule to be irradiated in the EBR-II. The target burnup for the capsule (helium bond) is 3 a/o. At the end of EBR-II Run 42, the capsule had accumulated a total of 4435 reactor megawatt-days for a total burnup of about 2.4 a/o at an estimated maximum specific fuel-pin power of 25.8 kw per ft [9.1×10^{13} fissions/(cm³)(sec)].

Plans for Future Work

The irradiation of Capsules BMI-1-1, -1-2, -1-4, -2-1, -2-2, -2-3, -2-5, -2-6, -2-7, and -2-8 will be initiated. It is anticipated that these capsules will be incorporated into Subassembly X-086 and inserted in the EBR-II for Run 45.

The irradiation of Capsule BMI-1-3 will be terminated at the end of EBR-II Run 44; a peak fuel burnup of about 3.8 a/o is anticipated. The capsule will be returned to Battelle-Columbus in Cask BCL-4 for postirradiation examination after an appropriate cooling period.

The assembly of the four remaining capsules in the BMI-3 series will be completed, and the capsules will be nondestructively tested. All seven capsules will then be shipped to the EBR-II Irradiation Coordinator. The Quality Assurance Description for the four capsules with sodium-bonded fuel pins will be prepared. The Data Package containing the Experiment Description, Hazards Analysis, and the Quality Assurance Description (for all seven capsules) will be forwarded to the EBR-II Project for review.

TABLE A-25. BMI-3 SERIES OF (U,Pu)N FUEL-PIN IRRADIATIONS IN THE EBR-II

Capsule	Fuel-Pin Cladding ^(a) , in.		Bond Medium	Radial Bond Thickness, in.	Fuel Diameter ^(b) , in.	Fuel Density ^(c) , percent of theoretical	Specific Power ^(d) , kw per ft	Smear Density, percent of theoretical	Burnup ^(e) , a/o		Planned Burnup ^(f) , a/o	Reactor Megawatt-Days to Achieve Planned Burnup ^(g)
	ID	Wall							To Contact Cladding	To Yield 1 Percent Cladding Strain		
BMI-3-2	0.285	0.015	Sodium	0.005	0.275	88	31.0-38.9	81.8	7.6	10.2	7.5	19,687.5
BMI-3-3	0.285	0.015	Sodium	0.005	0.275	91	32.1-40.2	84.6	7.6	10.2	9	26,250
BMI-3-4	0.285	0.015	Sodium	0.007	0.271	95	32.4-40.8	85.9	11.0	13.8	12	26,250
BMI-3-5	0.295	0.010	Sodium	0.005	0.285	91	34.4-43.2	85.0	7.4	9.8	7.5	13,125
BMI-3-6	0.275	0.020	Helium	0.0025	0.270 (0.070)	95	30.0-37.8	85.4	3.8	--	6	13,125
BMI-3-7	0.275	0.020	Helium	0.0025	0.270	88.5	30.0-37.7	85.3	4.1	--	6	13,125
BMI-3-8	0.275	0.020	Helium	0.0025	0.270 (0.070)	88.5	28.0-35.2	79.6	4.1	--	6	13,125

(a) All cladding is Type 316 stainless steel, 0.315 in. in OD.

(b) Values in parentheses are the ID's of annual pellets.

(c) Based on a theoretical density of 14.3 g per cm³ for (U_{0.8}Pu_{0.2})N.

(d) Ranges of axial centerline specific power levels in Core Row 5 of EBR-II operating at 62.5 Mw.

(e) Based on a fuel swelling rate of 0.48% $\Delta V/V$ per 10²⁰ fissions per cm³ (1.5% $\Delta V/V$ per a/o burnup), and assuming that internal porosity is ineffective in accommodating fuel swelling. Cladding swelling due to fast neutron irradiation is assumed to be given by $\Delta V/V = 10^{-38} (\phi t)^{1.69} \left[\exp\left(-\frac{7808}{RT}\right) - \exp\left(-\frac{25300}{RT}\right) \right]$.

(f) For 95 percent dense (U_{0.8}Pu_{0.2})N, 1 a/o burnup equals 3.27 x 10²⁰ fissions per cm³; for 88.5 percent dense (U_{0.8}Pu_{0.2})N, 1 a/o burnup equals 3.05 x 10²⁰ fissions per cm³.

(g) Assuming irradiation in an average Core Row 5 location.

A 19-pin subassembly containing unencapsulated, Type 316 stainless steel-clad mixed-nitride fuel pins will be designed and the pins fabricated and sent to the EBR-II for irradiation. The pins will be designed for irradiation at 30 kw per ft to a burnup goal of 150,000 MWD/T. The tentative fuel and cladding parameters are as follows:

(U,Pu)N pellet diameter	0.260 in.
Fuel density	95 percent of theoretical
Sodium annulus	10-mil radial
Cladding material	Type 316 stainless steel
Cladding OD	0.310 in.
Cladding thickness	15 mil

The Approval in Principle will be prepared and forwarded to the AEC for review. The Data Package consisting of the Experiment Description, Safety Analysis, and Quality Assurance Description will be prepared and forwarded to the EBR-II Project for review.

The irradiation surveillance and performance analysis for all capsules in the EBR-II will be continued.

UN-PuN TREAT Analysis

R. Artigas, H. D. Gronbeck, and P. Cybulskis

Objective of Program

To perform transient irradiation tests on (U,Pu)N fuels in the TREAT reactor.

Approach Being Used

(U,Pu)N fuel specimens will be subjected to transients in the TREAT reactor in order to simulate the dynamic thermal behavior of fast-reactor fuels. These tests should help to establish the acceptability of mixed-nitride fuels for LMFBR application under operational transient and accident conditions.

Significant Accomplishments During Fiscal Year 1970

In order to accomplish the objectives of the program a tentative capsule design has been adopted. The adequacy of the conceptual capsule design from a heat-transfer viewpoint has been demonstrated by means of time-dependent thermal calculations for the (U,Pu)N-fueled experiments for a range of reactor transients. The correlation between neutron flux in the (U,Pu)N-fueled capsule and the TREAT reactor has been established through the computation of a figure of merit for this experiment.

Summary of Research Carried Out

Transient testing programs for other ceramic fuels, performed by General Electric, Atomics International, and Argonne National Laboratory in the TREAT facility, were reviewed in order to take full advantage of the background of information developed previously. The physical properties of the mixed nitrides were compared with those of the carbides and oxides in order to predict the relative behavior of the fuels under transient conditions. Parameters which must be varied in the program have been identified as: type of bond between fuel and cladding, transient magnitude, preirradiation history, fuel density, and transient time scale.

Analytical comparisons were made between the thermal response of nitride fuels during TREAT transients and the response of a nitride-fueled LMFBR to an accident

reactivity excursion. It was found that TREAT can provide an adequate simulation of the thermal transients that may be expected under LMFBR accident conditions.

Analytical and experimental questions concerning the TREAT tests were discussed with representatives of ANL, and energy releases of up to 80 Btu per in. of fuel were tentatively selected as the basis for the TREAT capsule design. The primary cause of cladding failure for the unirradiated specimens in the temperature range of interest (2500 to 5000 F) is expected to be the pressure due to the vaporization of the sodium bond during the transient, and not the vapor or decomposition pressure of the fuel.

In order to establish the pulse characteristics that would subject the (U,Pu)N fuels to temperatures in the range of interest (2500 to 5000 F) for significant periods of time and to develop the requirements for the detailed capsule design, a preliminary conceptual capsule design was adopted. A one-dimensional geometrical model suited for computational purposes and based on the conceptual design is illustrated in Figure A-37. The time-temperature behavior of the mixed-nitride-containing capsule when subjected to a flat-top transient was calculated by means of the ARGUS⁽¹⁾ code*.

Initially, the sodium-bond region in the capsule was treated as a stationary region with thermal conductivity k , but this resulted in the selection of prohibitively small time steps to assure numerical stability, especially at times near or during sodium-bond vaporization. The treatment of sodium-bond regions as flow regions with zero velocity and the use of three axial nodes permitted the use of somewhat larger computational time steps. The heat-transfer coefficient, h , for these sodium annuli was defined such that,

$$\bar{h} = \frac{\int \frac{1}{r} \left[\frac{\partial}{\partial r} \left(k_r \frac{\partial T}{\partial r} \right) \right] r dr}{\left(\frac{\Delta T}{\Delta r} \right) \int r dr},$$

where k , the thermal conductivity, was taken to be constant over r , and the very thin annular regions were approximated by slabs. This model was tested by making two ARGUS runs, one with an "exact" treatment and another with the above definition of h . The temperature distributions as a function of time for the two cases were essentially identical.

The thermal properties of sodium in the range of 400 to 1700 F were fitted to the following parabolic equations by means of a linear regression subroutine for use in the ARGUS code:

$$\text{Thermal conductivity} = k = 54.274258 - (0.0213924)T + (4.787088 \times 10^{-6})T^2,$$

$$\text{Density (liquid)} = \rho_L = 58.727 - (0.00756757)T,$$

$$\text{Volumetric specific heat} = \rho C_p = 20.139277 - (6.556376845 \times 10^{-3})T + (1.812281168 \times 10^{-6})T^2,$$

where T is the Fahrenheit temperature, k is given in Btu/(hr)(ft)(F), ρ in lb per ft³, and ρC_p in Btu/(ft³)(F).

It is expected that the helium overpressure, which is for all practical purposes the dominant pressure at any point in the sodium bond, will not exceed 2 atm before boiling can occur. The vapor pressure of sodium exceeds 2 atm at about 1800 F. For the present analysis it was, therefore, assumed that, if the bond temperature exceeds the temperature at which the vapor pressure of sodium is greater than 2 atm, all the liquid sodium in the bond is displaced by sodium vapor. This is a reasonable, although conservative, assumption since at 2 atm the ratio of liquid to vapor densities is approximately 1300 and only a small portion of the sodium must vaporize to fill the fuel-cladding gap.

* Extensive programming modifications to the ARGUS code were made for adaptation to Battelle's CDC-6400 computer.

Radius		Region Material	
0		UN - PuN	⌀
0.125 in.		Sodium	
0.135 in.		304 SS	
0.155 in.		Sodium	
0.245 in.		Beryllium	
0.37 in.		Sodium	
0.40 in.		Niobium	
0.50 in.		Sodium	
0.55 in.		1100 aluminum	
1.50 in.			

FIGURE A-37. ONE-DIMENSIONAL MODEL OF TREAT CAPSULE
(Not Drawn to Scale)

In an attempt to determine the performance of the capsule, several "flat-top" transients, all within the tentative design criterion for total energy release of 80 Btu per in., were investigated. In a flat-top or clipped-burst transient the power rises rapidly to a high level and is maintained at that level for a brief period of time, of the order of seconds, followed by reactor shutdown. This is in contrast to a fast burst in which the power rises exponentially to a peak value and then immediately decreases to a low level. A flat-top transient generating a specified power density in the experimental fuel can be related to the average TREAT reactor power by use of the figure of merit. For these initial calculations, the figure of merit derived by General Electric for mixed-oxide irradiations in the TREAT reactor (1.12×10^{-4} watt/cm³ per watt of reactor power) was utilized. The time-temperature behavior of a representative UN-PuN experiment is illustrated in Figures A-38 through A-41 for the inner four capsule regions for four different flat-top transients. The four transients produced pseudo-steady-state power levels in the UN-PuN fuel specimen of 75, 60, 50, and 40 kw per ft at TREAT powers of 70, 55.5, 47, and 37 Mw, respectively. Figure A-42 shows the time-temperature behavior after the end of the reactor pulse for the case involving a power level of 50 kw per ft. Figure A-43 illustrates the time-temperature behavior of the fuel hot-spot for the four pulses. Figures A-44 and A-45 show the time-temperature history of the outer capsule components (beryllium, sodium, niobium, sodium, 1000 aluminum) for two different pulses.

The results of these calculations illustrate the effect of the rate of energy release on the behavior of the experimental fuel pin. For the pulse producing 75 kw per ft, under the assumptions previously noted, sodium-bond vaporization is seen to occur 2.65 sec after the start of the transient at a total TREAT energy release of about 186 Mw-sec. For the pulse producing 60 kw per ft, bond vaporization is predicted at 6.5 sec at an energy release of about 361 Mw-sec. For the pulse producing a pseudo-steady-state power level of 50 kw per ft, sodium-bond vaporization is not predicted to occur during the approximately 10-sec duration of the pulse even though the total TREAT energy release is 432 Mw-sec. As would be expected, the lower-power transients provide more time for the redistribution of energy to the heat sinks within the capsule, thus allowing a greater total energy release without exceeding a threshold such as the vaporization of the sodium bond. When sodium-bond vaporization does occur, essentially adiabatic heatup of the fuel can follow as a result of the low thermal conductivity of the sodium vapor. However, prediction of the thermal behavior of the fuel after bond vaporization is highly uncertain because of the lack of understanding about the thresholds and mechanisms of sodium boiling, the expulsion and reentry of liquid sodium in narrow annuli, etc. The situation is further complicated by the possibility of cladding failure as the temperatures in the experiment approach the regime in which bond vaporization (boiling) may be expected. The study of the above phenomena is, of course, one of the prime objectives of the fuel-element transient testing program.

In view of the uncertainties in predicting such effects at the onset of sodium-bond vaporization and the subsequent thermal behavior of the fuel pin, the calculations discussed herein can only be used to identify the types of transients which will take the experiments into regimes of interest. Thus, consideration of the results of the above calculations indicate that TREAT transients of the flat-top or clipped-burst type should produce pseudo-steady-state power levels in sodium-bonded UN-PuN fuels of about 50 kw per ft or more to reach conditions of interest from an overpower transient or accident viewpoint.

It should be noted that the experiment power levels (kw per ft) cited in this discussion do not correspond directly to the specific powers of steady-state irradiations. In the latter, the flowing coolant is maintained at a constant temperature and, from the point of view of the experimental fuel pin, provides an infinite heat sink. In the TREAT capsule, on the other hand, there is no heat transferred external to the capsule and only a finite heat capacity is available for accommodating the transient energy release. Thus, in the

FIGURE A-38. TIME-TEMPERATURE BEHAVIOR OF THE INNER FOUR REGIONS OF A UN-PuN CAPSULE SUBJECTED TO A 3.1-SEC FLAT-TOP TRANSIENT GENERATING 75 KW PER FT IN A FUEL AT A TREAT POWER OF 70 MW

FIGURE A-39. TIME-TEMPERATURE BEHAVIOR OF THE INNER FOUR REGIONS OF A UN-PuN CAPSULE SUBJECTED TO A 6.98-SEC FLAT-TOP TRANSIENT GENERATING 60 KW PER FT IN A FUEL AT A TREAT POWER OF 55.5 MW

FIGURE A-40. TIME-TEMPERATURE BEHAVIOR OF THE INNER FOUR REGIONS OF A UN-PuN CAPSULE SUBJECTED TO A 9.2-SEC FLAT-TOP TRANSIENT GENERATING 50 KW PER FT IN A FUEL AT A TREAT POWER OF 47 MW

FIGURE A-41. TIME-TEMPERATURE BEHAVIOR OF THE INNER FOUR REGIONS OF A UN-PuN CAPSULE SUBJECTED TO A 10-SEC FLAT-TOP TRANSIENT GENERATING 40 KW PER FT IN A FUEL AT A TREAT POWER OF 37 MW

FIGURE A-42. TIME-TEMPERATURE BEHAVIOR OF THE UN-PuN CAPSULE AFTER THE END OF THE REACTOR PULSE OF 50 KW PER FT

FIGURE A-43. TIME-TEMPERATURE BEHAVIOR OF FUEL HOT SPOT FOR THE FOUR PULSES

FIGURE A-44. TIME-TEMPERATURE BEHAVIOR OF THE OUTER CAPSULE FOR A 10-SEC PULSE GENERATING A POWER LEVEL OF 40 KW PER FT IN A FUEL

FIGURE A-45. TIME-TEMPERATURE BEHAVIOR OF THE OUTER CAPSULE FOR A 9.2-SEC PULSE GENERATING A POWER LEVEL OF 50 KW PER FT IN A FUEL

latter case, operation at even very low power levels for prolonged periods of time could lead to overheating of the experiment. In this connection, thermal reactor irradiations of UN-PuN fuels have been performed at continuous power levels approaching 50 kw per ft, whereas such an experimental power level could not be extended for more than 10 sec in the TREAT capsule without the expectation of possible fuel-pin failure. The energy release prior to sodium-bond vaporization and/or fuel-pin failure in TREAT will also depend on the initial temperature of the experiment; in the present analysis a starting temperature of 400 F has been assumed.

When the thermal behavior of the outer capsule components is examined it appears that the conceptual design includes sufficient heat capacity in the outer aluminum heat sink to accommodate the transient energy releases of interest (see Figures A-44 and A-45). The maximum average temperature rise in the outer aluminum region, as defined in Figure A-37, can be found to be

$$T_{\max} - T_{\text{initial}} = \left[\int_{\Delta t} Q dt \right] \times 0.5578, \quad (\text{A-12})$$

where Q refers to the experiment power in kw per ft and Δt is the duration of the pulse in seconds. The temperatures are in degrees Fahrenheit. For a flat-top transient

$\int_{\Delta t} Q dt = Q \cdot \Delta t$. Using Equation (A-12), the maximum average temperature rise in the outer aluminum, when a power level of 60 kw per ft is used for 6.5 sec, is 217 F. At any given time the hottest spot in the aluminum could be somewhat higher than this temperature rise predicts; see, for example, Figure A-46. This is an asymptotic, adiabatic, temperature increase and the actual temperature should be somewhat less than this value.

For the calculations discussed above, the power distribution and figure of merit as determined by GE for the mixed-oxide transient irradiations were utilized. In order to determine a figure of merit for the (U,Pu)N TREAT capsule, a one-dimensional, multi-group, integral transport code, THERMOS⁽²⁾, was used. A set of thermal cross sections to be used with a two-group diffusion code, 2DB⁽³⁾, was also generated. The transport-theory code gave the detailed neutron flux distribution within the UN-PuN experiment and the diffusion code was used to normalize the THERMOS results to an average reactor power.

For the THERMOS calculations, the reactor, or that part of the reactor immediately surrounding the test cell, was taken to be "normal core fuel cells"⁽⁴⁾; that is, 89.05 percent by volume of core graphite, 2.31 percent Zircaloy-3 cladding, and 8.64 percent void. The core graphite, also described in Reference (4), is 99.9585 percent by volume carbon (with a density of 1.72 g per cm³), 4.21 x 10⁻⁴ percent boron, 2.188 x 10⁻² percent iron, 1.787 x 10⁻² percent U²³⁵, and 1.32 x 10⁻³ percent U²³⁸. The (U,Pu)N capsule was assumed to be placed in a 4 by 4-in. test cell at the center of the TREAT core. The results of the THERMOS calculations are given in Figures A-47 and A-48, and the number densities used in the calculation in terms of nuclide per unit volume are given in Table A-26. The mixed-nitride fuel was assumed to have a density of 95 percent of theoretical, and to contain 80 w/o UN and 20 w/o PuN. The uranium isotopic composition was assumed to be that of natural uranium, and the plutonium as 89.6 percent Pu²³⁹, 8.8 percent Pu²⁴⁰, and 1.6 percent Pu²⁴¹. For computational purposes the test capsule was divided into four regions: (1) fuel; (2) a volume-homogenized region of sodium, stainless steel, and sodium; (3) a volume-homogenized region of beryllium, sodium, niobium, and sodium; and (4) an aluminum outer region. Figure A-48 also compares the power distribution for the (U,Pu)N TREAT capsule with the power distribution of mixed-oxide fuel of General Electric Company. A higher peak-to-average power ratio is present in the mixed-nitride fuel. The differences are attributed primarily to differences in fuel radii and fuel composition. These differences in power distribution are expected to make little difference in the overall temperature distribution within the capsule.

FIGURE A-46. ALUMINUM HOT-SPOT TEMPERATURE AS A FUNCTION OF TIME

FIGURE A-47. SOURCE-NORMALIZED THERMAL FLUX AS A FUNCTION OF POSITION IN (U, Pu)N FUEL IN TREAT TEST CELL

FIGURE A-48. POWER DISTRIBUTION (ARBITRARY NORMALIZATION) OF (U, Pu)N FUEL AND A MIXED OXIDE AS A FUNCTION OF POSITION IN TREAT TEST CELL

TABLE A-26. NUMBER DENSITIES USED IN THE INTEGRAL TRANSPORT CALCULATION

Element	Number Density, nuclides per unit volume $\times 10^{-24}$	
	(U,Pu)N Capsule	TREAT Reactor
Carbon	--	0.076796
Boron	--	5.1144×10^{-7}
Iron	0.0843	2.0445×10^{-5}
Uranium-235	0.0001871	7.706×10^{-6}
Uranium-238	0.025798	5.622×10^{-7}
Tin	--	1.794×10^{-6}
Zirconium	--	0.0009722
Plutonium-239	0.0057945	--
Plutonium-240	0.0005691	--
Plutonium-241	0.0001035	--
Sodium	0.0254	--
Aluminum	0.0602	--
Beryllium	0.1236	--
Niobium	0.0545	--
Nitrogen	0.03245	--

From the THERMOS results, thermal-group parameters were determined for the (U,Pu)N capsule. A cell calculation was also performed for the TREAT core using the same code, and a set of thermal cross sections was determined for the reactor.

A set of fast cross sections was generated by the use of a Fourier Transform 54-group Fast Spectrum Code, FORM⁽⁵⁾. Cross sections were generated for both the TREAT core and the (U,Pu)N capsule.

The reflector compositions were taken as described in Table A-27; some of the impurities were neglected. The thermal spectrum was assumed to be Maxwellian for the computation of thermal parameters for the reflector materials. The fast cross sections were obtained from FORM.

TABLE A-27. REFLECTOR COMPOSITIONS

Material	Amount of Indicated Element Present, v/o		
	Carbon	Zirconium	Aluminum
Upper reflector	65.39	3.98	15.67
Zirconium-clad reflector	89	2.31	--
Aluminum-clad reflector	87	--	4.6
Permanent reflector	90.57	--	1.89

For computational purposes the TREAT reactor was geometrically represented in a symmetrical R-Z geometry, as shown in Figure A-49. The two-dimensional diffusion code, 2DB, was used to compute power and flux distributions. Modification was required in using this code for a thermal reactor, since it defines Σ_{tr} as $\Sigma_a + \Sigma_s$. Σ_{tr} was computed properly, with Σ_s defined as $\Sigma_s (1-\mu)$ for the present calculations. Axial and radial flux distributions were computed. The reactor core was assumed to have no control rods; control rods were included in the upper reflector, and no test hole was included. Figures A-50 and A-51 compare the fission rates as measured by the use of

FIGURE A-49. R-Z REPRESENTATION OF THE TREAT REACTOR
(NOT TO SCALE)

FIGURE A-50 COMPUTED AND MEASURED AXIAL FISSION RATES IN THE TREAT CORE

FIGURE A-51 COMPUTED AND MEASURED RADIAL FISSION RATES IN THE TREAT CORE

U^{235} foils⁽⁴⁾, and as computed by this analysis. The measured fission rate in the lower half of the reactor is higher because it contains no control rods.

An experiment was next included in the test hole with a (U, Pu)N specimen about 2.5 in. long. A combination of the THERMOS run and the normalizing 2DB run was used to determine a figure of merit, i. e., the ratio of the specific power in the experimental fuel specimen to the average reactor power. This figure of merit determines the desired TREAT reactor power for a specified energy release in the fuel. The figure of merit, FOM, is somewhat dependent upon the particular fuel specimen to be irradiated, and it therefore must be determined for each case. The computed FOM for this analysis, which should be representative for UN-PuN experiments, is

$$\text{FOM} \approx 1.8 \times 10^{-4} \text{ watt/cm}^3 \text{ per watt of reactor power}$$

This compares with a FOM of 1.12×10^{-4} watt/cm³ per watt of reactor power, as computed by G. E. for mixed-oxide irradiations in TREAT. The higher fissile atom density of the nitride fuel compared to the oxide fuel is believed to account for the larger figure of merit. The difference implies that lower TREAT reactor powers are going to be required than indicated previously. The value of the FOM will be recomputed for each particular experiment and experimental capsule using many of the neutronics parameters and results determined here.

Plans for Future Work

During the next fiscal year the detailed design of the TREAT capsule will be performed and the capsule will be fabricated; engineering and safety analyses will be performed and the transient irradiation of one UN-PuN-fueled experiment will be carried out.

- (1) Schoeberle, D. F., et al., "A Method of Calculating Transient Temperatures in a Multiregion, Axisymmetric, Cylindrical Configuration, The ARGUS Program", ANL-6654 (November, 1963).
- (2) Toppel, B. J., and Boksys, I., "The Argonne-Revised THERMOS Code", ANL-7023 (March, 1965).
- (3) Little W. W., Jr., and Hardie, R. W., "2DB User's Manual", BNWL-831 (1968).
- (4) Okrent, D., et al., "The Reactor Kinetics of the Transient Reactor Test Facility (TREAT)", ANL-6174 (September, 1960).
- (5) McGoff, D. J., "FORM, A Fourier Transform Fast Spectrum Code", NAA-SR-5766 (September, 1960).

Model Development for Irradiation-Induced Swelling of (U, Pu)N

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Objective of Program

To develop a mathematical model for the swelling of nuclear fuels whose mechanical behavior is representative of that believed to be occurring when (U, Pu)N fuel surrounded by liquid sodium is irradiated in the low flux range, and to evaluate the influence various parameters may have on the rate of swelling.

Approach Being Used

A purely mechanical approach is adopted in meeting this objective. A quantitative analysis of an idealized model of a clad fuel element is used to provide further insight into the swelling problem and to suggest ways in which swelling can be retarded.

Significant Accomplishments During Fiscal Year 1970

The possibility of controlling fuel swelling during irradiation by incorporation of a controlled porosity in the fabricated pellets is being evaluated. Preliminary results show no influence of variations in density, from 80 to 95 percent of theoretical, on the diametral expansion of the clad fuel.

Summary of Research Carried Out

The motivation for the development of the model is to evaluate the possibility of eliminating liquid sodium in the annulus around nitride fuels in LMFBR fuel rods. The model developed makes the following assumptions to analyze the problem:

- (1) The fuel has uniform porosity throughout the deformation process. No new pores are generated during burnup.
- (2) The fuel material is approximated by polyhedral cells associated with each pore. The deformation behavior of the polyhedral cell is approximated by the deformation behavior of a spherical shell of equivalent volume.
- (3) The pressure of the fission gases in the pores is assumed to be negligible; surface-tension effects are neglected.
- (4) The fuel and cladding obey a nonlinear creep law.
- (5) Isothermal creep is assumed.
- (6) The solid fuel material is allowed to change its density through volumetric expansion due to fission atom production. This is introduced in the creep law in the following manner:

$$\dot{\epsilon} = A \left| \sigma^* \right|^n \exp (-Q/RT).$$

The equation expresses the relationship between the strain rate, $\dot{\epsilon}$, and the stress, σ^* , at the temperature, $T(K)$, for a material whose activation energy is Q (cal per mole). R is the gas constant and A and n are constants describing the creep properties of the material. The stress is so formulated as to make dilation, a measure of the volume change locally, equal to zero for all times. Therefore, to introduce volumetric expansion due to fission-product atoms, the equation is modified as

$$\dot{\epsilon} = A \left| \sigma^* \right|^n \exp (-Q/RT) + 1/3 \mu \Gamma,$$

where μ is the volumetric expansion caused by a fission-product atom and Γ is the fission rate.

- (7) A state of hydrostatic stress, which is equal to the reactive internal pressure of the cladding, is assumed to prevail in the fuel material.
- (8) Changes in volume of a radial slice of fuel are constrained to occur in the radial direction.
- (9) No gap is assumed to exist between the fuel and the cladding.
- (10) The cladding acts to restrain volume expansion of the solid fuel into the available porosity of the fuel.

This mathematical model is analyzed numerically. The program has been coded in the G. E. time-sharing system.

Using the model, the following case for nitride fuel was evaluated:

Inner radius of the cladding = 0.1475 in.
 Outer radius of the cladding = 0.1575 in.
 Fission rate = 7.5×10^{13} fissions/(cm³)(sec)
 Creep strength of the fuel (1000 C) = 3000 psi
 Creep strength of the cladding (700 C) = 7500 psi
 Creep stress exponent of the fuel = 3.0
 Creep stress exponent of the cladding = 4.0
 Volumetric expansion = 3×10^{-22} cm³ per fission.

Calculations were obtained for initial material density levels of 95 and 80 percent of theoretical. The results of these calculations are plotted in Figure A-52. They show

that for the postulated behavior and for these density levels the influence of porosity on swelling is negligible. Thus, the initial indication is that little benefit can be gained by increases in the fuel porosity, irrespective of the swelling restraint by the cladding.

FIGURE A-52. SWELLING BEHAVIOR OF CLAD NITRIDE FUELS INITIALLY 80 TO 95 PERCENT DENSE

The volumetric expansion caused by fission-product atoms is assumed to be $3 \times 10^{-22} \text{ cm}^3$ per fission in this mathematical model, and the fission rate is $7.5 \times 10^{13} \text{ fissions}/(\text{cm}^3)(\text{sec})$.

Plans for Future Work

To fully explore the potential of the porosity level in the retardation of fuel swelling, further parametric evaluations are planned. In particular, lower values of μ , more characteristic of fuel swelling due to solid fission-product atoms, will be examined. Alternatively, the porosity of an annular fuel geometry as a possible mechanism for retarding fuel swelling will be explored.