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EFFECT OF STOICHIOMETRY ON THE BEHAVIOR OF MIXED-OXIDE FUEL DURING EXTENDED OPERATION IN FAILED PINS

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ABSTRACT

Two mixed-oxide fuel pin sections containing fuel of 1.95 and 1.99 O/M were purposely defected and irradiated to an average burnup of 8700 MWd/Te in a flowing sodium environment. Stoichiometry was a sensitive parameter in this experiment. The pin section containing 1.99 O/M fuel sustained a total volumetric increase of 21% compared to 10% for the 1.95 O/M fuel section. Average peak diametral changes in the 1.99 and 1.95 O/M fuel pin sections were, respectively, 7% and 23%. Analysis suggests that in a reactor where the oxygen content of the sodium is maintained at a low value, the extent of sodium-fuel reaction and consequent swelling would be smaller. Less than 1% of the fuel was lost from either section.

1. INTRODUCTION

Experiment B9A, reported here, is one of a series of in- and out-of-reactor experiments being carried out to investigate the effects of mixed-oxide fuel failures on the operation of a liquid metal cooled fast breeder reactor (LMFBR). The objectives of this segment of the Fast Ceramic Reactor Development Program (FCR Program) are to provide basic data for reactor design and safety analysis. Successful development of technically and economically feasible LMFBR's is contingent upon the development of knowledge of the origin of fuel failure and of the consequences of operating with failed fuel. Determination of whether a reactor can continue to operate safely with failed fuel pins within the core for extended time periods is essential to the development of an economically feasible plant. In addition, knowledge of the erosion characteristics of the fuel, the amounts and species of fission products released from failed fuel pins, and the environment most suitable for amassing and/or depositing fission products must be obtained. This information will provide a basis for designing reactors for safe operation, safe habitation by operating personnel, and economic maintenance.

1.1 OBJECTIVES

The objectives of the B9A experiment were to determine:

- The effect of the initial stoichiometry (O/M) of the mixed-oxide fuel on the amount of fuel-sodium reaction occurring in a system with a constant oxygen inventory.*
- The effect of fuel-sodium reaction on the diametral cladding strain or deformation of initially failed fuel pins operated under conditions typical of the peak power and coolant temperatures of prototype liquid metal cooled fast ceramic reactors (LMFBR's).
- The amount of fuel released from a failed fuel pin due to erosion or dissolution of fuel by flowing sodium.
- The amounts and species of fission products released from a failed fuel pin to the sodium coolant.

* Thermodynamic calculations indicate that decreasing the fuel stoichiometry should decrease the amount of fuel-sodium reaction occurring.

2. SUMMARY

Two mixed-oxide fuel pin sections containing fuel with stoichiometries of 1.95 and 1.99 were defected and irradiated in-pile to an average burnup of 8700 MWd/Te. During irradiation the fuels were exposed to sodium flowing at a velocity of 10 to 14 ft/sec. A summary of the capsule operating conditions and irradiation history is shown in Table 1.

Post-irradiation examination of the fuel pin sections showed that each had increased in diameter; the diameter increase was a function of the fuel stoichiometry. The 1.95 and 1.99 O/M fuel sections were determined to have total volumetric changes of 10% and 21%, respectively. Maximum changes in the average diameter of the 1.95 and 1.99 O/M fuel sections were 7% and 23%, respectively. The total volumetric change observed was several times that calculated for uniform formation of fuel-sodium reaction product within the fuel sections, indicating that initially the reaction occurred locally in the region of the initial defect. Analysis of the thermodynamics and kinetics of mixed-oxide fuel-sodium reactions indicates that the volumetric increases in these experimental pins may be greater than would occur in an operating LMFBR because of the high oxygen content in the capsule.

The original defects, located in the center of each fuel section, were observed to have propagated toward each end of the fuel sections. Loss of small amounts of fuel from each fuel section was observed in regions away from the original defect. The total amount of fuel lost was ~1%.

Gamma spectroscopy indicated that cesium was readily removed from the fuel. Ruthenium and zirconium were not leached from the fuel.

Table 1
SUMMARY OF CAPSULE B9A OPERATING
CONDITIONS AND HISTORY

PEAK POWER ^(a)	
Fission ^(b)	23.8 kW/ft
Gamma	0.1 kW/ft
Total	23.9 kW/ft
SODIUM COOLANT	
Flow rate	7.4 gpm
Velocity	12 ft/sec (average)
Inlet Temperature	930°F
Outlet Temperature	1014°F
ΔT Over Test Section	84°F
ΔT Between Centers of 1.95 and 1.99 Fuel	42°F
OPERATING DAYS	25
AVERAGE BURNUP ^(c)	8700 MWd/Te

(a) Peak power determined from burnup analysis on fuel from each fuel pin section.

(b) Based on 180 MeV per fission.

(c) Determined by radiochemical analysis of neodymium isotopes.¹ Peak burnup is ~1.04 times average. Burnup value is average value of analyses from both fuel pin sections.

3. EXPERIMENT PARAMETERS

The experimental parameters and operating conditions selected for the experiment are listed in Table 2.

The 14-inch fuel pin was divided into two sections by a plug. Except for stoichiometry the two fuel sections were identical in design. The lower and upper sections of the pin contained fuel with stoichiometries (O/M's), respectively, of 1.95 and 1.99. These two stoichiometries were chosen based on the thermodynamic analysis made by Aitken² of possible chemical reactions between mixed-oxide fuel, sodium, and stainless steel. His analysis indicated that the amount of fuel-sodium reaction occurring in a failed mixed-oxide fuel pin should decrease with decreasing stoichiometry at comparable operating conditions. Therefore, it was predicted that the 1.95 O/M fuel section would exhibit less diametral cladding strain than the 1.99 O/M fuel section.

The planned operating conditions were selected on the basis of the predicted peak overpower conditions for the Fast Flux Test Facility (FFTF) and the General Electric Demonstration Plant. For this purpose a peak fuel pin power of 18–20 kW/ft and a bulk coolant exit temperature of 1000°–1050°F was selected. An average coolant velocity of 10 ft/sec was planned. This coolant velocity is significantly lower than the 20 to 30 ft/sec velocities expected in present LMFBR designs. This compromise from typical LMFBR conditions was required because of limited pump head available for this capsule design.

An average coolant velocity of 10 ft/sec was calculated to produce a ΔT of 90°F across the fuel test section and a temperature difference of 45°F between the centers of the fueled sections. It is believed that the final equilibrium state of each fuel section is governed by thermodynamic considerations and insensitive to reaction rate. For this reason the 45°F temperature difference is expected to cause negligible differences in the final end states of the two fuel sections.

Table 2
B9A EXPERIMENT AND OPERATING PARAMETERS

Number of Specimens	2		
Orientation of Specimens	Axially-Aligned		
Fuel Parameters	Upper Section		Lower Section
Type		Solid Pellet	
Powder Preparation Process		Coprecipitation	
Fabrication Process		Cold Press and Sinter	
Composition (Pu/U+Pu)		0.249 ± 0.001	
Stoichiometry	1.989 ± 0.001		1.955 ± 0.001
U-235 Enrichment (%)		93.06 ± 0.32	
Density (gm/cc)		10.23 ± 0.09	
Pellet Diameter (in.)		0.2160 ± 0.0002	
Length of Fuel (in.)		5.031 ± 0.017	
Cold Diametral Gap (in.)		0.005 ± 0.002	
Clad Parameters			
Material		316 SS	
Outer Diameter (in.)		0.250 ± 0.001	
Wall Thickness		0.015 ± 0.002	
Inside Diameter (in.)		0.220 ± 0.001	
Metallurgical Condition (a)		31-1/2% CW	
Operating Parameters	Planned		Actual
Peak Power (kW/ft) (b)	18–20		23.9
Coolant Exit Temperature (°F)	1000–1050		1014
Axial Fuel Pin ΔT (°F)	90		84
ΔT Between Centers of Fuel Pin Sections (°F)	45		42
Coolant Velocity (ft/sec)	10		12
Average Burnup (MWd/Te) (c)	7300		8700
Operating Days	25		25

(a) CW = Cold Worked

(b) Based on 180 MeV/fission

(c) Determined by Radiochemical Analysis for Neodymium Isotopes¹

4. CAPSULE DESIGN AND IRRADIATION FACILITY

A summary description of the capsule and irradiation facility is given in this section. A detailed description of each piece of equipment is given in Appendix D.

4.1 FORCED CONVECTION CAPSULE (FCC) DESIGN

A schematic of the B9A capsule design is shown in Figure 1. The capsule design essentially provides a flowing sodium loop, the sodium being pumped by an electromagnetic helical induction pump. The fuel pin test section was doubly contained by two concentric sealed tubular sections. Starting at the pump at the top of the capsule the sodium coolant is pumped down the outer sodium annulus to the capsule bottom, then up through the flowmeter and fuel pin test section to the pump inlet. On its upward pass through the fuel pin test section, the sodium conducted away the fission heat produced in the fuel. On its downward pass through the outer annulus, the heat was conducted radially out of the capsule to the reactor pool water. An insulating gas chimney filled with argon separates the inner and outer sodium annuli to prevent radial conduction from the "hot" sodium in the inner annulus to the "cold" outer annulus. A binary gas annulus surrounding the outer sodium annulus permitted the bulk temperature of the sodium coolant to be controlled by altering the resistance to radial heat flow to the reactor pool water. The transfer of heat radially was controlled by varying the ratio of nitrogen to helium in the gas annulus.

Capsule instrumentation consisted of thermocouples and a flowmeter. Thermocouples were located at the inlet and outlet of the fuel test section and at the top of the up flow annulus. The thermocouples permitted the measurement of the ΔT across the fuel pin test section. The electromagnetic flowmeter was of the permanent magnet type with a nominal accuracy of 5%. (It was inoperative as described in Section 5.)

The fuel pin test section consisted of three pins in a triangular array and surrounded by a fluted liner as shown in Figure 2. Two of the pins were dummies. The other pin consisted of two axially aligned sections containing mixed-oxide fuel. The pins were arranged with a pitch-to-diameter ratio of 1.2, had an 0.250-inch outer diameter and were 14 inches long. The pins were spaced by spirally wrapped wires (on dummies only) and spacers. The inner tube of the test section was shaped (fluted) to simulate additional pins surrounding the three-pin cluster.

The fueled pin consisted of two sections axially aligned as shown in Figure 3. Each section of the pin had a fuel column nominally 5 inches in length, clad with cold worked type 316 stainless steel tubing. The fuel columns were axially restrained as shown in Figure 3 and were identical except for stoichiometry. The fabrication parameters for both fuel sections are listed in Table 2.

Also shown in Figure 3 is the defect mechanism for the two fueled sections. The defects consisted of a 60-degree notch, 1 inch long by 0.012 inch deep, machined parallel to the axis near the middle of each fuel pin section. Only 0.003 inch of the original 0.015-inch thick cladding wall remained. Three 1/32-inch diameter holes on 1/4-inch centers were drilled radially through each notch. The drill holes were filled with a lead alloy braze to ensure containment of the fuel during fabrication. The braze composition was chosen to be chemically reactive in sodium so that it would be reacted away during the capsule sodium filling operation. The composition of the braze is given in Table D-1 of Appendix D.

4.2 IRRADIATION FACILITY

The B9A capsule was irradiated in the front row of the General Electric Test Reactor (GETR) pool in a special facility. The facility permitted both vertical and lateral (tilt) movement of the capsule with respect to the reactor core. This permitted the fuel pin to be maintained in a constant neutron flux so that the power in each fuel pin section could be maintained constant during the irradiation as the reactor flux profile changed. Figure 4 illustrates the relative position of the B9A capsule with respect to the reactor core. A detailed description of the facility is given in Appendix D.

Figure 1: Schematic of Capsule B9A Design

Figure 2. Cross Section Through Capsule Test Section

Figure 3. Capsule B9A Fuel Pin

Figure 4. Capsule B9A Reactor "Pool" Facility

5. OPERATION OF THE EXPERIMENT

The capsule was filled with sodium from an external cleanup loop. The sodium was circulated and cold-trapped in the external loop until a plugging temperature of less than 270°F was obtained. This corresponded to an oxygen impurity level in the sodium of less than 5 ppm.³ The sodium fill and cleanup loop is shown in Figure 5. After sodium filling, the capsule was maintained at 550°–800°F to prevent freezing of the sodium. An analysis of the oxygen available during the experiment is given in Appendix B. Prior to insertion of the capsule into the reactor pool, the binary gas annulus was purged with high purity xenon (xenon has a very low thermal conductivity) to insulate the capsule sodium. Immediately after insertion of the capsule in the reactor, the electromagnetic pump was turned on. The pump, because of its low efficiency, input enough heat to maintain the sodium at 450°F.

During the pretest checkout, all instruments except the flowmeter were found to be operating satisfactorily. The signal output from the flowmeter was found to be less than 10% of the expected value.* The loss of the flowmeter prevented direct calorimetric measurement of operating power. As a result, the capsule power was adjusted by positioning the capsule according to reactor physics calculations of capsule power versus distance from the pressure vessel.** During the reactor refueling outages, the sodium in the capsule was maintained molten by purging the binary gas annulus with xenon and continuing to operate the electromagnetic pump. The experiment was operated without incident for the entire 25-day reactor cycle.

The capsule operating conditions for the reactor cycle are summarized in Table 1.

* The problem was later diagnosed and found to be the result of misorientation of the poles of the flowmeter magnet.⁴

** See Appendix A, R9A Thermal Analysis.

Figure 5. Forced Convection Capsule Sodium Fill and Cleanup Loop

6. POST-IRRADIATION EXAMINATION

A detailed post-irradiation examination was carried out to determine the condition of the capsule and fuel pins and the distribution of fuel and fission products within the capsule. The examination consisted of the following:

- a. Gross gamma scans of the capsule to determine the extent and location of fuel and/or fission-product redistribution. Point gamma scans to identify source isotopes.
- b. Neutron radiography on the test section of the capsule to determine gross fuel dimensional changes and fuel erosion.
- c. The capsule was sectioned and the fuel pins removed for visual examination and dimensional measurements.
- d. The fuel pins were sectioned and particular sections were metallographically prepared. The metallographic specimens were examined and radiographed to determine redistribution of fuel and fission-product species and identification of fuel-sodium reaction products.
- e. Microprobe analysis of the fuel-sodium reaction product and the distribution of uranium and plutonium.
- f. Radiochemical burnup analysis.

Results obtained in each step of the examination are described in the following subsections.

6.1 GAMMA SPECTROSCOPY

A gross gamma scan was taken of the bottom portion of the capsule including the fuel pins. In addition, multichannel scans were taken at various points along the capsule. Figure 6 shows the gross gamma energy profile along the length of the capsule. Locations of the point scans are indicated by the numbers placed along the gross gamma scan shown in Figure 6. The relative concentrations of the fission products scanned for at the points indicated in Figure 6 are listed in Table 3.

The gross gamma scan showed that the relative gamma energy in each fuel pin was comparable, indicating that the capsule was correctly positioned during irradiation and that the two pins operated at approximately the same power. Only minor activity was detected in the sodium circuit of the capsule. No major areas of deposition of fuel or fission products were detected. These scans indicate that little erosion or dissolution of fuel and fission products occurred during the 25-day experiment.

The gamma scan of the 1.99 O/M fuel section shows a peak in activity near the center of the section. The position and width of the peak indicate it corresponds with the location of the initial defect notch in the cladding. Of the fission products scanned, the peak was found to contain Ru-103 and Zr-95. This peak is discussed further in the subsections on neutron radiographs and visual examination of the fuel pin. Cs-137 was detected only in the plenum of the capsule indicating that it was readily removed from the fuel.

6.2 NEUTRON RADIOGRAPHY

Neutron radiographs of the fuel pin test section before and after the irradiation are shown in Figure 7.

A pronounced increase in the diameter of the 1.99 O/M fuel section, especially in the region of the defect notch, was clearly evident in the post-irradiation neutron radiographs. Material subsequently confirmed to be fuel or fuel-sodium reaction product was observed protruding from the surface of this fuel section. The mass of material protruded far enough into the coolant channel to contact the inner surface of the fluted liner surrounding the fuel pin cluster, a distance of approximately 0.080 inch. The extent of the protruding material corresponded with the gamma activity peak observed in the gross gamma scan. The velocity of the sodium flowing past this material was calculated to range from 10 to 14 ft/sec.

Only small dimensional changes were observed in the neutron radiographs of the 1.95 O/M fuel pin section.

Since the fuel pins were fabricated to the same dimensional specifications, the pronounced difference in diameter observed in the neutron radiographs was concluded to be directly related to the initial fuel stoichiometry. Quantitative differences in the diametral changes of the fuel sections are discussed in subsection 7.3.

6.3 VISUAL EXAMINATION AND DIMENSIONAL MEASUREMENTS

The three-pin cluster is shown in Figure 8 just after removal from the test section of the capsule. The mass of fuel or fuel-sodium reaction product protruding from the 1.99 fuel section can be seen in Figure 8. The fuel pin after disassembly of the three-pin cluster is shown in Figure 9. The protruding mass of fuel or fuel-sodium reaction product had been dislodged during the disassembly of the fuel pin cluster prior to taking the photographs shown in Figure 9.

Figure 6. Gross Gamma Scan of Capsule B9A

Table 3
LOCAL CONCENTRATIONS OF GAMMA ACTIVITIES
FOR FISSION PRODUCTS IN CAPSULE B9A

Relative Amount of Isotope (Background Subtracted)*

Position	Ru-103	Cs-137	Zr-95	Co-58	Co-60
Background	1	N.D.	N.D.	N.D.	
7	3	N.D.	4	3	20
12	40	N.D.	17	7	30
13	170	N.D.	50	13	30
14	70	N.D.	14	5	10
15	N.D.	N.D.	8	2	15
16	4	N.D.	4	2	25
18	15	N.D.	4	2	20
19	N.D.	N.D.	4	3	18
20	80	N.D.	26	7	12
21	230	N.D.	44	<0.1	11
22	70	N.D.	25	6	11
23	30	N.D.	7	4	13
24	60	N.D.	7	7	12
25	60	N.D.	18	6	9
26	50	N.D.	20	6	10
27	N.D.	N.D.	<0.5	2	8
28	1	N.D.	2	1	9
29	1	N.D.	1	1	6
30	2	2	1	1	6
31	2	2	3	0.5	3
32	0.5	4	0.5	0.5	3
33	N.D.	N.D.	<0.5	<0.1	<0.5
Plenum	N.D.	4	1	N.D.	N.D.
34	N.D.	4	0.9	N.D.	N.D.
35	N.D.	1	1	N.D.	N.D.
36	N.D.	1	N.D.	N.D.	N.D.
37	N.D.	1	0.6	N.D.	N.D.
38	N.D.	6	1	N.D.	N.D.
39	N.D.	1	0.9	N.D.	N.D.

N.D. = None Detected

* Relative disintegrations per unit time. Values are only relative for a particular isotope. Comparisons between isotopes are not valid.

TOP PIN - O/M 1.99

BOTTOM PIN - O/M 1.95

PRE-IRRADIATION NEUTROGRAPH

DEFECT AREA

DEFECT AREA

POST-IRRADIATION NEUTROGRAPH

Figure 7. Pre- and Post-Irradiation
Appearance of B9A Fuel Pins

Figure 8. B9A Fuel Pin Cluster Immediately After Disassembly of Capsule

Figure 9. Appearance of B9A Fuel Pin After Disassembly of Three-Pin Cluster

Visual examination indicated diametral increases occurred along the length of both the fuel pin sections. The cold worked cladding of each section was observed to be cracked an additional 2 inches above and below the original 1-inch long defect notches.

Micrometer measurements of the diameter of each fuel section are shown in Figures 10 and 11. The largest diametral increases occurred in both sections over the purposely defected area. The average* peak diametral changes measured in these regions were 7% and 23%, respectively, for the 1.95 and 1.99 O/M fuel sections. The total volume changes for the 1.95 and 1.99 O/M fuel sections were 10% and 21%, respectively.

The cross section of the 1.99 O/M pin was elliptic in the vicinity of the original defect. The major axis of the ellipse was approximately 45 degrees from the cladding defect. The cross section of the 1.95 O/M fuel section was round to within ± 0.010 inch.

6.4 METALLOGRAPHIC EXAMINATION

Each fuel section was sectioned and metallographic samples from the initial defect and peak power regions prepared.

No center void was observed in the defect region of the 1.99 O/M fuel. The 1.95 O/M fuel had a center void 0.020 inch in diameter at the center of the fuel section. In the remainder of the fuel sections, the center void was generally offset from the center of the pin toward the side opposite the cladding defect. It is believed that these observations reflect nonuniform heat transfer around the circumference of the pins caused by the presence of sodium or fuel-sodium reaction product after the cladding was defected.

Figures 12 and 13 show beta-gamma and alpha autoradiographs of the fuel in the initially defected and nondefected (peak power) regions of the fuel sections. The variation in the center void described above can be seen in these figures.

Figure 14 shows the appearance of the fuel in the defect region of the 1.99 O/M fuel section. Fuel in this section was difficult to retain during preparation as evidenced by the large areas of fuel fallout. Substantial amounts of fuel-sodium reaction product can be seen around the periphery of the fuel. Although the fuel shows no evidence of columnar grain growth or of a center void the increase in the volume of the fuel plus fuel-sodium reaction product was sufficient to crack the cladding directly opposite the initial defect notch.

The appearance of the fuel in the initially defected and nondefected region of the 1.95 O/M fuel section is shown in Figures 15 and 16. Unlike the 1.99 O/M fuel, the fuel in the defect region exhibits some columnar grain growth and a small center void as well as the reaction product at the fuel periphery. The amount of fuel fallout was significantly less than that observed for the 1.99 O/M fuel, indicating that the sodium did not react with or attack the fuel adjacent to the grain boundaries of the 1.95 O/M as it did the 1.99 O/M fuel.

The initially nondefected region of the 1.95 fuel exhibited a large center void and a well developed columnar grain structure. Fuel in the initially nondefected (peak power) region was very similar in appearance to that of the initially defected region.

Fuel erosion was observed to be minor in both fuel sections. Neither fuel section exhibited erosion in the region of the initial defect. However, indications of erosion were observed in the fuel adjacent to the defect notches. The alpha and beta-gamma autoradiographs of the initially nondefected regions shown in Figures 12 and 13 show regions where fuel was lost from the periphery of pellets exposed to the flowing sodium coolant by the breach in the cladding. There was no significant difference observed in the amount of fuel which appeared to be lost by erosion from the 1.95 and 1.99 O/M fuel sections. The amount of fuel lost from each section was estimated to be less than 1% based on the examination of the as-cut cross sections of the fuel sections.

6.5 MICROPROBE ANALYSIS

A sample of the 1.99 O/M fuel section was examined with a shielded electron microprobe. The cross section examined is shown in Figure 17. The microprobe examination showed that sodium had permeated throughout the fuel except for a small region located near the center of the fuel cross section. This apparently unaffected region of fuel is indicated in Figure 17. Sodium was detected only in the grain boundaries around the periphery of this "island" of fuel.

No variation in the ratio of uranium to plutonium was observed in the fuel sodium reaction product or the fuel. Variations in the concentration of sodium in the fuel-sodium reaction product were detected in regions of the reaction product adjacent to the cladding. Specimen current image pictures of two of the regions exhibiting variations in sodium concentration are shown in Figure 18.

* The average diameter for the fuel sections was determined by averaging the diameters from the profilometer traces taken at 0-, 45-, 90-, and 135-degree orientations. The profilometer traces are shown in Figures 10 and 11.

Figure 10. Profilometry Data for 1.95 O/M Fuel Section

Figure 11. Profilometry Data for 1.99 O/M Fuel Section

$\beta-\gamma$

α

FABRICATED DEFECT REGION

$\beta-\gamma$

α

INITIALLY NON-DEFECTED REGION

Figure 12. Alpha and Beta-Gamma Autoradiographs of B9A Bottom Fuel Pin (O/M 1.95)

$\beta-\gamma$

α

FABRICATED DEFECT REGION

$\beta-\gamma$

α

INITIALLY NON-DEFECTED REGION

Figure 13. Alpha and Beta-Gamma Autoradiographs of B9A Top Fuel Pin (O/M 1.99)

UNETCHED

Figure 14. Appearance of Fuel in Fabricated Defect Region of B9A Top Pin (O/M 1.99)

UNETCHED

Figure 15. Appearance of 1.95 O/M Fuel In Region Of Defect Notch

UNETCHED

Figure 16. Representative Appearance of Fuel in Initially Non-Defected Region of B9A Bottom Pin (O/M 1.95)

Figure 17. Micrograph of Microprobe Sample From 1.99 O/M Fuel Section

(a) PICTURE OF AREA 1 NOTED
IN FIGURE 17.

(b) PICTURE OF AREA 2 NOTED
IN FIGURE 17.

*Figure 18. Specimen Current Image Pictures of Fuel-Sodium Reaction Product in 1.99 O/M Fuel Section
Showing Variations in Sodium Concentration*

6.6 BURNUP ANALYSIS

Fuel burnup was determined by two techniques: (1) neodymium and (2) heavy element analyses. Results of the analyses are listed in Table 4. The neodymium values are judged to be the more accurate.

Table 4
BURNUP ANALYSES OF SAMPLES FROM PEAK
POWER REGION OF FUEL SECTIONS*

Fuel Section	Nd-148 (MWd/Te)	Heavy Element (MWd/Te)
1.95 O/M	8600	8300
1.99 O/M	8800	10000

* Average burnup based on 180 MeV/fission. Peak burnup is ~1.04 times average. Nd-148 values for each fuel pin section were averaged and the average value (8700 MWd/Te) used in calculating the power in each fuel pin section. The Nd values were used because they are deemed to be more accurate than the heavy element values at low burnups.

7. DISCUSSION OF RESULTS

The objective of the B9A experiment was to study the behavior of defected, stainless steel clad, mixed-oxide fueled pins operating at typical LMFBR conditions. A specific test variable was the comparison of the performance of two different fuel stoichiometries, 1.95 and 1.99. In addition, data on the erosion of mixed-oxide fuel in a flowing sodium environment and the release and distribution of fission products were to be obtained.

7.1 THERMODYNAMIC AND KINETIC CONSIDERATIONS

A short review of the data available on the thermodynamics and kinetics of mixed-oxide fuel-sodium reactions is given in this section and applied to the analyses of the results of the B9A test. Available data on the thermodynamics and kinetics of mixed-oxide fuel-sodium reactions were recently compiled and interpreted by Aitken, et al.^{2,5,6}

Thermodynamically, the amount of reaction product formed from the reaction of mixed-oxide fuel and sodium is dependent on the amount of oxygen available in the system and the temperature of the components. In a mixed-oxide fuel pin the primary source of oxygen is the fuel. In the event of a fuel pin failure the oxygen present in the sodium coolant may also enter into the reaction. Thermodynamic thresholds exist for the formation of the fuel-sodium reaction product for each source contributing oxygen.

An oxygen threshold dependent on the fuel stoichiometry was calculated using the equations and data developed by Aitken.² For the operating conditions of the B9A experiment, it was calculated that no fuel-sodium reaction would occur if the initial O/M of the fuel in contact with the sodium was less than 1.949. If the average fuel stoichiometry was greater than 1.949 the fuel and sodium would react producing fuel-sodium reaction product. When the fuel O/M was reduced to 1.949, the reaction would then cease.* In the B9A experiment the 1.95 and 1.99 O/M fuel sections both operated under similar conditions. Therefore, the fuel in each section should have been reduced to an average O/M of 1.949 as a result of the formation of fuel-sodium reaction products. The amount of oxygen available to react from each of the fuel sections was:

Initial Fuel O/M**	Final Fuel O/M	Oxygen Available for Reaction (O/M Units)
1.955	1.949	0.006
1.989	1.949	0.040

Other sources of oxygen for reaction are the oxygen atoms released by fissioning of the uranium and plutonium atoms. Using the method of Anselin,⁷ it was estimated that 0.009 O/M units per 10,000 MWd/Te of oxygen would be available from fully enriched mixed-oxide fuel assuming that oxygen combined with fission-product cesium will be released in the presence of sodium. The total amount of oxygen available from reaction from the fuel in each section of the B9A pin after 8700 MWd/Te of burnup was calculated to be:

Source of Oxygen	1.95 O/M Fuel (O/M Units)	1.99 O/M Fuel (O/M Units)
Excess in fuel	0.006	0.040
Fissioning [0.009(8700/10,000)]	0.008	0.008
TOTAL	0.014	0.048

Under B9A operating conditions, the equilibrium concentration of oxygen in the sodium for the initiation fuel-sodium reaction is in the range of 70 to 130 ppm.⁶ If the sodium contains more than 70–130 ppm oxygen, oxygen would be taken from the sodium to react with the fuel (and sodium) until the oxygen in the sodium was reduced to 70–130 ppm. The sodium would tend to reduce the fuel and any fuel-sodium reaction products formed if its oxygen

* This is rigorously true only if the presence of the fuel-sodium reaction product does not alter the temperature conditions of the fuel and sodium.

** The actual U/M of the 1.95 and 1.99 O/M fuel sections were 1.955 and 1.989, respectively.

concentration was less than 70–130 ppm until the oxygen concentration in the sodium increased to 70 to 130 ppm. No further reduction of the fuel-sodium reaction product would then occur. Sodium, however, would continue to react with fuel and any excess oxygen available from the fuel to form the fuel-sodium reaction product.

Taking into account the sources of oxygen available in the B9A capsule and in the sodium used to fill the capsule, it was calculated that the initial oxygen concentration in the sodium coolant of the capsule could range from about 5 to 45 ppm (see Appendix B). As a result, the sodium coolant in the B9A experiment would reduce the fuel-sodium reaction products formed in each fuel section until the concentration of oxygen in the sodium increased to 70 to 130 ppm. At this point, the sodium would continue to react with fuel and any excess oxygen available from the fuel, and the total inventory of fuel-sodium reaction products in each fuel section would increase.

The kinetics of the reactions occurring is just as important as whether or not a reaction will occur. If the rate of formation of the fuel-sodium reaction products were slow, no fuel pin deformation would occur over the lifetime of the pin. However, the results of the B9A experiment and others indicate that this is not the case since significant cladding deformation was observed in failed pins. No data are presently available on the kinetics of the fuel-sodium reaction except those obtained through experience with in-reactor failures. These data provide only qualitative information on the kinetics of the reaction since the exact time and conditions under which the failures occurred are not accurately known. The most significant information provided by these "failures" is that the rate of formation of the fuel-sodium reaction product is initially much greater than the rate of reduction and dissolution of the reaction product, even in flowing sodium of very low oxygen content (<5 ppm). It is noted, however, that none of the in-reactor failures referenced above contained fuel of 1.95 or lower stoichiometry.

7.2 EFFECT OF FUEL STOICHIOMETRY ON FUEL PERFORMANCE

Based on the thermodynamic and kinetic considerations discussed in subsection 8.1, it would be predicted that the 1.95 O/M fuel would exhibit superior performance as compared to 1.99 O/M fuel because of the smaller amount of oxygen available for reaction with fuel and sodium. The 1.95 O/M fuel had less than one-third the amount of oxygen available for reaction as the 1.99 O/M fuel: 0.014 O/M units versus 0.048 O/M units. The amount of diametral increase observed in the two B9A fuel sections supports this prediction.

Aitken, et al.⁵ report that the volume change associated with the formation of the reaction product is about 1.2% $\Delta V/0.01$ O/M units entering into the reaction. On this basis the maximum volume change of the 1.95 and 1.99 O/M fuel sections should have been 1.68% and 5.76%, respectively. The average volume increase determined for the 1.95 and 1.99 O/M fuel sections by graphical integration of the area under the profiles plotted in Figures 10 and 11 was 10% and 21.4%, respectively. The large difference between the volume increases calculated and determined from the profile traces is attributed to nonuniform formation of the fuel-sodium reaction product. Instead of reaction occurring uniformly around the periphery of the fuel sections, it most likely formed in the region of the defect, and this hindered the access of sodium to other regions of the fuel. Nonuniform formation of reaction products could also explain the mass of fuel-sodium reaction product observed protruding from the initial defect region of the 1.99 O/M fuel section. If the breach in the cladding became filled with sodium-fuel reaction product, the only way for the reaction to continue would be for sodium to diffuse through the reaction product to the fuel interface where it would react, increasing the thickness of the reaction product layer. As the layer increased in thickness, it would apply a greater stress to the cladding opening up the defect further which would increase the surface area for diffusion and promote further reaction. At some point the breach in the cladding would open to a width where the reaction product would tend to "extrude" through the breach. If the reaction product was not plastic, it might crack exposing the fuel surface for further reaction with sodium. The formation of this reaction product at the base of the layer of reaction product would also tend to push the reaction product outward from the fuel surface through the defect in the cladding.

In an operating LMFBR, it is very unlikely that the fuel-sodium reaction product observed extending into the coolant channel at the position of the defect in the 1.99 O/M fuel section would ever occur. The reason for this is that the oxygen concentration in the sodium in an LMFBR will be maintained at less than 10 ppm. As a result of the low oxygen content and large sodium inventory the sodium coolant in an operating reactor will act as an infinite sink for reduction of the fuel-sodium reaction product. As a result of the infinite reducing capability of the sodium, any fuel-sodium reaction product formed and exposed directly to the flowing sodium would be reduced or dissolved away. Experience with the failure of mixed-oxide pins exposed to sodium of <5 ppm oxygen has confirmed that the reaction product will be reduced. In the B9A experiment the sodium was calculated to initially contain ~40 ppm oxygen. Since there was no way of removing oxygen from the sodium in the capsule, the sodium would reduce only the reaction product formed until the oxygen in the sodium increased to 70–130 ppm. Since the amount of oxygen available from the fuel in the B9A experiment would increase the oxygen content of the sodium, a maximum of ~220 ppm, only

about 13% to 40% of the sodium reaction product which could be formed would be reduced. Therefore, it is feasible that the amount of fuel-sodium reaction product observed in the B9A experiment was larger than would be predicted in an operating LMFBR in which oxygen is continuously removed from the sodium if necessary to maintain concentration below 10 ppm.

7.3 FUEL EROSION

Little evidence of fuel erosion or loss was detected during the examination of the B9A experiment. Based on the examination of the transverse sections cut from each fuel section the amount of fuel lost was estimated to be less than 1%. No significant difference was observed in the amount of erosion which occurred in the 1.95 versus 1.99 O/M fuel.

In an operating LMFBR it is possible that the higher coolant velocities (20–30 ft/sec) might produce greater erosion than observed in the B9A experiment where the sodium velocity ranged from 10–14 ft/sec.

Erosion was principally observed in regions of the fuel away from the initial defect. Loss of fuel from these regions is not believed to be the result of fuel erosion per se, but the thermal shocking of the fuel resulting from the propagation of the cladding rupture. Initially, the fuel in the region of the defect was in contact with sodium. Upon going to power, the fuel and sodium in the region of the defect reacted and strained the cladding. As the cladding strain increased locally, the cladding ruptured axially along the length of the fuel sections exposing the "hot" fuel surfaces to relatively cold sodium. The thermal shock resulting from the coolant (~1000°F) coming in contact with the fuel surface (~2600°F) caused local cracking of the fuel. Unbonded fuel particles would then be swept away by the flowing coolant.

7.4 FISSION-PRODUCT RELEASE

Gamma spectroscopy of the capsule showed that essentially no Ru-103 or Zr-95 was released to the coolant. Cs-137 was detected only in the plenum of the capsule indicating that cesium in the fuel was readily removed by dissolution in sodium.

8. CONCLUSIONS AND RECOMMENDATIONS

- The amount of diametral cladding strain occurring in a failed mixed-oxide fuel pin will be a function of the initial fuel stoichiometry. The peak diametral cladding strain was observed to be approximately three times larger at an initial O/M of 1.99 than it was at 1.95.
- Loss of fuel by erosion of the flowing sodium coolant was less than 1% for both the 1.95 and 1.99 O/M fuels. The coolant velocity ranged from 10–14 ft/sec.
- Transport of the fission products Ru-103 and Zr-95 into the sodium coolant was not detected. Essentially all of the Cs-137 was removed from the fuel.
- The conditions of the B9A experiment were atypical of those expected in an operating LMFBR in two respects:
 1. The sodium velocity was 12 ft/sec. Coolant velocities in operating LMFBR's are expected to range from 20 to 30 ft/sec.
 2. The total amount of oxygen in the capsule was constant. As a result the oxygen concentration in the sodium in the capsule increased to the level where reduction of the fuel-sodium reaction product no longer occurred. This may have caused diametral cladding strains and volumetric increases greater than those which would occur in an operating LMFBR. The oxygen content of the coolant in an operating LMFBR will be maintained below 10 ppm which will result in continuous reduction of the fuel-sodium reaction product.

RECOMMENDATION

It is recommended that future experiments on defected fuel behavior utilize getters to maintain low oxygen concentrations.

It is also recommended that further work be carried out to determine the kinetics of the fuel-sodium reaction and to provide data which will more accurately determine the thermodynamics and rate of the reaction.

ACKNOWLEDGMENTS

This report is the final step of many steps required to complete the first defected fuel pin experiment in a forced convection capsule. It is appropriate to acknowledge the key people involved in the design, fabrication, irradiation, and examination of this first-of-a-kind experiment. W. O. Wade was responsible for schedule and cost control in the design and fabrication of the capsule. G. W. Tunnell was the principal design engineer for the capsule and facility.

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APPENDIX A

B9A THERMAL ANALYSIS

During the operation of the experiment the power level calculated by reactor physics was confirmed by operating the capsule for a brief time in a mode which permitted detailed thermal analysis. The binary gas annulus was purged with helium during full power operation. Measured temperatures were compared to temperatures calculated for the detailed thermal analysis of the capsule under the same binary gas composition. The results are summarized in Table A-1.

Table A-1
B9A CALIBRATION TEST WITH 100% HELIUM
IN THE BINARY GAS ANNULUS

	Detailed Thermal Analysis Calculations	Values Calculated From Measurements During Operation with 100% Helium
Power:		
Fission	18.25 kW	18.72 kW
Gamma*	1.11 kW	1.14 kW
Total	19.36 kW	19.86 kW
Coolant: (Sodium)		
Flow Rate	0.6778 lbm/sec	0.76 lbm/sec
Inlet Temperature	607.1°F	625°F**
Outlet Temperature	694.7°F	705°F**
ΔT	87.6°F	70°F**
GETR Pool: (Water)		
Bulk Temperature	110°F	110°F

* Includes gamma heating in fuel, stainless steel and sodium.

** Experimentally measured value.

The actual total power is approximately

$$Q_T = Q_0 \left[\frac{\bar{T} - \bar{T}_{H_2O}}{\bar{T}_0 - \bar{T}_{H_2O}} \right]$$

Where,

$$\begin{aligned}
 Q_0 &= 19.36 \text{ kW,} && \text{total power input in detailed thermal analysis (Table A-1).} \\
 \bar{T}_0 &= \frac{694.7 + 607.1}{2} && \text{average of inlet and outlet sodium temperatures calculated} \\
 &&& \text{in detailed thermal analysis. Table A-1)} \\
 &= 650.9^\circ\text{F}
 \end{aligned}$$

$$\bar{T}_{H_2O} = 110^{\circ}F, \quad \text{average pool water temperature.}$$

$$\bar{T} = \frac{705 + 625}{2}, \quad \text{average of inlet and outlet sodium temperatures measured (Table A-1).}$$

$$\begin{aligned} Q_T &= 19.36 \left[\frac{665 - 110}{650.9 - 110} \right] \\ &= 19.86 \text{ kW} \end{aligned}$$

The actual gamma heat is approximately

$$Q_{\gamma} = Q_{\gamma_0} \left[\frac{Q_T}{Q_0} \right]$$

Where,

$$Q_T = 19.86 \text{ kW}, \quad \text{actual total power.}$$

$$Q_0 = 19.36 \text{ kW}, \quad \text{total power input in detailed thermal analysis.}$$

$$Q_{\gamma_0} = 1.11 \text{ kW}, \quad \text{total gamma heating input in detailed thermal analysis.}$$

$$\begin{aligned} Q_{\gamma} &= (1.11) \left[\frac{19.86}{19.36} \right] \\ &= 1.14 \text{ kW} \end{aligned}$$

The actual fission heat is approximately

$$Q_F = Q_T - Q_{\gamma}$$

Where,

Q_T and Q_{γ} are as defined above

$$Q_F = 19.86 - 1.14$$

$$= 18.72 \text{ kW}$$

The actual flowrate is approximately

$$\dot{M}_T = \dot{M}_0 \left[\frac{(\Delta T)_0}{(\Delta T)_T} \right] \times \frac{Q_T}{Q_0}$$

Where,

Q_T and Q_0 are as defined above

$$\begin{aligned}\Delta T_T &= 705 - 625, && \text{measured temperature difference} \\ &= 80^\circ\text{F}\end{aligned}$$

$$\begin{aligned}\Delta T_0 &= 694.7 - 607.1, && \text{calculated temperature difference from detailed thermal analysis.} \\ &= 87.6^\circ\text{F}\end{aligned}$$

$$\dot{M}_0 = 0.6778 \frac{\text{lbm}}{\text{sec}}, \quad \text{mass flowrate input in detailed thermal analysis}$$

$$\begin{aligned}\dot{M}_T &= (0.6778) \left[\frac{87.6}{80} \times \frac{19.86}{19.36} \right] \\ &= 0.76 \text{ lbm/sec}\end{aligned}$$

The normal operating power for the B9A experiment can now be calculated using the flowrate calculated in the previous section. A summary of the operating conditions is given in Table A-2.

Table A-2
SUMMARY OF B9A OPERATING CONDITIONS
CALCULATED FROM CALIBRATION TEST RESULTS*

	Calculated From Data Experimentally Measured	GETR Physics Calculations
Power:		
Fission	22.5 kW/ft	20.0 kW/ft
Gamma (in fuel only)	0.1 kW/ft	0.1 kW/ft
TOTAL	22.6 kW/ft	20.1 kW/ft
Coolants (Sodium):		
Flowrate	6.7 gpm	6.0 gpm
Inlet Temperature	930°F	930°F
Outlet Temperature	1014°F	1014°F
ΔT	84°F	84°F

* The power level presented in the main text was calculated from post-irradiation burnup values and differs from these values.

Total power during operation was:

$$Q = \dot{M} C_p \Delta T$$

Where,

$$\dot{M} = 0.76 \frac{\text{lbm}}{\text{sec}}, \quad \text{actual mass flowrate as calculated in the previous section.}$$

$$C_p = 0.3 \frac{\text{Btu}}{\text{lbm} \cdot ^\circ \text{F}}, \quad \text{specific heat of sodium.}$$

$$\begin{aligned} \Delta T &= 1014 - 930, \quad \text{measured temperature difference during normal operation} \\ &= 84^\circ \text{F} \quad \text{(Table A-2).} \end{aligned}$$

$$K = 1.05 \frac{\text{kW sec}}{\text{Btu}}, \quad \text{conversion fraction from Btu to kW sec}$$

$$\begin{aligned} Q &= (0.76) (0.3) (84) (1.05) \\ &= 20.1 \text{ kW} \end{aligned}$$

Total gamma heat (i.e., in fuel, stainless steel, sodium) was:

$$Q_\gamma = Q_{\gamma_0} \left[\frac{Q}{Q_0} \right]$$

Where,

Q_0 and Q_{γ_0} are as defined in previous section.

Q is as defined above

$$\begin{aligned} Q_\gamma &= (1.11) \left[\frac{20.1}{19.36} \right] \\ &= 1.2 \text{ kW} \end{aligned}$$

Gamma heating in the fuel was approximately

$$Q_{\gamma F} = \text{kW}$$

Where,

$$K = 1.5 \text{ watts/gram}, \quad \text{gamma heating in first row of GETR pool.}$$

$$W = 62 \text{ grams}, \quad \text{weight of fuel}$$

$$Q_{\gamma F} = 0.1 \text{ kW}$$

Total fission heat is:

$$Q_F = Q - Q_\gamma$$

Where,

Q and Q_γ are defined above

$$\begin{aligned} Q_F &= 20.1 - 1.2 \\ &= 18.9 \text{ kW} \end{aligned}$$

Linear fission power is:

$$\left(\frac{Q}{L} \right)_F = \frac{Q_F}{L}$$

Where,

$$Q_F = 18.9 \text{ kW, total fission heat}$$

$$L = 5/6 \text{ ft, length of fuel}$$

$$\begin{aligned} \left(\frac{Q}{L} \right)_F &= \frac{18.9}{5/6} \\ &= 22.5 \text{ kW/ft} \end{aligned}$$

Linear gamma heat in fuel pin is:

$$\left(\frac{Q}{L} \right)_{\gamma_F} = \frac{Q_{\gamma_F}}{L}$$

Where

Q_{γ_F} and L are defined above

$$\begin{aligned} \left(\frac{Q}{L} \right)_{\gamma_F} &= \frac{0.1}{5/6} \\ &= 0.1 \text{ kW/ft} \end{aligned}$$

Total linear power in the fuel pin is:

$$\left(\frac{Q}{L} \right)_T = \left(\frac{Q}{L} \right)_F + \left(\frac{Q}{L} \right)_{\gamma_F}$$

Where $\left(\frac{Q}{L} \right)_F$ and $\left(\frac{Q}{L} \right)_{\gamma_F}$ are defined above

$$\begin{aligned} \left(\frac{Q}{L} \right)_T &= 22.5 + 0.1 \\ &= 22.6 \text{ kW/ft} \end{aligned}$$

Coolant flowrate in GPM is:

$$\dot{Q} = K \frac{\dot{M}}{\delta}$$

Where,

$$K = 450 \frac{\text{gal-sec}}{\text{ft}^3 \cdot \text{min}}, \quad \text{conversion factor}$$

$$\dot{M} = 0.76 \text{ lbm/sec}, \quad \text{mass flowrate}$$

$$\delta = 51.2 \text{ lbm/ft}^3, \quad \text{sodium density}$$

$$\begin{aligned} \dot{Q} &= 450 \left[\frac{0.76}{51.2} \right] \\ &= 6.7 \text{ gal/min} \end{aligned}$$

APPENDIX B

CALCULATION OF OXYGEN AVAILABLE IN B9A

There were four possible sources of oxygen in the B9A experiment. They were (1) the initial oxygen impurity level in the sodium, (2) the oxide layer on the stainless steel surfaces, (3) the oxygen absorbed onto the fuel surface, (4) the oxygen released during the fissioning, and the oxygen released by fuel due to a temperature gradient:

- (1) The initial oxygen level in the sodium was determined by a plugging temperature indication of $<270^{\circ}\text{F}$. This according to Eichelburger³ corresponds to <2 ppm oxygen in sodium.
- (2) The amount of oxygen in the oxide layer on stainless steel surfaces which have been cleaned and passivated has been experimentally determined to be from $0.15 (10^{-2}) \text{ gm/meter}^2$ to $0.3 (10^{-2}) \text{ gm/meter}^2$. * Another source⁹ indicates from calculations that the reduction of oxygen from stainless steel surfaces by sodium is $\sim 5 (10^{-2}) \text{ gm/meter}^2$. The exposed surface area in B9A was $\sim 0.8 \text{ meter}^2$. This gives the following equivalent oxygen impurity level in the sodium.

By reference (8)

$$\begin{aligned} \text{wt of oxygen} &= 0.15 (10^{-2}) \frac{\text{gm}}{\text{m}^2} (0.8 \text{ m}^2) \\ &= 0.120 (10^{-2}) \text{ gm} \end{aligned}$$

$$\begin{aligned} \text{impurity level} &= \frac{0.0012 \text{ gm of oxygen}}{1000 \text{ gm of sodium}} \frac{10^3}{10^3} \\ \text{of oxygen} &= 1.2 \text{ wppm} \end{aligned}$$

$$\text{for } 0.3(10^{-2}) \text{ gm/m}^2 \text{ impurity level of oxygen} = 2.4 \text{ wppm}$$

By reference (9)

$$\begin{aligned} \text{wt of oxygen} &= 5 (10^{-2}) \frac{\text{gm}}{\text{m}^2} (0.8 \text{ m}^2) \\ &= 4 (10^{-2}) \text{ gm} \end{aligned}$$

$$\begin{aligned} \text{impurity level} &= \frac{0.04 \text{ gm O}_2}{1000 \text{ gm Na}} \left(\frac{10^3}{10^3} \right) \\ \text{of oxygen} &= 40 \text{ wppm} \end{aligned}$$

- (3) The average value for total off-gassing of 22 samples for the B9A fuel batch was $27.6 \mu\text{l/gm}$. Of this amount ~ 2.2 was CO and $<1\%$ was O_2 . Neglecting the O_2 there is $\sim 2.2 \mu\text{l/gm}$ of CO. For the 61.9 gm of fuel in B9A this corresponds to $\sim 137 \mu\text{l}$ of CO. This corresponds to $\sim 1(10^{-4}) \text{ gm}$ of oxygen which in terms of impurity level in 1 kg of sodium is

$$\text{sodium is } \frac{1(10^{-4})}{10^3} \left(\frac{10^{+3}}{10^3} \right) = 0.1 \text{ wppm.}$$

* See reference 8.

- (4) The reaction which describes the oxygen uptake from the fuel phase can be written as follows:

Where X is the initial departure from stoichiometry

Y is the final departure from stoichiometry.

For the B9A experiment the final O/M of the fuel was calculated to be 1.949. However, excess oxygen would be available for formation of Na_3MO_4 as a result of fissioning of the fuel. This would amount to ~0.009 O/M units per 10,000 MWd/Te burnup. The average burnup of the B9A fuel was 8700 MWd/Te, therefore, the amount of oxygen available from fission was:

$$\begin{aligned} \text{O}_{2f} &= 8700 \text{ MWd/Te} (0.009 \text{ O/M units per } 10,000 \text{ MWd/Te}) \\ &= 0.008 \text{ O/M units} \end{aligned}$$

In calculating the amount of oxygen available for reaction or the amount of Na_3MO_4 formed during the life of the B9A experiment this oxygen can be thought of as an increase in the initial fuel O/M. Therefore, the 1.95 and 1.99 O/M fuels would have O/M's of 1.963 and 1.997, respectively. X and Y and the coefficients for each of the components in equation (1) for these O/M's are given in the table below,

Fuel Section	X	Y	$\frac{Y-X}{2+Y}$	$\frac{2+X}{2+Y}$	2-Y	2-X
1.95	0.037	0.051	0.00683	0.9932	1.949	1.963
1.99	0.003	0.051	0.0234	0.9766	1.949	1.997

The grams of oxygen available from the 1.95 O/M fuel is:

$$W_{\text{MO}_{1.963}} = 30.942 \text{ gm}$$

$$\text{Moles}_{\text{MO}_{1.963}} = 0.249 (239) + 0.751 (235) + 1.963 (16)$$

$$= 59.511 + 176.485$$

$$= 267.404$$

$$\text{Moles}_{\text{MO}_{1.949}} = 0.9932 \left\{ \left[0.249 (239) + 0.751 (235) \right] + 1.949 (16) \right\}$$

$$= 0.9932 \left[(59.511 + 176.485) + 31.184 \right]$$

$$= 234.391 + 30.972$$

$$= 265.363$$

$$\begin{aligned}
 W_{\text{MO}_{1.949}} &= 265.363 \left(\frac{30.942}{267.404} \right) \\
 &= 30.706 \text{ g} \\
 W_{\text{O}_2 \text{ Reacted}} &= 30.942 \left(\frac{31.408}{267.404} \right) - 30.706 \left(\frac{30.972}{265.363} \right) \\
 &= 3.643 - 3.584 \\
 &= 0.050 \text{ g}
 \end{aligned}$$

The grams of oxygen available from the 1.99 O/M fuel is:

$$\begin{aligned}
 \text{MO}_{1.997} + 0.072 \text{ Na} &\rightleftharpoons 0.0234 \text{ Na}_3 \text{ MO}_4 + 0.9766 \text{ MO}_{1.949} \\
 W_{\text{MO}_{1.997}} &= 30.965 \\
 \text{Moles}_{\text{MO}_{1.997}} &= 0.249 (239) + 0.751 (235) + 1.997 (16) \\
 &= 59.511 + 176.485 + 31.952 \\
 &= 267.948 \\
 \text{Moles}_{\text{MO}_{1.949}} &= 0.9766 \left\{ \left[0.249(239) + 0.751(235) \right] + 1.949 (16) \right\} \\
 &= 0.9766 \left[(59.511 + 176.485) + 31.184 \right] \\
 &= 230.474 + 30.454 \\
 &= 260.928 \\
 W_{\text{MO}_{1.949}} &= 260.928 \left(\frac{30.965}{267.948} \right) \\
 &= 30.154 \text{ g} \\
 W_{\text{O}_2 \text{ Reacted}} &= 30.965 \left(\frac{31.952}{267.948} \right) - 30.154 \left(\frac{30.454}{260.928} \right) \\
 &= 3.692 - 3.521 \\
 &= 0.173 \text{ g} \\
 \text{Total O}_2 \text{ Reacted} &= 0.050 + 0.173 \\
 &= 0.223 \text{ g}
 \end{aligned}$$

If all of this oxygen were taken up by the sodium the ppm O_2 in the 1000 g of Na would be:

$$\frac{0.223 \text{ g O}_2}{1000 \text{ g Na}} \left(\frac{10^3}{10^3} \right) = 223 \text{ wppm}$$

Table B-1
OXYGEN AVAILABLE IN B9A EXPERIMENT

Source	Oxygen Level in Sodium, wppm
(1) Initial Sodium Purity	<2
(2) Oxide Layer on Stainless Steel Surfaces	2-40
(3) Off-Gassing from Fuel Pellets	<1
(4) Oxygen Redistribution in Fuel and Oxygen Released in Fissioning	70-223*
 TOTAL	 75-226 wppm

-
- * Lower limit assuming that oxygen is taken from the surface of the fuel until its O/M is reduced to 1.97. The upper limit if all of the oxide in the pin reached thermodynamic equilibrium in the time of operation.

APPENDIX C

CALCULATION OF THE TEMPERATURE AT THE FUEL-SODIUM INTERFACE

The instantaneous temperature change at the fuel-sodium interface as given in Carslaw and Jaeger¹¹ is from ~2560°F to ~1400°F.

$$\begin{aligned}
 T_{\text{INTERFACE}} &= T_{\text{SODIUM}} + \frac{(T_{\text{FUEL}} - T_{\text{SODIUM}})}{1 + \left(\frac{K}{\sqrt{\alpha}} \right)_{\text{SODIUM}} \left(\frac{\sqrt{\alpha}}{K} \right)_{\text{FUEL}}} \\
 &= T_s + \frac{(T_F - T_S)}{1 + \left(\frac{K_s}{K_F} \right) \left(\sqrt{\frac{\alpha_F}{\alpha_s}} \right)}
 \end{aligned}$$

Where,

- T_s = Temperature of sodium
- K_s = Thermal conductivity of sodium
- α_s = Thermal diffusivity of sodium
- T_F = Fuel surface temperature
- K_F = Thermal conductivity of fuel
- α_F = Thermal diffusivity of fuel

$$\begin{aligned}
 &= 1000 + \frac{2556 - 1000}{1 + \left(\frac{37.7}{1.32} \right) \left(\sqrt{\frac{0.0242}{2.3902}} \right)} \\
 &= 1000 + \frac{1556}{1 + 2.872} \\
 &= 1000 + 402
 \end{aligned}$$

$$T_{\text{INTERFACE}} \cong 1400^\circ\text{F}$$

APPENDIX D

DETAILED DESCRIPTION OF CAPSULE AND IRRADIATION FACILITY

FORCED CONVECTION CAPSULE DESIGN

The capsule was designed to provide a flowing sodium environment typical of an LMFBR. This required control of the rate of sodium coolant flow by the test section and the rate of heat dissipation from the fuel pin. The coolant flowrate was controlled by an electromagnetic pump. The bulk sodium temperature and rate of heat dissipation was controlled by varying the ratio of helium/nitrogen in a binary gas annulus (see Figure D-1).

FLOW CONTROL AND MEASUREMENT

The electromagnetic (EM) pump was controlled by a motorized autotransformer. Pump voltage and current were monitored for each of the three phases of the pump stator. In addition, phase-to-ground detectors were provided to indicate short circuit in the stator. The flow and developed head of the EM pump varied with the temperature of the sodium. The nominal flowrate was achieved with the pump operating at ~28% of rated power.

An electromagnetic flowmeter was located near the bottom of the capsule at the inlet to the test section. The flowmeter was of the permanent magnet type.

TEMPERATURE CONTROL AND MEASUREMENT

The bulk sodium temperature was controlled by an insulating gas chimney and the binary gas annulus. As shown in Figure 1 an inert gas (argon) chimney separated the sodium up-pass from the down-pass. This prevented regenerative heat transfer between the legs of the flow loop. The binary gas annulus was formed by the primary and secondary containment tubes. The annulus was filled with a mixture of helium and nitrogen. The rate of radial heat transfer from the sodium down-pass to the reactor pool water was controlled by varying the composition of the helium/nitrogen gas mixture. The gas composition could be automatically or manually controlled. In the automatic mode the signal from a thermocouple at the exit to the test section operated a valve which adjusted the gas mixture to maintain a constant temperature.

A total of eight thermocouples (T/C's) were installed in the capsule. Two thermocouples monitored temperature in the pump stator. All of the T/C's were chromel-alumel, the two in the pump stator were glass insulated. The eight capsule T/C's were insulated with MgO and sheathed in 304 ss. The stator winding T/C's were attached at the end turn hot spots. The eight capsule T/C's were placed in the sodium flow channel and arranged in three groups. Two T/C's were located at the inlet to the fuel region, three at the exit from the fuel region, and the last three were in the up-flow path midway between the top of the fuel and the top of the up-flow annulus. The positions of the T/C's in the capsule are shown in Figure D-1.

The argon chimney surrounding the up-flow annulus ensured that the sodium temperature would not drop more than 4°F, so there were in effect six T/C's monitoring the exit temperature. During reactor shutdown periods the capsule sodium was maintained molten by the heating losses from the EM pump. In addition, two heaters were installed to provide auxiliary heating capacity. The heaters had nichrome wire elements insulated in MgO and sheathed in inconel.

TEST SECTION AND DEFECT MECHANISM

The test section was designed to mock up a multipin array. This was accomplished by surrounding the fuel pin with two dummy pins and a fluted liner as shown in Figure 2. The dummy pins were also used as conduits for the flowmeter and thermocouple leads. The dummy pins were spiral wire-wrapped to promote turbulent mixing of the sodium coolant and to minimize the radial and circumferential temperature gradients in the test section.

The fuel pin consisted of two axially-aligned sections as shown in Figure 3. The fuel pin was not wire-wrapped to maintain a clean flow path around the pin and to direct as much of the coolant flow around it as possible. Each fuel section had a defect mechanism to breach the cladding and expose the fuel to sodium. The defect mechanism was designed to provide containment of the fuel until fabrication of the capsule was complete. The defects were designed to open immediately on exposure to molten sodium. The defect mechanism in each fuel section consisted of a 60-degree notch and three holes (1/32-inch diameter) filled with braze material. The braze material had a low melting point (514°F) and was chemically reactive in sodium. Table D-1 shows the composition of the braze material and the impurities added to the sodium as a result of dissolution. These impurity levels were so low as to have negligible effect on the experiment.

Table D-1
COMPOSITION OF BRAZE FILLER MATERIAL
USED IN B9A DEFECT MECHANISM

Composition:	Pb	—	75%
	Zn	—	15%
	Sn	—	10%
	Cu	}	Trace
	Fe		
	Ni		
Melting Temperature:	514°F		
Density:	9.63 gm/cc		
Total Volume Used in B9A (2 pins)	V	=	1.78(10 ⁻³)cc
Impurity Levels in Sodium (0.9 Kg)			
Due to Braze	Pb	—	15 ppm
	Zn	—	3 ppm
	Sn	—	2 ppm
	Total	—	20 ppm

IRRADIATION FACILITY AND PUMP STATOR

The irradiation facility was designed to support the capsule in the reactor* pool and provide controls for positioning the capsule relative to the reactor core. The pump stator was attached to the facility and supported the capsule. The pump stator could be raised/lowered and tilted relative to the facility thereby providing positioned control of the capsule as shown in Figure D-2.

The capsule pump stator generated the traveling electromagnetic field to pump the capsule sodium. A water manifold provided cooling water (~30 gpm) to the stator to prevent overheating of the insulated stator windings.

* General Electric Test Reactor (GETR).

197R441		MULTI-PHASE POOL FACILITY	
		15000 GPM, 15000 GPM, 15000 GPM	
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97	1/2" 1/2" 1/2"	161 452-010	
98	1/2" 1/2" 1/2"	161 452-010	
99	1/2" 1/2" 1/2"	161 452-010	
100	1/2" 1/2" 1/2"	161 452-010	

Figure D-2. Forced Convection Capsule B9A V-RAFT Assembly

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