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IRRADIATIONS OF
U-20Pu-10Fs ALLOY FUEL RODS

by

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ABSTRACT

Instrumented capsule irradiations were performed with uranium-20 w/o plutonium-10 w/o fission compositions sodium bonded to various high-temperature jacketing alloys. The specimens were EBR-II-diameter fuel rods that had been reduced to a 51-mm fuel length. The jacketing materials tested were: niobium, Nb-1w/oZr alloy, Nb-33w/o-Ta-1w/oZr alloy, Nb-4w/oV alloy, vanadium, molybdenum, Ta-0.1w/oW alloy, Inconel-X, Type 304 stainless steel, and Hastelloy-X. A total of 78 specimens were irradiated over a temperature range of 420 to 900°C and burnups of 1.2 to 4.2 a/o heavy metal. Other specimens presently under irradiation have exceeded burnups of 7.2 a/o without indication of failure. It was found that rupture strength of the jacketing material is not the only criterion of the restraint characteristics of the jacket during irradiation. The optimum relationship appears to be a jacket that retains a high rupture strength with a high degree of ductility. The lifetime of the fuel specimens before failure occurred was also significantly extended by increasing the void space within the jacket.

INTRODUCTION

Argonne National Laboratory has designed and recently constructed the Experimental Breeder Reactor II (EBR-II) at the National Reactor Testing Station in Idaho. EBR-II is an unmoderated, heterogeneous, sodium-cooled reactor with a thermal power output capacity of 62.5 MW. The reactor plant includes a pyrometallurgical reprocessing facility, in which discharged fuel from the reactor is remotely processed, fabricated into rods, and reassembled into fuel elements for return to the reactor.

The pyrometallurgical reprocessing operation permits certain fission products to remain in the fuel. These fission products are collectively designated "fission" (symbol Fs). They include principally zirconium, molybdenum, technetium, and ruthenium, with lesser amounts of rhodium and palladium. Eventually these fission products reach equilibrium concentration

ratios in the fuel. The total amount of fission in the fuel can be varied, however, by the frequency of reprocessing and by the amount of fresh fuel added to each reprocessing cycle.

Core I has been fabricated from U-5w/oF's alloy. The fission in this alloy is synthetic, being compounded of natural isotopes and hence no technetium. Irradiation experiments completed on this fuel material indicate that it has a swelling temperature of 600 to 650°C in the range of 1.2 to 2.5 a/o burnup.*⁽¹⁾ The expected maximum central fuel temperature during reactor operation is expected to be only somewhat above the 600 to 650°C swelling range. Irradiations of jacketed fuel indicate that the restraint imposed by the 0.23-mm (0.009-in.)-thick stainless-steel jacketing will be sufficient to permit the fuel to achieve atom burnups on the order of 1.5% or more.

Fuel-alloy compositions for future metallic cores include plutonium, as well as additional fission. The originally selected composition was U-20w/oPu-10w/oF's. Irradiation experiments on unclad specimens of this alloy, containing synthetic fission, showed that it swelled rapidly above temperatures of about 370°C.⁽²⁾ Since this is 300°C below the normal maximum fuel temperature expected during reactor operation, considerable reliance must be placed on the jacket to maintain dimensional stability of the fuel elements.

An additional complicating feature of this plutonium fuel alloy is its tendency to form a liquid eutectic with iron or nickel base alloys at temperatures as low as 550°C. This temperature will be experienced at the fuel-jacket interface in the hotter regions of the core during normal reactor operation. The jacket must therefore be made of the refractory metals that do not form low-melting eutectics with plutonium alloys. Alternatively, a thin barrier of a refractory metal can be placed between the fuel and the iron- or nickel-alloy jacket.

As part of the development of a satisfactory EBR-II plutonium-alloy fuel element, a series of irradiations of fuel pins was begun in April 1961. The elements duplicate the cross-sectional dimensions and geometry of the standard EBR-II jacketed fuel pin, but the length, internal void space, and amount of bond sodium have been scaled down by a factor of about six. The irradiations are being conducted in instrumented capsules in the CP-5 reactor.

The main design parameter in the initial experimental irradiations has been the jacket composition. Other experimental design variables studied subsequently have included void volume, the effect of venting the

*The burnup figures reported are in terms of percent of heavy metal atoms.

jacket, and the effect of jacket thickness. The two principal test parameters are irradiation temperature and fuel burnup. Studies of the effect of varying composition were started more recently and will be reported later. This report describes the results of the experimental irradiations completed before July 1, 1964.

SPECIMEN MATERIALS AND ASSEMBLY

A. Description of the EBR-II Fuel Rod

The EBR-II fuel rod is a cylindrical element having overall dimensions of 461-mm (18.187-in.) length and 4.5-mm (0.18-in.) diameter. The fuel material is an injection-cast U-5w/oFs alloy rod measuring 361 mm (14.221 in.) in length and 3.65-mm (0.144-in.) diameter, inserted in a tube having a nominal wall thickness of 0.23 mm (0.009 in.). The fuel is bonded to the jacket with sodium, which fills the 0.15-mm (0.006-in.) annulus formed by the fuel and the jacket. The fuel can experience 15% radial expansion before making contact with the jacket. The sodium covers the top of the fuel by 16.5 mm (0.65 in.). From the top of the sodium to the bottom of the top seal plug is a helium-filled space referred to as void volume. The void volume was provided to accommodate the thermal expansion of the sodium. At the time of assembly, the helium gas in the void volume is at one atmosphere pressure. The percentage void volume in this report will refer to the ratio of the initial gas volume to the fuel pin volume at room temperature. The void volume of the EBR-II fuel rod with the fuel restrainer is 14.9%, but at operating temperatures this volume is reduced to 7.7% because of expansion of the sodium bond. Figure 1 is a drawing of the fuel rod.

Figure 1. Diagram of EBR-II Fuel Rod Assembly

B. The Fuel Specimen

The specimens included the reference cross-section dimensions, but for convenience, the fuel length dimension was reduced to 51 mm (2.0 in.). The jacket length was also reduced proportionately to provide the identical void volume relative to the fuel volume as exists in the full-length rod. The spacer wire spiraled around the outside of the production rod was omitted. The fuel material in the jacketed specimens was injection-cast U-20w/oPu-10w/oFs, which had been selected originally as the reference fuel for a plutonium alloy loading of the EBR-II reactor. Additional molybdenum and ruthenium were added to the fissium as a substitute for technetium. The composition of the alloy is shown in Table I.

Table I
TYPICAL COMPOSITION OF ALLOYS USED
FOR FUEL SPECIMENS

Element	w/o	Element	w/o
Uranium	70.6	Ruthenium	4.1
Plutonium	20.0	Rhodium	0.8
Molybdenum	2.6	Palladium	1.9

Pertinent physical characteristics of the fuel in the as-cast condition are: density, 16.8 g/cc; melting point, $\sim 820^{\circ}\text{C}$; and hardness, 74.0 R_A. A plot of the thermal conductivity of the fuel versus temperature is shown in Figure 2.⁽³⁾ Thermal cycling tests conducted on the fuel material between 475 and 750°C (the anticipated maximum temperature fluctuation in the reactor), showed no significant change in length and diameter.⁽⁴⁾ The coefficient of thermal expansion has been measured at an average of $21.5 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ between the temperatures of 605 and 720°C.

The alloy was made by induction heating under vacuum and in a graphite crucible which was coated with an Aquadag-thoria wash. The casting temperatures ranged from 1100 to 1250°C. The molds used were made of Vycor tubes and duplicated the dimensions of the molds for the production fuel elements. From the 382-mm (15-in.) long cast rods, 51-mm (2-in.) sections were cut for use as irradiation specimens. The jacket of the specimens duplicated the cross-section dimensions of the production fuel rod, but had been reduced in length to provide a ratio of internal void volume to fuel volume equal to that of a full-size fuel rod.

Figure 2
Thermal Conductivity versus Temperature
for U-20w/oPu-10w/oFs Alloy⁽³⁾

Figure 3 is a diagram of an irradiation specimen. The overall length approximated 89 mm (3.5 in.), with a diameter of 4.44 mm (0.175 in.). The fuel pin, which had an average diameter of 3.65 mm (0.144 in.), was sodium-bonded to the jacket. The sodium bond extended 1.5 mm (0.060 in.) above the top of the fuel. No centering or positioning arrangement was used to locate or restrict the fuel within the jacket. A liquid-metal bond was used in the EBR-II fuel rod to allow some longitudinal and radial expansion of the fuel without straining the jacket. Sodium was used to provide good thermal conductivity and to permit handling of the fuel rods at room temperatures without disturbing the bond layer.

The fuel pin was sealed in the jacket by two welded end plugs. The top plug was shaped so as to provide a flat portion on which the identification number of the specimen could be stamped.

Before the assembly of the specimens, the jacket material was subjected to rupture tests. The tubing was also nondestructively inspected by eddy-current, point-probe techniques and X-radiography. Defects such as inclusions, pits, or cracks greater than 10% of the wall thickness were considered a basis for rejection. Micrometric measurements were made of wall thicknesses. Unusual surface conditions were recorded and photographed. The fuel pin was measured for length, diameter, and density. Source radiography was performed on specimens in which density determinations indicated internal voids or porosity. Surface roughness was noted and recorded.

Figure 3
Diagram of Irradiation Specimen

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The specimens were assembled in a helium-filled glovebox whose atmosphere had an oxygen concentration less than 200 parts per million. The welding of the jacket, the loading of the specimen, the sodium filling, and the final seal welding were performed in this environment. However, closing the void space at the welding temperature resulted in a pressure less than one atmosphere at room temperature. After assembly, the specimens were removed from the glovebox and the welds were checked for leaks with a helium mass spectrometer. Wetting of the sodium bond to all contacting surfaces was attained by centrifuging the specimens for 5 min at 350°C. This bonding technique had proven superior to other methods previously attempted, which include: impacting, high-frequency vibration, ultrasonic vibration, spinning, or high-frequency, low-amplitude vibrations. Verification of the sodium bond was nondestructively determined by an eddy-current, point-probe technique. Any bonded specimen that had an area or bubble in the sodium layer larger than 1.6 mm (1/16 in.) was not considered acceptable. The specimens were then X-radiographed to ascertain the proper positioning of the fuel pin and the level of the sodium above the fuel. A volume measurement was made of the complete specimen, and length and diameter measurements were taken. The diameter measurements were taken at 90° positions at 13-mm (0.5-in.) intervals along the length of the specimen.

A total of 78 specimens have been irradiated in this phase of the refractory alloy jacketing study. The jacket materials selected for these experiments were those that were known to be compatible with the fuel material and to have relatively high rupture strengths. The materials that were tested up to the time of the writing of this report are listed below. The order of presentation does not signify the sequence of test, but rather a categoric grouping according to materials and application.

- A. Niobium
 - 1. Niobium
 - 2. Niobium-1 w/o zirconium
 - 3. Niobium-33 w/o tantalum-1 w/o zirconium
 - 4. Niobium-4 w/o vanadium
- B. Vanadium
 - 1. Vanadium
- C. Molybdenum
 - 1. Molybdenum
- D. Tantalum
 - 1. Tantalum-0.1 w/o tungsten
- E. Duplex Tubing
 - 1. Inconel-X (vanadium liner)
 - 2. Type 304 stainless steel (vanadium liner)
 - 3. Hastelloy-X (tungsten-coated)

The tubing for the specimen jackets were procured from commercial sources. Much of the material had to be discarded because of nonconformance to dimension as well as quality specifications. In addition to the non-destructive tests performed on the tubing, hydrostatic pressure tests were conducted on some materials with the aid of a manually operated hydraulic pump that developed a maximum pressure of 700 kg/cm² (10,000 psi). The results of the pressure tests on the material are listed in Table II.

Table II
RESULTS OF (SHORT-TIME AND ROOM-TEMPERATURE) PRESSURE TESTS CONDUCTED
ON 4.422-mm-OD x 0.23-mm-WALL-THICKNESS TUBING

Material	Test Pressure*		Yield Pressure		Burst Pressure	
	psi	kg/cm ²	psi	kg/cm ²	psi	kg/cm ²
Niobium	2,700	189	3,000-3,500	211-246	7,100-7,300	499-513
Nb-1w/oZr	4,000	281	4,500-5,000	316-351	6,300-6,700	443-471
Nb-33w/oTa-1w/oZr	6,700	471	7,200-7,500	506-527	8,700-8,900	611-625
Nb-4w/oV	-	-	7,500-8,000	527-562	10,000-11,000	703-773
Vanadium	3,700	260	4,100	288	11,600-11,800	815-829
Molybdenum**	-	-	7,200	506	9,750	685
Ta-0.1w/oW	-	-	7,800-9,100	548-639	9,200-9,300	646-653
Inconel-X	6,500	457	7,500	527	10,200-10,700	717-752
Type 304 SS	-	-	4,000-5,000	281-351	7,500-8,000	527-562
Hastelloy-X	8,000	562	8,500-8,800	597-618	11,300-11,600	794-815

*Some tubing which was to be used as jackets of fuel specimens was subject to a pressure test as a means of quality control.

**Data calculated from tensile test information.

EXPERIMENTAL PROCEDURES

The specimens were irradiated in instrumented, temperature-controlled capsules in the CP-5 reactor. The irradiation facilities were located in the intermediate ring of fuel tubes of the reactor core. The capsules were inserted in vertical thimbles that extended from the bottom of the fuel tubes to the top of the reactor. The capsules were cooled by the heavy-water reactor coolant flowing at a rate of 4.5 m/sec (15 ft/sec) through the 1.5-mm (0.063-in.) annulus formed by the capsule and the thimble.

The capsules were NaK-filled double-wall containers, 38 mm (1.5 in.) in diameter and 533 mm (21 in.) in length.⁽⁵⁾ Figure 4 is a diagram of the capsule. The specimens were arranged in two tiers, three specimens at each tier. They were held in place by spacer disks which restricted lateral as well as vertical motion but allowed a 0.8-mm (0.312-in.) preferential radial displacement about the fueled portion of the specimen. These spacer disks also restricted excessive NaK convection within the capsule. The arrangement of three specimens located at one tier was advantageous in that a direct comparison could be made between specimens which had been irradiated at comparable temperatures and burnups.

Temperature of the capsules was controlled by electric resistance heaters that encircled the two tiers of specimens. A maximum of 2 kW of auxiliary heating power was available, which was not sufficient to maintain temperature during reactor shutdown. The auxiliary heat was therefore used to compensate for neutron flux variations in the reactor core. Temperature control could be maintained at $\pm 4^{\circ}\text{C}$ for extended periods of time.

Adjacent to each specimen, at the midpoint of the fuel, a thermocouple continuously monitored the NaK temperature. With this instrumentation it was possible to determine the maximum fuel and jacket temperatures. These determinations were based on electrical analogue plots,⁽⁶⁾ which were later verified by directly measuring with thermocouples the center fuel temperatures in several specimens.

The irradiation period of each group of specimens was governed mainly by the integrity of the jackets. The condition of the specimens was determined by periodic removal of the capsule from the irradiation facility and nondestructively examining the contents by pinhole autoradiography⁽⁷⁾ and/or neutron radiography.⁽⁸⁾ When the autoradiographic technique was used, dimensional changes only of the fuel could be detected and measured. As an aid to this inspection, each capsule included a tantalum rod of known dimensions which, on being irradiated, provided a gamma-emitting standard whose image could be measured. Evidence of jacketing failure was usually seen as small localized bulges of the fuel rod. Neutron radiography offered

a considerable advantage in that in addition to the fuel pin, the capsule hardware, NaK level, and jacketing materials could be observed with maximum resolution.

Another surveillance technique, which was indirect but fairly reliable, was the instrumenting thermocouples. Any gross change in irradiation temperature as recorded by the thermocouple usually indicated that the specimen it was monitoring had deformed. A jacketing failure generally occurred at the portion of the specimen that was at the highest temperature. The thermocouples had been purposely positioned to read the highest temperature and consequently usually detected the initial deformation of the fuel rod as a result of jacket failure.

In the advent of a jacket failure, the specimen was identified by one of the nondestructive techniques and a decision was made as to whether the irradiation period should be continued. Since three different jacketing materials were generally irradiated as a group at one tier in the capsule, it was occasionally advantageous, for comparison purposes, to extend the irradiation period till a second cladding failed, thereby isolating the most promising material. By a systematic selection of irradiation specimen groups, many jacket materials could be evaluated.

The capsule was disassembled in a shielded cell equipped with a manipulator-operated glovebox. The nitrogen atmosphere within the glovebox was maintained at less than 2.5% oxygen content. The unirradiated fuel material is not pyrophoric in air of normal humidity, but tends to become pyrophoric after being irradiated to burnups in excess of one percent. The pyrophoricity is attributed to decay heating, increased surface area, and the presence of slight amounts of sodium.

Most of the capsules were filled with eutectic NaK. Some, however, were filled with sodium. Others were assembled with NaK that had a potassium concentration of only 13%. This composition, which has a consistency of soft putty at room temperature, had a thermal conductivity value 50% higher than that of eutectic NaK.⁽⁹⁾ The various NaK compositions were selected to vary the internal heat-transfer characteristics of the capsule, thereby setting the initial irradiation temperature of the specimens. Disassembly of capsules containing sodium and the Na-13w/oK alloy required the installation of furnaces in the glovebox to maintain the sodium or NaK in a liquid state. After removal from the capsule, the specimens were photographed while still in the supporting spacer disks. This was done to later determine the orientation of the jacket failure and to insure that sections of fuel material bound to the spacer disks could be recovered if broken off during handling. Wires containing cobalt were used as flux monitors and were located between specimens. These were removed, sectioned, dissolved, and analyzed radiochemically for cobalt-60. These results were used for a preliminary computation of the fuel burnup.

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Figure 4. Diagram of CP-5 Irradiation Capsule for Six Specimens Arranged in Two Tiers of Three Each

An optical inspection was made of each specimen, and photographs were taken of each defect area. Length, diameter, and density measurements were made of all intact specimens. Ruptured specimens were measured for length, but diameter measurements were taken only on portions of the jacket that were still intact. Bowing of the specimen was determined from magnified photographs of the full-length rod. Some intact specimens were selected for determinations of fission-gas release. This was accomplished by puncturing the jacket at a point above the fueled portion of the specimen. The gas was analyzed mass-spectrographically for fission product isotopes. The internal gas pressure was calculated from the volume of the gas obtained.

Transverse sections as well as longitudinal sections were made of specimens for metallography studies. One small section of the fuel was analyzed mass-spectrographically for uranium and plutonium isotopes so that fuel burnup could be calculated.

The irradiation capsule and components were closely inspected for structural damage, corrosion, and irradiation damage. It was noted in the earlier experiments that loose particles of fuel material were producing pits in the walls and bottom of the Zircaloy-2 capsules. A tantalum liner was added that completely encircled the two tiers of specimens thereby eliminating this occurrence. Irradiated thermocouples were recalibrated within the shielded cell and found to be accurate within the manufacturer's tolerances. The thermocouples were chromel-alumel, sheathed in stainless steel, and insulated with MgO.

RESULTS AND DISCUSSION

The irradiation conditions and the experimental results are summarized in Table III. The data are grouped according to jacket material and arranged in order of increasing fuel burnup.

Table III
EFFECTS OF IRRADIATION ON CAST U-20w/oPu-10w/oFs SPECIMENS

Spec. No.	Melt No.	Jacket Composition, w/o	Void Volume, %	Heavy Metal Burnup, a/o	Max. Temp., °C		Time Ave. Temp., °C		Condition of Specimen**
					Jacket Surface	Fuel Center	Jacket Surface	Fuel Center	
17-2	R21-10B	Niobium	13	1.4	655	760	595	690	B
17-6	R21-10C	Niobium	13	1.6	790	920	740	855	B
16-4	R21-10A	Niobium	13	1.7	780	910	715	830	B
18-2	R21-10D	Niobium	13	2.0	680	790	665	770	B
22-5	S2-3-2	Nb-1Zr	36*	0.7	615	715	555	640	C
22-2	S2-3-1	Nb-1Zr	36*	0.7	655	760	595	690	C
19-1	R21-19C	Nb-1Zr	28	1.2	520	595	490	570	B
19-6	R21-20C	Nb-1Zr	28	1.2	535	615	510	590	B
19-4	R21-21C	Nb-1Zr	28	1.2	535	615	510	590	B
17-1	R21-15D	Nb-1Zr	13	1.4	655	760	595	690	B
23-1	S2-3-4	Nb-1Zr	50	1.4	560	650	535	620	D
23-4	R21-11A	Nb-1Zr	50	1.4	630	730	560	650	A
20-2	R21-21F	Nb-1Zr	28	1.5	555	640	540	625	B
20-4	R21-22A	Nb-1Zr	28	1.5	580	670	550	635	B
20-6	R21-22D	Nb-1Zr	28	1.5	580	670	550	635	B
16-2	R21-15B	Nb-1Zr	13	1.6	695	810	630	730	B
17-4	R21-15E	Nb-1Zr	13	1.6	790	920	740	855	B
16-5	R21-15C	Nb-1Zr	13	1.7	780	910	715	830	B
18-4	R21-17B	Nb-1Zr	13	2.0	680	790	665	770	A
18-1	R21-17A	Nb-1Zr	13	2.0	680	790	665	770	B
21-4	S2-1-1	Nb-1Zr	25	2.1	520	600	395	490	B
21-1	R21-22B	Nb-1Zr	25	2.1	520	600	495	580	B
21-2	R21-24B	Nb-1Zr	25	2.1	520	600	495	580	B
27-4	R21-11E	Nb-1Zr	Vented	2.1	570	660	520	600	C
27-1	R21-11C	Nb-1Zr	Vented	2.1	570	660	520	600	A
25-4	R21-7B	Nb-1Zr	50	2.1	575	670	520	600	B
25-1	R21-7A	Nb-1Zr	50	2.1	580	670	530	620	C
28-1	R21-11B	Nb-1Zr	57	2.5	580	670	410	480	B
28-4	R21-11D	Nb-1Zr	57	2.5	580	670	520	610	C
32-1	R21-3B	Nb-1Zr(a)	23	4.2	590	690	460	535	A
32-2	R21-3C	Nb-1Zr(a)	24	4.2	590	690	460	535	C
32-3	R21-3D	Nb-1Zr(b)	24	4.2	590	690	460	535	A
32-4	R21-3E	Nb-1Zr(b)	22	4.2	615	720	480	560	A
32-5	R21-3F	Nb-1Zr(b)	23	4.2	615	720	480	560	A
32-6	R21-4A	Nb-1Zr(a)	17	4.2	615	720	480	560	A
22-4	S2-2-3	Nb-33Ta-1Zr	36*	0.7	615	715	555	640	A
22-1	S2-2-2	Nb-33Ta-1Zr	36*	0.7	655	760	595	690	B
23-3	R21-9A	Nb-33Ta-1Zr	50	1.4	560	650	535	620	B
23-6	R21-9B	Nb-33Ta-1Zr	50	1.4	630	730	560	650	A
27-6	R21-25F	Nb-33Ta-1Zr	Vented	2.1	570	660	520	600	A
27-3	R21-25E	Nb-33Ta-1Zr	Vented	2.1	570	660	520	600	A
25-6	R21-26D	Nb-33Ta-1Zr	50	2.1	575	670	520	600	B
25-3	R21-26C	Nb-33Ta-1Zr	50	2.1	590	680	575	670	A
29-1	R21-29E	Nb-4V	57	3.0	490	560	450	510	A
29-5	R21-3A	Nb-4V	57	3.0	500	570	460	520	A
29-4	R21-29F	Nb-4V	57	3.0	500	570	460	520	A
22-6	S2-2-4	Vanadium	50*	0.7	615	715	555	640	G
17-3	R21-17E	Vanadium	13	1.4	655	760	595	690	B
16-3	R21-17D	Vanadium	13	1.6	695	810	630	730	B
18-5	R21-18B	Vanadium	13	2.0	680	790	665	770	G
18-3	R21-17E	Vanadium	13	2.0	680	790	665	770	B
19-3	R21-20E	Molybdenum	30	1.2	520	595	490	570	B
20-1	R21-21B	Molybdenum	30	1.5	555	640	540	625	C
22-3	S2-2-6	Ta-0.1W	36*	0.7	655	760	595	690	B
19-2	R21-20A	Ta-0.1W	28	1.2	520	595	490	570	C
19-5	R21-20B	Ta-0.1W	28	1.2	535	615	510	590	C
23-2	R21-9D	Ta-0.1W	50	1.4	560	650	535	620	B
23-5	S2-3-3	Ta-0.1W	50	1.4	630	730	560	650	A
20-3	R21-27B	Ta-0.1W	28	1.5	555	640	540	625	A

Table III (Contd.)

Spec. No.	Melt No.	Jacket Composition, w/o	Void Volume, %	Heavy Metal Burnup, a/o	Max. Temp., °C		Time Ave. Temp., °C		Condition of Specimen**
					Jacket Surface	Fuel Center	Jacket Surface	Fuel Center	
20-5	R21-27D	Ta-0.1W	28	1.5	580	670	550	635	B
21-5	R21-H9	Ta-0.1W	25	2.1	520	600	360	420	C
21-6	S2-2-5	Ta-0.1W	36*	2.1	520	600	460	540	B
21-3	R21-24E	Ta-0.1W	25	2.1	520	600	495	580	B
27-2	R21-25A	Ta-0.1W	Vented	2.1	570	660	520	600	A
27-5	R21-25B	Ta-0.1W	Vented	2.1	570	660	520	600	A
25-5	R21-14A	Ta-0.1W	50	2.1	575	670	520	600	B
25-2	R21-7D	Ta-0.1W	50	2.1	590	680	530	620	A
16-1	R21-18B	Inconel X V Barrier	13	1.6	695	810	630	730	E
17-5	R21-18E	304 SS V Barrier	13	1.6	790	920	740	855	E
16-6	R21-18C	304 SS V Barrier	13	1.7	780	910	715	830	E
18-6	R21-18F	304 SS V Barrier	13	2.0	680	790	665	770	A
28-3	R21-28F	Hastelloy X W Lined	57	2.5	590	670	410	480	E
28-2	R21-28B	Hastelloy X W Lined	57	2.5	590	670	410	480	F
28-5	R21-28C	Hastelloy X W Lined	57	2.5	590	670	520	610	A
28-6	R21-28D	Hastelloy X W Lined	57	2.5	590	670	520	610	A
29-3	R21-29D	Hastelloy X W Lined	57	3.0	490	560	450	510	A
29-2	R21-29C	Hastelloy X W Lined	57	3.0	490	560	450	510	A
29-6	R21-29B	Hastelloy X W Lined	57	3.0	500	570	460	520	A

*No sodium bond.

**A. Specimen straight and intact.

B. Longitudinal split in jacket with protrusion of fuel.

C. Longitudinal split in jacket without protrusion of fuel.

D. Several small cracks around center section of specimen.

E. Fuel penetrated barrier and formed extensive eutectic with jacket.

F. Fuel penetrated barrier and formed small hole in jacket.

G. Specimen bowed but intact.

(a) Jacket thickness 0.38 mm (0.015 in.)

(b) Jacket thickness 0.64 mm (0.025 in.)

A. The Niobium Alloy Jacketing

The failures in the niobium and niobium alloys were characterized by brittle fracture of the jacketing material. The failures generally occurred at the hottest portion of the specimen, which was near the center fueled section of the specimen and extended along the length of the jacketing. In some instances there were several small longitudinal fractures around the circumference of a center portion of the specimen. Figure 5 shows typical failures. Once the jacketing had ruptured, the fuel, now being irradiated in an unrestrained condition, extruded through the opening. The initial failure was aggravated as the fuel continued to emerge from the break.

Occasionally a failure was followed by rapid movement of the fuel out of the defect area. This could be observed during irradiation when the specimen was at operating temperature. In most cases, the temperature-monitoring thermocouple adjacent to the specimen would record a

	EI-150	EI-1222	EI-1224	EI-1227	EI-1609
Specimen No.	18-4	25-1	25-3	25-6	29-4
Jacket Comp., w/o	Nb-1Zr	Nb-1Zr	Nb-33Ta-1Zr	Nb-33Ta-1Zr	Nb-4V
Burnup, a/o	2.0	2.1	2.1	2.1	3.0
Max. Jacket Temp., °C	680	580	590	575	500
Void Volume, %	13	50	50	50	57

Figure 5. Typical Results of U-20w/oPu-10w/oFs Fuel Material Irradiated in Niobium Alloy Tubings Having 0.23-mm (0.009-in.) Wall Thickness.

temperature excursion that would trip the instrument interlock system and shut down the reactor. Postirradiation examination of the specimen in question usually showed that the fuel had moved toward the thermocouple and on occasions engulfed the thermocouple sheath. This occurrence was common only to specimens having internal gas void volumes less than 28% and a central metal irradiation temperature above 550°C.

Diameter measurements on irradiated niobium-base jacketed specimens showed no deformation due to internal pressure even though the specimen had experienced a localized jacketing failure. There was no measurable bowing of the jackets in either the intact or failed specimens. This of course excludes the condition of complete separation of the jacketing where fuel deformation would produce an angular deformity between two straight sections of jacketing. The density measurement of the intact specimens also confirmed the absence of significant volumetric changes.

Metallographic examination of jacket and core showed incipient cracking of the inside diameter of the jacket (see Figure 6). Edges of the jacketing where complete separation had occurred had a jagged appearance characteristic of brittle fracture. Figure 7 is a photograph of a typical specimen at the point of failure.

106-6998

Figure 6. Incipient Jacket Failures in U-20w/oPu-10w/oFs Alloy Jacketed with Nb-1w/oZr Alloy after 1.6 a/o Burnup at 695°C

106-6996

Figure 7. Brittle Jacket Failure in U-20w/oPu-10w/oFs Alloy Jacketed with Nb-1w/oZr Alloy after 1.6 a/o Burnup at 790°C

An analysis of the irradiation results on the four niobium-base alloys listed in Table III failed to show a pronounced relationship between the room-temperature rupture strengths of the tubing and the ability of the tubing to prevent fuel swelling during irradiation. Specimens having void volumes less than 28% failed before 1.2 a/o burnup had been attained. Increasing the void volume or venting the jacket permitted a distinction to be made indicating that at the reference jacket temperatures the niobium and Nb-1w/oZr alloy jackets should be satisfactory up to 2.5 a/o fuel burnup. Increasing the jacket thickness from 0.23 mm (0.009 in.) to 0.64 mm (0.025 in.) enabled burnups in excess of 6 a/o to be obtained. The results of the tests conducted on these jackets are discussed in detail later in Section G of this report. The Nb-33w/oTa-1w/oZr alloy (0.23 mm thick) should be suitable up to 2.7 a/o burnup, and the Nb-4w/oV alloy might permit burnups up to 3.3 a/o burnup. This last-mentioned jacketing material remained slightly ductile, as evidenced by measurable changes in diameter of the irradiated specimens.

B. Vanadium

Five specimens having vanadium jackets were irradiated to maximum jacket temperatures of 695°C and burnups of 2.0 a/o. The tubing from which the jackets were fabricated was in the recrystallized condition and had been pressure-tested at 260 kg/cm² (3700 psi). It was therefore known

that although the tubing did not have high rupture strength it was highly ductile. The irradiation results showed that the jacket maintained ductility after prolonged exposure to neutron irradiation and high temperatures. When a jacket ruptured, it was a ductile failure and did not propagate as the fuel material extruded from the once-established opening.

Jacket failures were also accompanied with a noticeable bowing of the specimen. In addition, diameter changes of an average of 2% were measured, a diameter change of 14% being noted in one specimen. In specimens where the jacketing had not failed, bowing of the specimen was also prevalent.

Figure 8 is an enlarged view of a transverse section of the failed area of specimen 17-3. This specimen previously had a circular cross section and after failure approached an egg shape. The fuel material that extruded from the opening in the jacket was very porous. Though not clearly depicted in this photograph, porosity is generally largest in the center of the fuel material, becoming smaller near the jacket-fuel interface. Observations of areas where the jacket had ruptured showed ductile failure of the material. Figure 9 is a photograph of a transverse section of an irradiated specimen showing an area adjacent to such a failure.

106-6995

Figure 8. Release of U-20w/oPu-10w/oFs Alloy Fuel through Failed Vanadium Jacketing after 1.4 a/o Burnup at Maximum Jacketing Temperature of 655°C

106-6997

Figure 9. Ductile Jacket Failure in U-20w/oPu-10w/oFs Alloy Jacketed with Vanadium after 1.4a/o Burnup at Maximum Jacket Temperature of 655°C

C. Molybdenum

Two specimens that were jacketed with molybdenum were irradiated. The postirradiation examination of these specimens showed that the jacketing had become extremely brittle. The failures were represented by numerous longitudinal splits that extended the full length of the specimen. The end plugs of one specimen had separated from the jacket during irradiation, and additional fracturing occurred when the specimen was handled in the hot cell. From the amount of fuel material that extruded from the fractured areas it was possible to estimate that the failure occurred in the early stages of irradiation.

The annulus that separates the fuel material from the jacket in this series of specimens was 15% of the fuel volume. Calculations showed that after 0.13 a/o burnup at reference temperatures, the fuel had displaced the sodium from the annulus and was in direct contact with the jacketing. The instrumenting thermocouples for these specimens indicated that the fuel was not restrained by the jacketing after 0.4 a/o burnup. The results of this test are not considered conclusive in that the quality of the tubing material was known to be poor.

D. Tantalum

1. Tantalum-0.1 w/o Tungsten

This jacket material duplicates the composition that was selected for the LAMPRE reactor.⁽¹⁰⁾ The intended application is different, but the temperature conditions were similar.

Fourteen specimens were irradiated. By comparison, this jacketing offered a higher degree of restraint than the jacket materials previously described. Unlike the niobium-alloy jacket materials, the fractures did not lengthen as the fuel extruded from the opening. No failed jackets had long longitudinal cracks, but small cracks around the circumferences of several of the specimens resulted in localized bulged sections. Several specimens had more than one failure along the length of their fueled portions. This behavior is attributed to weakened areas in the jacketing resulting from undetected defects existing in the tubing before irradiation.

The irradiation results make it possible to establish that with 0.23-mm (0.009-in.)-thick jackets a probable maximum operating range at the reference temperature is in the neighborhood of 3.5 a/o burnup.

E. Duplex Tubing

1. Inconel-X (Vanadium Liner)

One specimen assembled had a standard-dimension Inconel-X jacket, the interior of which was lined with 0.013-mm (0.0005-in.)-thick vanadium foil. The foil was overlapped to preclude the possibility of the fuel contacting the jacket. The bottom plug, on which the specimen rests, was also protected by foil. This barrier was incorporated to prevent the formation of the eutectic at 550°C between the fuel and the jacket. The specimen was irradiated to 1.6 a/o burnup at a maximum clad temperature of 695°C. Postirradiation examination of the jacket showed that the fuel had penetrated the barrier and formed a eutectic with the jacket. A portion of the specimen that was intact was sectioned and examined metallographically. Figure 10, a photograph of this section, shows that the fuel is bearing directly against the barrier, and the cladding material is uniform and intact.

2. Type 304 Stainless Steel (Vanadium Liner)

Three specimens, assembled in the manner described above for the Inconel-X jacketing, were irradiated at a maximum jacket temperature of 790°C and fuel burnup of 2.0 a/o. The temperatures were considerably higher than the eutectic formation temperature between the jacketing and the fuel. It was essential to determine if this configuration could be operated at critical temperatures since a production fuel loading would be subject to

localized hot spots or temperature gradients that would exceed the eutectic temperature. A postirradiation examination of two of the specimens showed that the fuel material had penetrated the barrier. A third specimen, which had been irradiated at maximum jacket temperatures of 680°C, was intact. Measurements taken on this specimen failed to show any elongation, deformation, or diameter changes.

106-7002

Figure 10. U-20w/oPu-10w/oFs Alloy Jacketed with Inconel-X and Vanadium Barrier Foil after 1.6 a/o Burnup at Maximum Jacket Temperature of 695°C

3. Hastelloy-X Tungsten (Duplex Tubing)

Hastelloy-X was attractive in that reports of irradiations performed on the material indicated that it was not subject to excessive neutron irradiation embrittlement.⁽¹¹⁾ The material is also compatible with sodium at the reference reactor operating temperatures. Isolating the fuel material from the clad by a barrier, or operating the fuel at reduced temperatures, precluded the formation of eutectic.

Specimens were fabricated with a 0.05-mm (0.002-in.) layer of tungsten vapor-deposited on the inner surface of the tubing. Figure 11 is a macrophotograph of a section of tubing showing the layer of tungsten.

106-7001

Figure 11. Hastelloy-X Tubing with Tungsten-coated Inner Surface

Seven specimens were irradiated at maximum clad temperatures of 590°C and at fuel burnups of 3.0 a/o. Five of the seven specimens were recovered intact. Failures in the other two were characterized by small holes in the jacket where the fuel had made contact with the Hastelloy-X, evidently through defects in the tungsten coating. In the fabrication of this duplex jacketing, difficulties had been encountered in obtaining a uniform tungsten layer free of pinholes.

Figure 12 is a photograph of two typical irradiated specimens. The location of the break in the failed specimen is near the top of the fueled portion of the specimen, and not in the middle as is common with other jacketings. This supports the supposition that a defect in the tungsten barrier resulted in a eutectic formation.

Diameter and length measurement of the specimens jacketed in duplex tubing did not show any significant length changes and only minor diameter changes. The specimens were straight and maintained a cylindrical cross section.

F. Effect of Increasing the Void Volume

The first specimens assembled for this series of irradiations had the same percentage void volume as a full-length EBR-II fuel pin (i.e., ~13%).

EI-1608

EI-1277

Specimen No.
Jacket Comp., w/o
Burnup, a/o
Max. Jacket Temp., °C
Void Volume, %

29-3
Hast. X-W
3.0
490
57

28-2
Hast. X-W
2.5
590
57

Figure 12. Failures of Duplex Tubing Which Occurred When Fuel Material Penetrated the Barriers and Formed a Eutectic with the Outer Tubing

The fuel burnup that could be achieved without failure was usually less than 1.2 a/o. The jacket failures were violent and accompanied with the ejection of considerable amounts of fuel material. Nondestructive examinations of the irradiated specimens showed that the fuel did not elongate within the jacket. Calculations made on probable volumetric expansion of the sodium, accompanied with thermal expansion and swelling of the fuel, indicated that internal jacket pressures due to these factors alone could create enough pressure to exceed the rupture strength of the jacketing. If the void volume was increased to 28%, which is equivalent to what would exist if the fuel restrainer was removed from an EBR-II pin, the fuel rupture was not as severe, but failures were still occasionally experienced below 1.2 a/o burnup. As in the 13% void volume specimens, the fuel material did not elongate within the jacket. Only by increasing the void volume to a value greater than 50% was it possible to observe a tendency for the fuel to elongate and achieve higher burnups without jacket failure. With specimens having this large void volume, fuel burnups of 2.5 a/o were realized and an average fuel elongation of 10% was noted.

Venting of the gas void volume to the capsule environment also significantly extended the burnup capability of the specimens. The fuel elongated within the cladding as in the case of the specimens with large void volumes. However, venting did not yield results that surpassed those obtained for the large void volumes.

The jackets of four intact specimens were punctured, and the gases within the void volume above the fuel were collected for measurement and analyses. The compositions of the gas samples are listed in Table IV. The calculated internal pressures due to this gas are included in Table V. Though the specimens that were punctured for gas recovery were intact, companion specimens irradiated at comparable temperatures and atom percent burnups had failed. Jacket stresses resulting from the calculated internal pressures due to the gas were not significant compared to the assumed rupture strengths of the jacketing; nevertheless, the apparent reduction of this pressure, either by increasing the void volume or by venting

Table IV
ANALYZED GAS CONCENTRATIONS RECOVERED FROM VOID SPACE

Specimen No.	Gas Concentration, v/o													
	H ₂	He	H ₂ O	N ₂	O ₂	Ar*	Kr ⁸³	Kr ⁸⁴	Kr ⁸⁵	Kr ⁸⁶	Xe ¹³¹	Xe ¹³²	Xe ¹³⁴	Xe ¹³⁶
22-6	0.3	14.3	10.1	0.9	0.4	12.3	0.62	0.91	0.24	1.40	7.12	10.6	15.8	25.1
23-2	3.9	16.6	22.0	6.8	4.5	3.1	0.69	0.83	0.28	0.16	4.77	7.18	10.9	17.4
23-5	2.1	18.4	1.1	0.7	0.3	3.8	0.85	1.24	0.33	1.99	8.33	12.4	18.8	29.8
23-6	3.5	14.1	7.3	1.6	0.7	12.6	0.66	1.04	0.27	1.57	6.86	9.97	15.2	24.5

*Note: Argon was introduced into the specimen void space during welding of the top seal plug.

Table V
CALCULATED GAS PRESSURES WITHIN VOID VOLUME

Specimen No.	Burnup, a/o	Void Volume, %	Sodium Fill, g	Measured Gas Recovered, cc	Calculated Pressure of Gas at 500°C, kg/cm ²
22-6	0.7	50	0	0.42	0.73
23-2	1.4	50	0.1175	0.158	0.41
23-5	1.4	50	0.1185	0.85	2.19
23-6	1.4	50	0.1105	0.65	1.49

the jacketing, had a pronounced effect on extending the performance of the specimen. At present two possible explanations exist for this behavior: (a) there may have been an error in the volumetric determinations, and (b) the jacketing may have been near its rupture stress as a result of fuel swelling, so that the slight increment of stress resulting from the gas pressure was sufficient to cause the jacketing to fail.

G. Effect of Increasing the Jacket Thickness

Six specimens were irradiated which were jacketed in Nb-1w/oZr tubing having a wall thickness of 0.38 mm (0.015 in.) or 0.64 mm (0.025 in.). Two of the jackets were in the ductile-recrystallized condition, while the other four were in a stress-relieved condition. The specimens were irradiated at maximum jacket temperatures of 615°C for 4.2 a/o burnup. Surveillance of specimen integrity was maintained by periodic nondestructive examinations of the rods by neutron radiography.

When it was evident that one of the fuel rods was deformed, the capsule was disassembled, and the specimens (shown in Figure 13) were examined in detail. The failed jacket had a wall thickness of 0.38 mm (0.015 in.) and was in a stress-relieved condition. By comparison, a companion rod irradiated at the same temperature but having a jacket in the ductile fully recrystallized condition was intact. The failed jacket was characterized by longitudinal splits about the midportion of the fueled section of the specimens. The fuel material had exerted pressure against a segment of the cracked jacket producing a noticeable bulge. Metallographic examination of transverse sections of the defective jacket showed failures that were characteristic of brittle fracture. In addition, cracks in the inside diameter of the tubing similar to those shown in Figure 7 were observed 10 mm (0.40 in.) from the area where the jacket had split open.

At the date of writing, a similar group of specimens has been irradiated to greater than 7.2 a/o burnup at a fuel temperature near 625°C without indication of specimen failure.

Jacket Thickness, mm	0.38	0.38	0.64	0.64	0.64	0.38
Burnup, a/o	4.2	4.2	4.2	4.2	4.2	4.2
Max. Fuel Temp., °C	690	690	690	720	720	720
Max. Jacket Temp., °C	590	590	590	615	615	615

106-8044

Figure 13. Effect of Irradiation on U-20Pu-10Fs Alloy Jacketed with Nb-1w/oZr Alloy Having a Thickness of 0.38 mm (0.015 in.) and 0.64 mm (0.025 in.)

These results make it possible to establish that (1) the increase in jacket thickness substantially increased the attainable a/o burnup, and (2) the more ductile form of this alloy appeared superior to the alloy having initial high rupture strengths.

As previously mentioned, both 0.38-mm (0.015-in.) and 0.64-mm (0.025-in.) wall-thickness tubing was irradiated. It is realized that the latter tubing thickness may not be attractive for use in assembling production-type fuel rods. The intent of this application was to provide high-burnup specimens from which fission gas release measurements could be made. As of the date of this writing, these rods have not been sampled for fission gas.

H. Unbonded Fuel Specimens

Seven specimens were tested in which the usual sodium bond between the fuel and the clad had been purposely omitted. These specimens were presumably momentarily melted in their claddings during initial reactor startup. By the elimination of the sodium, the effects of pressure buildup due to the contribution of the sodium were not present. On the other hand, melting the fuel within the jacket eliminated the annulus between the fuel and jacket so that the fuel was in direct contact with the jacketing at the start of irradiation. This condition resulted in premature failure of the jacketing.

CONCLUSIONS

From the results of the irradiations performed to date on jacket-restrained U-20w/oPu-10w/oFs alloy, having the EBR-II cross-section reference dimensions and irradiated at the reference jacketing temperatures of approximately 600°C,⁽¹²⁾ it may be concluded that:

1. High rupture strength of the jacketing material is not the only important criterion of the restraint characteristics of the jacket during irradiation. The optimum relationship appears to be a jacketing that retains a high rupture strength with a high degree of ductility.
2. Jacket restraint of the fuel material in a radial direction results in an axial elongation of the fuel. At 2 a/o burnup, the elongation is on the order of 10% of the fuel length. Extending the operational range of the fuel rod requires adequate void volume to accommodate the displacement of the sodium, volumetric changes of the fuel, and released fission gas. If the jacket is vented, only the elongation of the fuel need be considered.
3. A promising jacketing material for this fuel is a duplex tubing consisting of Hastelloy-X with an internal layer of tungsten, vanadium, or other compatible metals.

4. Strict and exacting inspection of the jacketing stock and observance of the assembly procedures are important. Many premature failures of specimens could be attributed to defects in the jacket. As the program progressed, the improved quality of the tubing became apparent in the performance of the specimens and the greater consistency of the irradiation results.

5. Increasing the jacket thickness from 0.23 mm (0.009 in.) to 0.38 mm (0.015 in.) significantly increases the attainable burnup level of the specimen.

This report covers only the results of the irradiations performed on the specified fuel composition. The program is presently investigating other variables such as: (a) the effects of varying the plutonium concentration in the fuel, (b) the restraint characteristics of jackets having larger internal diameters, and (c) the investigation of additional jacketing materials.

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