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THE IRRADIATION BEHAVIOR OF HIGH-BURNUP URANIUM-PLUTONIUM ALLOY PROTOTYPE FUEL ELEMENTS

by

W. N. Beck, R. J. Fousek,
and J. H. Kittel

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SUMMARY

Alloys containing both uranium and plutonium are being developed as fuels for fast breeder reactors. The alloys selected for the study described in this report were U-Pu-Fs, U-Pu-Zr, and U-Pu-Ti. The irradiation behavior of the U-20 w/o Pu-10 w/o Fs alloy has been evaluated in the form of prototype EBR-II fuel elements and reported in Ref. 4. Additional work on this alloy and results obtained from studies on more advanced fuel alloys are reported here.

The irradiation studies were performed with cylindrical fuel pins sodium-bonded to tubular cladding. The cladding materials investigated included refractory alloys as well as iron- and nickel-base alloys. The investigations studied variations in the (1) plutonium concentration in the fuel pin, (2) cladding wall thickness, (3) irradiation temperature, (4) vented plenums, (5) type of cladding materials, and (6) type of fuel materials (solid fuel, disc-type fuel, and sodium-impregnated powder fuel). The investigations determined fission-gas release from the fuel as a factor of volume expansion, and maximum attainable fuel burnups.

Fuel-element design was the variable that resulted in the most significant improvement in the irradiation performance of the fuel; sufficient space for annular fuel swelling was allotted to accommodate solid fission products and to enable a significant fraction of the fission gas to be released from the fuel alloy. The most promising alloy tested for fast breeder reactors was the U-Pu-Zr composition, which contained approximately 15 w/o plutonium and 10-15 w/o zirconium. Successful irradiations were performed with this alloy at maximum cladding temperatures of 655°C and burnups of 12.5 a/o.

I. INTRODUCTION

Argonne National Laboratory is presently operating the Experimental Breeder Reactor No. 2 (EBR-II) at the National Reactor Testing Station in Idaho. The fuel discharged from the reactor is remotely reprocessed, refabricated into rods, and reassembled into subassemblies for return to the reactor.

The reactor is presently fueled with U-5 w/o Fs alloy. The term "Fs" refers to "fission," which is the collective designation of fission products that remain in the fuel after the pyrometallurgical reprocessing operation. These elements principally include molybdenum, technetium, and ruthenium, with lesser amounts of zirconium, rhodium, and palladium. The fission alloy first used in the reactor (before reprocessing) included all the mentioned elements except technetium, which would be present only after the fuel had been irradiated.

Future metallic cores for EBR-II and other fast breeder reactors may include alloys that contain plutonium. These fuels are of interest because they have the highest fertile-to-fissile atom density and thereby permit higher breeding ratios. The addition of plutonium to the U-Fs system, however, resulted in alloys characterized by relatively low melting temperatures (820-910°C).¹ Irradiation experiments on unclad specimens showed that the alloys swelled rapidly at temperatures $\geq 370^\circ\text{C}$.² In addition, a compatibility problem existed between the U-Pu-Fs alloy and the nickel- or iron-base alloy cladding materials at the desired operating temperatures.³ Apparently considerable reliance would have to be placed on cladding restraint to control fuel swelling, and the cladding would have to be made of refractory alloys that do not form low-melting eutectics with U-Pu-Fs alloy.

Screening irradiations were performed on clad U-20 w/o Pu-10 w/o Fs alloys.^{4,5} The prototype elements duplicated the cross-sectional dimensions and geometry of the standard EBR-II fuel pin, with the length, internal void space, and amount of sodium bond scaled down by a factor of seven. When the prototype elements were tested with various refractory-alloy tubing at cladding temperatures of 600°C, the maximum attainable burnups without cladding failure were near 4 a/o.

This report includes a detailed investigation of factors governing the irradiation behavior of plutonium-alloy fuel elements. The following areas of investigation were included:

1. Effect of composition variations on the irradiation behavior of U-Pu-Fs alloys.
2. Evaluation of duplex tubing* as cladding material.
3. Effect of operating temperature, cladding thickness, plenum volume, and effective density on maximum attainable burnup.
4. Relationship of fuel-element design to maximum attainable burnup before jacket failure.
5. Effects of enhanced fission-gas release from the fuel alloy.
6. Performance of improved high-melting U-Pu-Zr and U-Pu-Ti alloy systems.

*For this report, duplex tubing is referred to as a composite tube, the inner layer having been selected for its compatibility with the fuel, but not necessarily contributing to the burst strength of the tube.

The development of reprocessing methods that made possible the virtual removal of all fission products from uranium and plutonium alloy fuels⁶ permits consideration of other U-Pu alloys. The U-Pu-Ti and U-Pu-Zr alloys appeared the most promising of those considered. The solidus temperature of the two alloys was near 1150°C, and U-Pu-Zr alloys were found to be compatible with Type 304 stainless steel up to 800°C.⁷ Successful irradiations were performed with U-Pu-Zr alloys in both refractory alloy and iron- and nickel-base alloy tubing (see Section IV.B). Unprecedented burnups up to 12.5 a/o were achieved at maximum cladding temperatures of 655°C. The U-Pu-Ti alloy was irradiated in V-20 w/o Ti cladding to 10.7 a/o burnup at maximum cladding temperatures of 630°C.

II. DESCRIPTION OF PROTOTYPE ELEMENT

The initial configuration of the prototype elements was patterned after the EBR-II fuel element. This element was 18.187 in. long and

0.174 in. in diameter. The fuel pin was cast U-5 w/o Fs alloy, 14.22 in. long and 0.144 in. in diameter. The fuel pin was separated from the Type 304L stainless steel cladding by a 0.006-in.-thick sodium-filled annulus. The cladding has a nominal wall thickness of 0.009 in. and was sealed at both ends. A helium-filled plenum volume above the sodium will accommodate the thermal expansion of the sodium. The assembled plenum volume* is 14.9% of the fuel volume, but is reduced to 7.7% at operating temperature, primarily because of expansion of the sodium bond.

The effective (or smeared) density** of the fuel is ~85%. As previously mentioned, the lengths and void volumes of the specimens were scaled down by a factor of seven. Figure 1 is a drawing of a typical prototype element.

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Fig. 1. Typical Prototype Element Used in Irradiation Experiments

*The plenum volume expressed in "percent" refers to the ratio of the volume of the gas plenum to the fuel volume (at room temperature).

**Effective density is defined as the ratio of the fuel volume (assuming 100% of theoretical density) to internal cladding volume, neglecting end effects.

Before the specimens were assembled, the cladding was subjected to rupture tests and also nondestructively

examined by eddy-current point-probe techniques and X radiography.⁸ Defects greater than 10% of the wall thickness (i.e., inclusions, pits, or cracks) were considered a basis for rejection. Micrometric measurements were made of the cladding wall thicknesses.

The fuel pin was measured for length, diameter, and density. Source radiography was performed on specimens in which density determination indicated internal voids or porosity. Table I shows the identification and composition of the fuel pins.

TABLE I. Identification and Composition of Fuel Pins

Identification			Composition, w/o							
Pin No.	Alloy Designation, w/o	Melt No.	U	Pu	Mo	Ru	Pd	Rh	Zr	Ti
30-1	U-10Pu	495	88.87	11.13	-	-	-	-	-	-
30-2	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
30-3	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
30-4	U-15Pu	496	83.48	16.52	-	-	-	-	-	-
30-5	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
30-6	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
31-1	Uranium ↓	K-10	100	-	-	-	-	-	-	-
31-2		K-16	100	-	-	-	-	-	-	-
31-3		K-4	100	-	-	-	-	-	-	-
31-4		K-5	100	-	-	-	-	-	-	-
31-5		K-13	100	-	-	-	-	-	-	-
31-6		K-7	100	-	-	-	-	-	-	-
33-1	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
33-2	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
33-3	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
33-4	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
33-5	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
33-6	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
34-1	U-20Pu-10Fs	R-21	70.60	19.97	2.51	4.12	1.99	0.73	0.048	-
34-2	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
34-3	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
34-4	U-20Pu-10Fs	R-21	70.60	19.97	2.51	4.12	1.99	0.73	0.048	-
34-5	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.50	-
34-6	U-10Pu-10Fz	493	80.96	9.15	2.72	2.97	1.09	0.52	2.59	-
35-1	U-20Pu-10Fs	R-25	70.62	19.96	2.51	4.12	1.99	0.73	0.048	-
35-2	↓	R-25	↓	↓	↓	↓	↓	↓	↓	-
35-3		R-25	↓	↓	↓	↓	↓	↓	↓	-
35-4		R-25	↓	↓	↓	↓	↓	↓	↓	-
35-5		R-25	↓	↓	↓	↓	↓	↓	↓	-
35-6		R-25	↓	↓	↓	↓	↓	↓	↓	-
36-1	U-15Pu-10Fz	494	76.11	14.55	2.79	2.45	1.10	0.50	2.5	-
36-2	↓	494	↓	↓	↓	↓	↓	↓	↓	-
36-3		494	↓	↓	↓	↓	↓	↓	↓	-
36-4		494	↓	↓	↓	↓	↓	↓	↓	-
36-5		494	↓	↓	↓	↓	↓	↓	↓	-
36-6		494	↓	↓	↓	↓	↓	↓	↓	-
37-1		R-107	75.38	14.52	2.46	3.09	0.99	0.48	2.60	-
37-2	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
37-3		R-107	↓	↓	↓	↓	↓	↓	↓	-
37-4		R-107	↓	↓	↓	↓	↓	↓	↓	-

TABLE I (Contd.)

Identification			Composition, w/o							
Pin No.	Alloy Designation, w/o	Melt No.	U	Pu	Mo	Ru	Pd	Rh	Zr	Ti
37-5	U-15Pu-10Fz	R-107	75.38	14.52	2.46	3.09	0.99	0.48	2.60	-
37-6	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-1	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-2	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-3	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-4	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-5	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
38-6	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-1	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-2	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-3	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-4	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-5	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
39-6	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-1	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-2	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-3	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-4	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-5	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
40-6	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
41-1	↓	R-129	75.89	15.19	2.98	2.40	1.33	0.52	1.79	-
41-2	↓	R-129	↓	↓	↓	↓	↓	↓	↓	-
41-3	↓	R-129	↓	↓	↓	↓	↓	↓	↓	-
41-4	U-10Pu-10Fz	R-115	78.18	9.56	2.75	2.21	1.18	0.53	2.28	-
41-5	↓	R-115	↓	↓	↓	↓	↓	↓	↓	-
41-6	↓	R-115	↓	↓	↓	↓	↓	↓	↓	-
43-1	U-15Pu-10Fz	R-171	74.19	15.50	2.91	3.29	1.09	0.53	2.67	-
44-1	U-15Pu-10Ti	TM-41	75.25	15.40	-	-	-	-	-	9.54
45-1	U-18.5Pu-14.1Zr	TM-42	67.80	18.47	-	-	-	-	13.78	-
46-1	U-15Pu-10Fz	R-107	75.38	14.52	2.46	3.09	0.99	0.48	2.60	-
46-2	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
46-3	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
46-4	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
46-5	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
46-6	↓	R-107	↓	↓	↓	↓	↓	↓	↓	-
48-1	U-15Pu-12Zr	R-254	74.19	14.01	-	-	-	-	11.80	-
48-2	↓	R-254	↓	↓	-	-	-	-	↓	-
48-3	↓	R-254	↓	↓	-	-	-	-	↓	-
48-4	↓	R-254	↓	↓	-	-	-	-	↓	-
48-5	↓	R-254	↓	↓	-	-	-	-	↓	-
48-6	↓	R-254	↓	↓	-	-	-	-	↓	-

The specimens were assembled in a helium-filled glovebox with an oxygen concentration of less than 200 ppm. After assembly, the specimens were removed from the glovebox and the welds were checked for leaks with a helium mass spectrometer. The sodium bond was wetted to all contacting surfaces by centrifuging the specimens for 5 min at 350°C. This bonding technique had proved superior to the other methods previously attempted (i.e., impacting, high-frequency vibration, and spinning). The sodium bond was nondestructively inspected by an eddy-current point-probe technique.

Any specimen having an unbonded area or bubble in the sodium layer larger than 1/16 in. was not considered acceptable. The specimens were then X-radiographed to verify the proper positioning of the fuel pin and to determine the level of the sodium above the fuel. The volume of the specimen was measured by immersion, and length and diameter were measured. Table II shows the irradiation history of the capsules used for this fuel-evaluation program.

TABLE II. History of Capsule Irradiation

Capsule Number	Reactor Position	Date In Reactor	Date Out of Reactor	Exposure, MWd
CP-30	3 and 5	1-25-63	10-7-63	790
CP-31	10	3-11-63	5-13-63	240
CP-33	4	4-18-63	8-25-65	2584
CP-34	5 and 6	7-8-63	1-20-64	684
CP-35	14	9-30-63	7-14-64	724
CP-36	8	10-29-63	4-27-64	398
CP-37	15	11-14-63	9-3-64	573
CP-38	15	9-5-64	3-23-65	775
CP-39	15 and 8	6-8-64	2-9-65	664
CP-40	10	9-5-64	8-10-65	1269
CP-41	3 and 5	10-28-64	8-16-66	1892
CP-43	5 and 10	12-14-64	8-2-66	1440
CP-44	8	2-9-65	9-2-66	1768
CP-45	15	3-23-65	9-22-66	1596
CP-46	5	6-8-65	5-16-66	850
CP-48	6	1-26-66	5-16-66	405

III. 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 that flowed at 15 ft/sec through the 0.063-in.-thick annulus formed by the capsule and the thimble.⁹

The capsules were NaK-filled, double-wall containers, 1.5 in. in diameter and 21 in. long.¹⁰ Figure 2 is a diagram of the standard capsule. The specimens were arranged in two tiers, three specimens in each tier. They were held in place by the spacer discs, which restricted lateral as well as vertical motion, but allowed a 0.132-in. preferential radial displacement about the fuel portion of the specimen. These spacer discs also

106-7592

Fig. 2. Diagram of CP-5 Irradiation Capsule for Six Prototype Elements Arranged in Two Tiers of Three Each

restricted excessive NaK convection within the capsule. The arrangement of three specimens in one tier was advantageous in that three different specimens irradiated at comparable temperatures and burnups could be compared directly.

106-8709

Fig. 3. Diagram of CP-5 Irradiation Capsule Vessel for Single Prototype Element (Reduced 50% photographically)

The temperature of selected capsules was controlled during irradiation by electric resistance heaters, which encircled the two tiers of specimens. A maximum of 2 kW of auxiliary heating power was available. The auxiliary heat was used to compensate for normal neutron-flux variations in the reactor core. Temperature during irradiation could be controlled at $\pm 4^\circ\text{C}$ for extended periods of time.

Temperature was also controlled by using a flux-positioning capsule assembly.¹¹ The heat generation of the ele-

ments was controlled by positioning the capsule in a desired neutron-flux level. The assembly was operated by a remotely controlled extension tube. The capsule could be positioned at any elevation from the reactor midplane to 1 in. above the top of the core. With this equipment, the irradiation temperature of the fuel pins could be varied approximately 50°C . Average temperature fluctuation during reactor operation was $\pm 7^\circ\text{C}$. Figure 4 is a diagram of this capsule assembly.

The irradiation temperature of each specimen was continuously monitored by a central thermocouple or by a thermocouple located in the NaK immediately adjacent to the external surface of the cladding; thus maximum fuel and cladding temperatures could be determined. For specimens without a central thermocouple, central fuel temperatures were extrapolated, based on electric-analog plots¹² from adjacent thermocouple readings. The extrapolated temperatures were verified by direct measurements of fuel temperatures in specimens containing a central thermocouple.

The condition of specimens undergoing irradiation was determined by periodic removal of the capsule from the irradiation facility and non-destructive examination of the capsule contents by neutron radiography.¹³ This inspection provided information on fuel dimensions, possible cladding deformation, and proper placement of all components in the capsule. In most of the tests, a capsule was deliberately irradiated until the cladding of one or more of the specimens ruptured. Periodic neutron radiographs proved invaluable in establishing the sequence of cladding failure.

The capsule thermocouples were also used to indicate cladding failure. Any gross change in irradiation temperature as recorded by the thermocouples usually indicated that the adjacent specimen had deformed. The cladding generally failed at the portion of the specimen where the temperature was the highest. The thermocouples had been purposely positioned where previous irradiations performed on this type of fuel configuration had indicated the highest temperature. Consequently, the thermocouple usually detected the initial deformation of the fuel rod as a result of cladding failure.

The capsules were disassembled in shielded cells that contained a helium atmosphere with less than 200 ppm water and oxygen. After removal from the capsule, the two tiers of specimens were photographed to ensure that the orientation of the individual specimens with regard to the centerline of the capsule could be re-established. Wires containing cobalt were used as flux monitors and were located between specimens. The wires were removed, sectioned, dissolved, and analyzed radiochemically; the analytical results were used for a preliminary computation of the fuel burnup.

Each specimen was optically inspected, and each defect area was photographed. Length, diameter, and density of all intact specimens were measured. Ruptured specimens were measured for length, but diameter measurements were taken only on portions of the cladding that were still intact. Bowing of the specimen was determined from magnified photographs of the full-length rod. Fission-gas release was determined in some specimens by puncturing the cladding at a point above the fuel pin. The gas was analyzed mass-spectrographically for fission-product isotopes. Irradiated thermocouples were recalibrated within the shielded cell and found to be accurate within the calibration tolerances.

IV. RESULTS AND DISCUSSION

A. Investigation of Variables Affecting the Irradiation Behavior of Clad U-Pu-Fz Alloys1. Properties of U-Pu-Fz Alloys

A series of irradiations was previously conducted on U-20 w/o Pu-10 w/o Fz alloy.^{4,5} The irradiations described in this report are principally on U-15 w/o Pu-10 w/o Fz and U-10 w/o Pu-10 w/o Fz alloys. A distinction must be noted between the designation Fz and Fs; the Fs alloy contains <0.1 w/o Zr; the Fz alloy contains ~2.8 w/o Zr. The existence of the two different fissium alloys was due to anticipated variations in the EBR-II pyrometallurgical reprocessing cycle that would strongly influence the level of residual zirconium. Comparisons of irradiation results performed on both Fs and Fz alloys failed to show a significant difference that could be attributed to the presence of additional zirconium. Table III compares the properties of several U-Pu-Fz alloys.

TABLE III. Properties of Uranium-Plutonium-Fissium Alloys¹⁵

Alloy Composition, w/o	Uranium	U-10Pu	U-10Pu-10Fz	U-15Pu-10Fz	U-20Pu-10Fz	U-20Pu-5Fz	U-20Pu-15Fz
Solidus, °C	1132	1025	910	~865	820 ^a	860 ^a	805 ^a
Liquidus, °C	-	1060	1010	~1000	990 ^a	~1010	~960
Transformation Range, °C							
Start	668	585	550	520	510	510	500
End	775	740	630	605	605	700	550
Density at 25°C, g/cm ³	19.07	18.7	16.8	16.5	16.8	17.7	16.1
Thermal Expansion							
Avg Coef, 10 ⁻⁶ /°C							
20°C to start of trans.	19.4	19.3	16.3	16.5	17.5		
End of trans. to 720°C	25.6	23.8	20.5	22.5	21.7		
20-720°C	24.2	23.8	21.2	20.6	21.2		
Transformation, %	0.45	0.28	0.45	0.36	0.33		
Enthalpy (25°C to mp), cal/g	52	46	44 ^b	~43	~42	~43	~42
Thermal Conductivity, cal/(sec)(cm)(°C)							
20°C	0.071	0.056	0.046	~0.046	0.047	0.041	0.043
400°C	0.080	0.069	0.057	~0.056	0.056	0.054	0.053
600°C	0.094	0.087	0.070	~0.070	0.069	0.066	0.067
800°C	0.115	0.100	0.084	~0.085	0.086	0.077	0.080
Thermal Cycling							
% Change in Density		-30	+1	0	-1		
% Change in Length	-7	+25	0	-1	+2		
Hardness (DPN), kg/mm ² ^c							
Gamma-quenched	.d	.d	255		215		
Alpha-transformed	291	205	411	433	430		
Tensile Strength, kg/mm ²							
200°C	37	26	58	42			
400°C	17	19	40	27			
600°C	5	9 ^e	8	6	3		
800°C	1	0.5	2		1		
Brittleness,							
Extreme Fiber Stress, kg/mm ²							
Gamma-quenched		.d	136		116		
Alpha-transformed		101	83		51		
Strain at Fracture, %							
Gamma-quenched		.d	1.3		1.0		
Alpha-transformed		0.7	0.9		0.4		

^aThese values were obtained on the fissium composition (low zirconium).

^bFour cal/g were added to the observed value to approximate the heat of transformation.

^cConverted from Rockwell B and C scales by use of the Steel Hardness Conversion Chart.

^dGamma phase cannot be retained in either U or U-10 w/o Pu by quenching.

^eThe high strength is probably caused by beta phase.

The phase-transformation temperatures of the alloys are influenced by the concentration of plutonium and fissionium. The liquidus, solidus, and solid-state transformation¹⁵ temperatures are lowered as

plutonium and fissionium additions to the uranium are increased. Figure 5 is a graphic comparison of phase-transformation temperature of U-Pu-Fz alloys.

106-8078

Fig. 5. Transformation Temperatures in U-Pu-Fz Alloys.¹⁵ Upper shading shows interval between liquidus and solidus.

The U-15 w/o Pu-10 w/o Fz and U-10 w/o Pu-10 w/o Fz alloys thermally cycled through the transformation temperatures showed no appreciable volume change.¹⁶ Volume changes experienced during the irradiation would therefore be attributable to factors other than thermal-cycling effects.

Out-of-pile compatibility tests were conducted by Walter¹⁷ on diffusion couples of potential cladding materials and U-10 to 20 w/o Pu-10 w/o Fz alloys. The tests showed that only refractory-metal cladding could be used in contact with U-Pu-Fz alloys above 600°C. Alternatively, a refractory-metal-barrier layer could be used between the fuel alloy and iron- or nickel-base alloy cladding.

2. Design Parameters and Irradiation Results

A major objective of the experiments was to determine the effect of various parameters on the maximum achievable burnup before cladding failure. For this reason (as stated previously), most of the irradiations were deliberately continued until at least one specimen in each capsule showed indications of cladding failure.

a. Effect of Composition Variations on the Irradiation Behavior of U-Pu-Fz Alloys. Nine specimens of four different plutonium alloys were irradiated in Capsules CP-30 and CP-34. Table IV summarizes the irradiation conditions for these specimens. The cladding selected for these irradiations was Nb-1 w/o Zr having a wall thickness of 0.009 in. This cladding material was used in this test series and selected so that a correlation could be made with previous irradiations.⁴

The six specimens in Capsule CP-30 were irradiated at maximum cladding temperatures of 560°C. The first evidence of cladding failure occurred at 4 a/o burnup. An examination of the capsule revealed that the cladding of U-15 w/o Pu-10 w/o Fz alloy specimens (30-3, 30-5) had failed. The failures were longitudinal splits at the midsection of the

fuel pin from which a small amount of fuel protruded. The cladding failures in Nb-1 w/o Zr were characteristic of brittle-type fracture. No significant changes in cladding diameter were noted (i.e., less than 0.1%).

TABLE IV. Summary of Information for U-Pu, U-Pu-Fz Comparison Irradiations

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Comp. w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Condition of Specimens ^b
									a/o (U + Pu)	fiss/cc x 10 ^{-20a}		
30-1	U-10Pu	81.7	Nb-1Zr	0.174	0.009	58.1	525	600	2.6	13.1	50	A
30-4	U-15Pu	81.7	Nb-1Zr	0.174	0.009	56.4	525	600	4.1	17.6	37	C
30-2	U-10Pu-10Fz	82.8	Nb-1Zr	0.174	0.009	58.1	525	600	3.1	9.8	25	A
30-6	U-10Pu-10Fz	81.7	Nb-1Zr	0.174	0.009	58.4	525	600	3.2	10.0	30	A
34-6	U-10Pu-10Fz	82.9	Nb-1Zr	0.174	0.009	23.4	600	690	2.5	7.9	4	D
30-3	U-15Pu-10Fz	83.1	Nb-1Zr	0.174	0.009	57.4	560	640	4.5	14.3	22	B
30-5	U-15Pu-10Fz	83.4	Nb-1Zr	0.174	0.009	56.5	525	600	4.6	14.7	19	B
34-5	U-15Pu-10Fz	82.1	Nb-1Zr	0.174	0.009	24.5	635	730	3.2	10.0	7	B
34-4	U-20Pu-10Fs	86.5	Nb-1Zr	0.174	0.009	23.4	625	720	3.6	11.9	12	B

^aBased on effective density.

^bA = Specimen straight and intact; B = longitudinal split in cladding with protrusion of fuel; C = longitudinal split in cladding without protrusion of fuel; and D = several small cracks around center section of specimen.

The U-10 w/o Pu-10 w/o Fz alloy specimens (30-2, 30-6) were intact. The fuel-length change of these specimens appeared to result principally from localized axial expansion. The upper 15% of the pins showed an area of low density; in specimen 30-2 there was a noticeable necking of the fuel. There were no significant changes in outer dimensions of the clad specimens.

The U-15 w/o Pu alloy specimen (30-4) had a longitudinal hairline crack in the cladding located in the upper third section of the fueled portion of the specimen. No significant change was noted in diameter, length, or density. The U-10 w/o Pu alloy specimen (30-1), which was irradiated to a maximum 2.6 a/o burnup, was intact. The fuel had elongated 50% and had also segregated into three high-density areas joined by fuel of lower density.

Three additional specimens were irradiated in Capsule CP-34; one was a U-20 w/o Pu-10 w/o Fs alloy-fuel pin. The irradiation temperatures were higher (i.e., 635°C maximum cladding temperature), and the plenum volumes had been reduced from ~57 to ~24%. When the fuel was examined at a maximum burnup of 3.6 a/o, the cladding of the U-20 w/o Pu-10 w/o Fs and the U-15 w/o Pu-10 w/o Fz alloy had failed, but the U-10 w/o Pu-10 w/o Fz alloy pin was intact. The fuel-length changes were respectively 12, 7, and 4%. The Nb-1 w/o Zr cladding for these three specimens was in the fully annealed condition, but the observed failures were brittle-type fractures. No measurable changes were noted in specimen length, diameter, or density.

The irradiation of these specimens showed that the reduction of plutonium from 20 to 15 w/o did not result in significant increases in achievable burnup before cladding failure. The fuel with the 10 w/o plutonium had not reached the burnup values of the other alloys and could

not be compared directly. As anticipated, the increase in plutonium concentration resulted in an increase in swelling rate. Also, increasing the plenum volume, which reduced the final internal operating pressures, resulted in a substantially greater fuel volume. The binary alloys showed large volume increases and had a tendency to segment.

b. Evaluation of Duplex Tubing as Cladding Material. Above 650°C,¹⁷ the U-Pu-Fz system was not compatible with nickel- or iron-base alloys. However, these cladding materials are attractive because they are commercially available and relatively inexpensive. Some of the alloys had high strength characteristics at desired operating temperatures. Compatibility difficulties may possibly be overcome by using a thin refractory-metal barrier to separate the fuel from the cladding.

Twenty-one specimens, principally U-15 w/o Pu-10 w/o Fz alloy pins, were irradiated. The pins were clad in Type 304 stainless steel or Hastelloy-X, lined with tungsten or vanadium. Table V summarizes the information for these irradiations. The 0.0015-in.-thick tungsten lining was achieved by a vapor deposition process developed by a commercial vendor for this size tubing. Figure 6 is a transverse section showing this type of duplex tubing.

Difficulties had been encountered in obtaining duplex tubing that had a uniform-thickness layer of tungsten free of pinhole defects.¹⁸ Although the duplex tubing was closely inspected, cladding failures were experienced that were attributed to undetected flaws in the barrier layer.

TABLE V. Summary of Information for Specimens Irradiated in Duplex-type Tubing

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Condition of Specimens ^b
									a/o (U + Pu)	fiss/cc x 10 ^{-20a}		
34-3	U-10Pu-10Fz	88.4	304 SS-W Lined	0.175	0.012	22.1	575	660	2.2	7.5	7	F
37-1	U-15Pu-10Fz	85.1	304 SS-W Lined	0.175	0.010	Vented	660	780	2.3	7.5	12	C
37-2	U-15Pu-10Fz	82.7	304 SS-W Lined	0.175	0.010	Vented	660	780	2.3	7.3	12	A
37-3	U-15Pu-10Fz	83.9	304 SS-W Lined	0.175	0.010	19.8	630	750	2.3	7.4	10	F
37-4	U-15Pu-10Fz	83.9	304 SS-W Lined	0.175	0.010	22.4	590	680	2.3	7.4	9	B
37-5	U-15Pu-10Fz	83.9	304 SS-W Lined	0.175	0.010	22.2	590	680	2.3	7.4	8	B
37-6	U-15Pu-10Fz	83.9	304 SS-W Lined	0.175	0.010	Vented	590	680	2.3	7.4	15	A
34-2	U-15Pu-10Fz	83.9	304 SS-W Lined	0.175	0.011	22.9	590	680	2.5	8.0	14	E
34-1	U-20Pu-10Fs	86.1	304 SS-W Lined	0.175	0.010	22.9	590	680	2.8	9.2	17	E
38-1	U-15Pu-10Fz	82.8	304 SS-V Lined	0.186	0.015	49.3	580	660	3.2	10.3	23	A
38-2	U-15Pu-10Fz	82.8	304 SS-V Lined	0.186	0.015	Vented	580	660	3.2	10.3	17	A
38-4	U-15Pu-10Fz	74.2	304 SS-V Lined	0.196	0.015	54.0	630	710	3.2	9.1	13	E
38-5	U-15Pu-10Fz	74.2	304 SS-V Lined	0.196	0.015	Vented	630	710	3.2	9.1	13	A
36-1	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	23.0	645	750	2.2	7.5	11	F
36-2	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	Vented	645	750	2.2	7.5	10	A
36-3	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	21.0	645	750	2.2	7.5	10	E
36-4	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	22.0	600	695	2.3	7.8	9	A
36-5	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	22.0	600	695	2.3	7.8	10	A
36-6	U-15Pu-10Fz	88.5	Hast.-X-W Lined	0.174	0.011	20.0	635	740	2.3	7.8	15	E
38-3	U-15Pu-10Fz	73.2	Hast.-X-W Lined	0.184	0.009	Vented	580	660	3.2	8.9	18	A
38-6	U-15Pu-10Fz	73.2	Hast.-X-W Lined	0.184	0.009	58.0	630	710	3.2	8.9	15	F

^aBased on effective density.

^bA = Specimen straight and intact; B = longitudinal split in cladding with protrusion of fuel; C = longitudinal split in cladding without protrusion of fuel;

E = fuel penetrated barrier and formed extensive eutectic with cladding; F = fuel penetrated barrier and formed small hole in cladding.

Fig. 6. Hastelloy-X Tubing with Tungsten Coating

The vanadium-liner was a 0.0005-in.-thick foil encircling the fuel pin. The bottom plugs of both the vanadium- and the tungsten-lined specimens were separated from the fuel by 0.0625-in.-thick refractory-metal discs.

In addition to the evaluation of the effectiveness of the refractory-metal layers, the performance of the specimens was evaluated relative to the effect of (1) an increase in cladding thickness from 0.009 to 0.015 in., (2) a variation of effective density from 73 to 88.5%, and (3) a change in plenum-volume range from 19.8% to vented cladding. The cladding was vented by drilling a hole in the plenum above the sodium bond.

Nine of the 21 irradiated specimens (listed in Table V) were intact at the termination of irradiation. Of the nine, six had vented claddings, two had plenum volumes of 22%, and one had a plenum volume of 49.3%. The average fuel-length change was 15.7% for the vented specimens and >50% for those with plenum volumes. The specimens with smaller plenum volumes had an average fuel-length increase of 9.5%.

Of the 12 failed specimens, nine were directly attributable to failure of the barrier layer, as evidenced by eutectic formation, and three were due to fracture of the cladding. The average fuel-length increase was 11%. Variations in cladding thickness or effective fuel density did not noticeably affect the burnup level at which cladding failure occurred.

c. Effect of Operating Temperature and Increased Cladding Thickness on Maximum Attainable Burnup. Most of the capsule irradiations were with the standard EBR-II Mark I cladding-wall thickness of 0.009 in. Considerable difficulty was experienced in obtaining a thin-wall, defect-free tubing. The limit of defect detection by nondestructive testing was approximately 10% of the wall thickness, or 0.0009 in. Defects of this depth are

common in refractory metal tubing. Therefore, the performance of specimens with thicker cladding was evaluated to make the results of the experiments less dependent on the characteristics of the defects in the cladding. Furthermore, the penalty in breeding ratio incurred by increased amounts of structural material in a fast-breeder-reactor core could be compensated for by increases in fuel burnup.¹⁹

A group of U-15 w/o Pu-10 w/o Fz and U-10 w/o Pu-10 w/o Fz alloy pins were assembled to evaluate the effect of thicker cladding and the cladding-irradiation temperatures near 550°C with Nb-1 w/o Zr cladding having wall thicknesses of 0.015-0.025 in. Table VI summarizes the information for these irradiations. The pins were irradiated to burnups up to 8.7 a/o at maximum cladding temperatures of 545°C. The postirradiation examination showed all specimens to be intact except 33-3, which had a longitudinal split in the cladding near the center section of the fuel. Figure 7 is a photograph of the intact specimens. The only significant difference between the failed specimen (33-3) and an identical intact specimen (33-5) was a 6.6% smaller plenum volume in the failed specimen, which produced a higher internal cladding pressure in the later stages of irradiation.

Periodic neutron radiographs were taken of the six specimens in Capsule CP-33 to observe fuel-dimension changes and the condition of the cladding. The fuel-pin lengths reached a maximum within a range of 2-4 a/o burnup. This was followed by a gradual reduction in fuel length. Figure 8 is a plot of the fuel-pin lengths versus burnup.

An average of only 4.8% of the fission gas had been released into the gas plenum. The contraction of the fuel-pin length cannot be fully explained because this contraction is not entirely dependent upon the fission-gas release at the later stages of irradiation. Means of determining the final plenum volumes were not available because the plenum pressures could not be established. From information on the internal volume of the specimen, the fuel volume, and the sodium volume, calculations of plenum pressures resulting from thermal expansion of the sodium and swelling of the fuel were possible. Assuming that the sodium was displaced into the plenums when the fuel had reached maximum volume

TABLE VI. Summary of Information for U-Pu-Fz Alloy Irradiated to High Burnup

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Final Fuel-length Change, %	Fission-gas Release, %
									a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a		
33-1	U-10Pu-10Fz	81.6	Nb-1Zr	0.186	0.015	22.2	510	590	7.1	22.2	2	3.6
33-2	U-10Pu-10Fz	78.6	Nb-1Zr	0.199	0.020	23.2	520	610	7.1	21.4	1	9.6
33-6	U-10Pu-10Fz	79.7	Nb-1Zr	0.209	0.025	24.3	530	610	7.1	21.7	1	3.7
33-3	U-15Pu-10Fz	85.2	Nb-1Zr	0.186	0.015	23.3	545	625	8.7	28.4	0	-
33-5	U-15Pu-10Fz	84.0	Nb-1Zr	0.186	0.015	29.9	545	625	8.7	27.9	0	4.9
33-4	U-15Pu-10Fz	87.4	Nb-1Zr	0.209	0.025	23.9	545	625	8.7	29.1	0	-

^aBased on effective density.

Fig. 7. Appearance of Intact Specimens (Capsule CP-33) Irradiated to a Maximum 8.7 a/o Burnup

Fig. 8

Change in Fuel-pin Length vs.
a/o Burnup for a Group of U-Pu-
Fz Specimens Irradiated to High
Burnup

expansion, the resulting pressure in the plenums (>7500 psi) should have caused cladding failure at the early stages of irradiation. The most probable explanation is that part of the sodium bond had infiltrated the fuel through open pores created by the fuel swelling. Sodium was noted in the pores during preparation of metallographic sections.

A microscopic examination of transverse and longitudinal sections of the specimens showed no reaction between the fuel and the cladding. The fuel material was characterized by pores approximately $4\ \mu$ in diameter near the outer edge. The pores in the center of the pin (see Figs. 9 and 10) were approximately $15\ \mu$ in diameter.

In summary, these irradiations showed that fuel swelling can be suppressed by the application of high external pressures. The greatest fraction of the fission gas is retained in the fuel. Increasing the cladding thickness by as little as 0.005 in. approximately doubled the achievable burnup before cladding failure, but considerable reliance must be placed on the structural integrity of the cladding to withstand the high internal pressures.

d. Effect of Increasing Plenum Volume and Reducing Effective Density of Fuel. A theoretical analysis was made by Foote to determine the relationship between gas-bubble pressure in sodium-bonded metallic-fuel elements with fuel burnup, cladding dimensions, and mechanical properties.⁵ Factors that limit the attainable specimen burnup were included in the analysis; factors that extended the burnup were not included.

Fig. 9

Transverse Section of U-10 w/o Pu-10 w/o Fz Alloy (Specimen 33-1) Contained in Nb-1 w/o Zr Cladding and Irradiated to 7.1 a/o Burnup at Maximum Cladding Temperature of 510°C

Fig. 10. Transverse Section of U-10 w/o Pu-10 w/o Fz Alloy (Specimen 33-2) Contained in Nb-1 w/o Zr Cladding. The specimen was irradiated to 7.1 a/o burnup at a maximum cladding temperature of 510°C. The photograph at the left shows center porosity; the photograph at the right shows smaller pores in the fuel adjacent to the cladding.

An assumption was made that no fission gas escaped from the fuel and none was trapped in the lattice as isolated atoms; therefore, all fission gas appeared as free-gas bubbles and obeyed the ideal gas law. Two additional assumptions were made: The fuel had no mechanical strength, and the pressure of fission gas in the bubbles was equal to the pressure of gas in the void volume.

The fuel-volume increase due to accumulation of solid and gaseous fission products was expressed by

$$\frac{Y}{Bt} = 2.50 + 100 \cdot n \cdot R \left(\frac{T_f}{P} \right), \quad (1)$$

where

Y = swelling of the fuel, %;

Bt = burnup of total atoms, a/o;

2.50 = percent volume increase of fuel per 1 a/o burnup due to solid fission products;

n = fission product gas (Xe + Kr) per cm^3 of fuel per 1 a/o burnup, moles;

R = gas constant = 84.781 (pressure in kg/cm^2 ; volume in cm^3);

T_f = volume-averaged temperature of fuel = effective temperature of gas bubbles, $^{\circ}\text{K}$;

and

P = pressure of the fission gas in the bubbles, kg/cm^2 .

Equation (1) shows that for large burnup values the swelling will also be correspondingly large. If the swelling of the fuel can be restricted or limited to a practical value, the Y/Bt ratio will be diminished, and this would signify higher achievable burnup.

The amount of swelling could be limited by incorporating an axial restrainer inside the cladding, directly above the fuel pin.

The swelling of the fuel could then be expressed as

$$\frac{Y}{100} = \left(1 + \frac{Z}{100}\right) \left(\frac{B}{A}\right)^2 \left(\frac{D_j}{D_f}\right)^2 - 1, \quad (2)$$

where

Y = swelling of the fuel, %;

Z = axial swelling over and above the increase in length due to thermal expansion alone, %;

$A = 1 + \alpha_f(T_f - 300)$;

$B = 1 + \alpha_j(T_j - 300)$;

where α_f and α_j are the linear thermal-expansion coefficients of the fuel and cladding materials, $^{\circ}\text{C}^{-1}$,

and T_f and T_j are the effective average fuel and cladding temperatures, $^{\circ}\text{K}$;

D_j = cold inside diameter of cladding, cm;

and

D_f = cold diameter of fuel pin, cm.

The following assumptions are made: The operating temperature (T_f) is 1000°K , the fuel material has zero creep strength, and the fission-gas bubbles are of such size that surface-tension effects are small. Then,

$$Y = \left[\left(\frac{B}{A} \right)^3 - \left(\frac{P_o}{T_o} \right) \left(\frac{1}{A^3} \right) \left(\frac{T_v}{P} \right) \right] X - 100 \left(\frac{C - B^3}{A^3} \right) \left[\left(\frac{D_j}{D_f} \right)^2 \left(1 + \frac{L_n}{L_f} \right) - \left(\frac{C - A^3}{C - B^3} \right) \right], \quad (3)$$

where

$$A = 1 + \alpha_f(T_f - 300);$$

$$B = 1 + \alpha_j(T_j - 300);$$

$$C = 1 + \alpha_n(T_n - 300);$$

α_f, α_j = linear thermal-expansion coefficients of fuel and cladding materials;

α_n = average volume thermal-expansion coefficient of sodium from room temperature (300°K) to operating temperature (Tn);

Tf, Tj, Tn = volume-averaged temperatures of fuel, cladding, and sodium, respectively, °K;

Po, To = initial pressure and temperature of gas trapped in gas plenum, kg/cm² and °K, respectively;

Tv = temperature of gas in plenum at operating temperature, °K;

P = pressure of gas in plenum or pressure of fission gas in bubbles in fuel, kg/cm²;

X = volume of gas in plenum expressed as X% of initial fuel volume, %;

Dj, Df = cold inside diameter of cladding and diameter of fuel pin, respectively, cm;

and

Ln, Lf = thickness of sodium layer above the fuel pin and length of fuel, respectively, cm.

Thirteen irradiation experiments summarized in Table VII were conducted to test the results predicted by the above analytical approach. The specimens were to be irradiated until a cladding failure occurred. The fuel alloy selected was U-10 w/o Pu-10 w/o Fz and U-15 w/o Pu-10 w/o Fz contained in V-20 w/o Ti cladding. This alloy had shown promise as a cladding material because of its high strength, neutronic properties, and compatibility with the fuel. A piston-type restrainer was incorporated to limit the axial swelling of the fuel.

Six specimens (40-1 through 40-6) were assembled, all with preselected effective densities, plenum volumes, and allowable axial expansions. The analytical approach predicted that all six specimens should experience cladding failure at 3.1 a/o burnup. The first cladding failure (40-1)

was not observed until 5 a/o burnup. The specimens were further examined at 6.1 a/o burnup; specimens 40-2, 40-4, and 40-5 had longitudinal cracks in the cladding from which a small amount of fuel was protruding. Specimens 40-3 and 40-6, which had the lowest effective densities (74.0 and 71.6%, respectively), had hairline longitudinal cracks around the center section of the specimen.

TABLE VII. Summary of Information for Specimens Having Increased Plenum Volume and Reduced Effective Density of Fuel

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Condition of Specimens ^b
									a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a		
41-4	U-10Pu-10Fz	76.0	V-20Ti	0.197	0.016	41.1	630	740	8.2	23.9	0	B
41-5	U-10Pu-10Fz	79.9	V-20Ti	0.193	0.016	39.9	630	740	8.2	25.1	5	A
41-6	U-10Pu-10Fz	84.1	V-20Ti	0.189	0.016	36.7	630	740	8.2	26.4	10	B
40-1	U-15Pu-10Fz	82.9	V-20Ti	0.189	0.016	23.5	620	720	6.1	19.4	0	B
40-2	U-15Pu-10Fz	82.9	V-20Ti	0.189	0.016	23.6	620	720	6.1	19.4	1	B
40-3	U-15Pu-10Fz	74.0	V-20Ti	0.197	0.016	26.5	620	720	6.1	17.3	0	A
40-4	U-15Pu-10Fz	78.8	V-20Ti	0.193	0.016	25.0	620	720	6.1	18.4	-2	B
40-5	U-15Pu-10Fz	78.8	V-20Ti	0.193	0.016	25.0	620	720	6.1	18.4	1	B
40-6	U-15Pu-10Fz	71.6	V-20Ti	0.201	0.016	27.5	620	720	6.1	16.7	4	A
41-1	U-15Pu-10Fz	75.1	V-20Ti	0.197	0.016	41.1	630	740	7.3	21.0	-4	B
41-2	U-15Pu-10Fz	79.9	V-20Ti	0.193	0.016	39.9	630	740	7.3	22.4	5	A
41-3	U-15Pu-10Fz	86.3	V-20Ti	0.187	0.016	36.7	630	740	7.3	24.1	9	C
43-1	U-15Pu-10Fz	69.0	V-20Ti	0.203	0.015	56.0	600	770	12.0	31.7	3	B

^aBased on effective density.

^bA = Specimen straight and intact; B = longitudinal split in cladding with protrusion of fuel; C = longitudinal split in cladding without protrusion of fuel.

A second group of six specimens (41-1 through 41-6) was assembled with three different configurations preselected to fail at 5.9, 6.0, and 6.4 a/o burnup. No failures were noted until the specimens were examined at a maximum burnup of 8.2 a/o, at which time four had deformed cladding. The specimens most severely damaged were the two calculated to fail at 5.9 a/o burnup. These specimens had effective densities near 75% and incorporated a fuel restrainer that prevented any axial swelling of the fuel. Specimens 41-3 and 41-6 had longitudinal cracks in the cladding from which fuel protruded. These specimens were calculated to fail at 6.4 a/o burnup. The effective density was ~85%, and the fuel was restricted to an axial expansion of 5%. The two intact specimens were calculated to fail at 6.0 a/o burnup. The effective density of the fuel was ~80%, and the fuel was restricted to an axial expansion of 8.8%.

An irradiation was performed on a specimen (43-1) designed for cladding failure at 8.5 a/o burnup. The effective density was 69%, the plenum volume was 56% of the fuel volume, and the axial expansion was limited at 5% of the fuel length. Cladding failure did not occur until 12 a/o burnup.

The significant conclusions that can be drawn from the results of these 13 experiments are:

(1) Specimens achieved higher burnups without cladding failure than predicted by the analytical approach.

(2) Specimens that did not fail generally had a lower effective fuel density than those that failed, indicating that radial clearance is more effective than axial clearance in extending fuel burnup.

e. Effects of Enhanced Fission-gas Disengagement from the Fuel Alloy. Metal fuels are characterized by relatively low release of fission gas. For high-burnup fuels, where the forces exerted by the fuel on the cladding become excessively high, release of the gas from the fuel into the plenum above the fuel pin could be advantageous. Pressures in the gas bubbles in the fuel matrix would thereby be reduced, and, in the ideal case, only volume expansion from solid fission products would exert significant pressure at the interface between the cladding and the fuel.

Three methods of enhancing fission-gas release were investigated: (1) The fuel was subdivided to provide shorter paths of release and to provide more surface for fission-recoil escape, (2) an axial hole was provided for release of fission gas migrating to the center of the specimen, and (3) a larger annulus between the fuel and cladding was used to permit the fuel to swell to a volume where bubbles could interlink, releasing fission gas to the surface of the fuel.

(1) Subdivided Fuel. The first series of experiments in this category were performed to evaluate the swelling behavior of metallic fuels that had a preselected number of paths for fission-gas release. The axial and radial gas-release paths were attained by stacking 0.152-in.-diam circular foils that had 0.056-in.-diam axial holes. The foils were beta-treated unalloyed uranium (20% enriched), either 0.001 or 0.006 in. thick and stacked to provide a 1-in. pin length. For comparison, solid pins of fuel with axial holes and identical dimensions were also irradiated. The foils were contained in Nb-1 w/o Zr cladding of 0.175-in. OD and 0.009-in. wall thickness. Six specimens were assembled, two specimens of each fuel-pin configuration (0.001-in. foil, 0.006-in. foil, and solid pin). Three pins (one of each configuration) were sodium-bonded to the cladding, and the plenum chambers were punctured to permit release of the fission gas. The remaining three pins were assembled into cladding that had been partially cut away to serve as open cages for the fuel; thus swelling could occur without complete cladding restraint. Figure 11 is a photograph of the assembled specimens. The specimens were irradiated to a burnup of 0.7 a/o. Table VIII summarizes the information for these irradiations.

Postirradiation examination of the specimens showed that all were intact. Cladding-diameter measurements showed no significant change. The caged specimens showed varying degrees of swelling: minor in the 0.001-in. foils, pronounced in the 0.006-in. foils, and even more severe for the solid specimen. Enhanced fission-gas release from the 0.001-in.-thick foils appears to prevent the swelling evidenced in the other two specimens. When the surface condition of the stacks of foils was examined, the individual 0.001-in.-thick foils were visible, but the 0.006-in.-thick foils were not readily identifiable.

EI-1084

2X

Fig. 11. Preirradiation Photograph of Specimens 31-1 through 31-6

TABLE VIII. Summary of Information for Fuel Specimens Having Selected Fission-gas Release Paths

Spec. No.	Fuel Composition, w/o	Foil Thickness, in.	Cladding Composition, w/o	Cladding OD. in.	Type of Restraint	Max Fuel Surface Temp. °C	Max Fuel Temp. °C	Burnup, a/o	Diameter Change, %
31-1	Uranium	0.001	Nb-1Zr	0.175	Clad	560	590	0.7	3
31-2	Uranium	Solid Pin	Nb-1Zr	0.174	Clad	560	590	0.7	3
31-3	Uranium	0.006	Nb-1Zr	0.175	Clad	560	590	0.7	3
31-4	Uranium	0.001			Caged	560	590	0.7	22
31-5	Uranium	Solid Pin			Caged	560	590	0.7	89
31-6	Uranium	0.006			Caged	560	590	0.7	83

Metallographic examination showed that the axial hole in each specimen, whether solid fuel or stacked foils, had either disappeared completely or was only identifiable by a small discontinuous central void. The foils were sintered, and it was difficult to establish the original interfaces. The only evidence of the original interface was the presence of rows of elongated pores, separated by an axial distance that coincided with the original thickness of the foil.

The object of the second series of experiments was to enhance the rate of fission-gas release by recoil. The six specimens

irradiated in these experiments consisted of particles of U-15 w/o Pu-10 w/o Fz alloy vibratorily compacted in Nb-1 w/o Zr cladding. The mesh size of the particles was -60 +120, -120 +200, and -200 +325.

Although the compacts were sodium-impregnated to improve the heat-transfer characteristics, the sodium-bonding operation was not completely successful. Nondestructive examination of the compacts before irradiation revealed isolated areas where the sodium had not completely infiltrated the fuel particles. Table IX is a summary of information for these specimens.

TABLE IX. Summary of Information for Specimens Fueled with Vibratorily Compacted Powders

Spec. No.	Powder Mesh Size	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %
								a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a	
46-1	-60 +120	57	Nb-1Zr	0.174	0.009	560	650	3.8	8.3	-10
46-2	-200 +325	58	Nb-1Zr	0.174	0.009	560	650	3.8	8.4	-11
46-3	-200 +325	59	Nb-1Zr	0.174	0.009	560	650	3.8	8.5	-7
46-4	-200 +325	58	Nb-1Zr	0.174	0.009	600	690	3.8	8.4	-9
46-5	-120 +200	61	Nb-1Zr	0.174	0.009	600	690	3.8	8.8	-4
46-6	-120 +200	59	Nb-1Zr	0.174	0.009	600	690	3.8	8.4	-7

^aBased on effective density.

One specimen (46-1) was instrumented with a central thermocouple, to continuously monitor center fuel temperatures of the compact and to verify the calculated temperature drop across the fuel. Periodic neutron radiographs of the capsule showed that the compacts were undergoing sintering and were subdividing into two or three sections.

The capsule was opened after the specimens had reached a calculated 3.8 a/o burnup. Four of the six specimens had several small longitudinal cracks from which the fission gas had escaped. Although the other two specimens appeared to be intact, no fission gas was recovered when the plenums were punctured, indicating that these also had undetected defects in the cladding.

These specimens will be examined metallographically, to assist in determining the cause of cladding failure.

(2) Irradiation of Fuel Pins Having Axial Holes. A fuel pin subjected to reactor operating conditions is in a steep radial temperature gradient, on the order of 50°C/mm. Under these conditions, fission-gas-bubble migration is toward the center of the fuel pin.²⁰ An axial void introduced in the fuel could facilitate the release of fission gas to the plenum above the specimen.

Six specimens were prepared to evaluate this concept (summarized in Table X). The fuel pins, fabricated of U-20 w/o Pu-10 w/o Fs alloy, were 0.144 in. in diameter and had axial holes 0.055 in. in diameter. One of these specimens (35-5) differed from the rest in that a

thermocouple was placed in the axial hole to measure central temperatures. The pins were contained in Nb-1 w/o Zr cladding (0.009-in.-thick walls). Two specimens (35-3 and 35-6) had solid fuel pins 0.132 in. in diameter, instead of the reference 0.144 in. The claddings of these two specimens were vented to the capsule plenum. The diameter of the solid fuel was purposely reduced to make the fuel volume equal that of a pin with an axial hole. Thus, two fuel configurations that represented the same effective density were compared, one having a central void and the other an annular void.

TABLE X. Summary of Information for Specimens Having Axial Holes

Spec. No.	Fuel Configuration	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Condition of Specimens ^b
									a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a		
35-1	Axial hole	71.5	Nb-1Zr	0.174	0.009	27	635	740	4.3	11.8	-6	B
35-2	Axial hole	72.0	Nb-1Zr	0.174	0.009	27	635	740	4.3	11.9	-6	B
35-4	Axial hole	72.1	Nb-1Zr	0.174	0.009	27	615	720	4.5	12.4	-3	B
35-5	Axial hole	72.0	Nb-1Zr	0.174	0.009	25	615	720	4.5	12.3	9 ^c	C
35-3	Solid fuel	69.8	Nb-1Zr	0.174	0.009	Vented	635	740	4.3	11.5	10	D
35-6	Solid fuel	70.2	Nb-1Zr	0.174	0.009	Vented	615	720	4.5	12.1	1	A

^aBased on effective density.

^bA = Specimen straight and intact; B = longitudinal split in cladding with protrusion of fuel; C = longitudinal split in cladding without protrusion of fuel; and

D = several small cracks around center section of specimen.

^cThermocouple in axial hole.

Periodic neutron radiographs of the capsule permitted observations on the swelling behavior of both fuel configurations. All pins having the axial hole showed a gradual reduction in length, which at the end of the irradiation averaged ~5%. The axial holes slowly disappeared, closing initially at the midsection of the fuel and eventually progressing to both ends until the hole was no longer visible on the radiographs. The two solid pins (smaller diameter in vented cladding) gradually elongated, reaching a final average fuel-length increase of 5%.

The specimens were examined at a maximum of 4.5 a/o burnup. All specimens had damaged cladding except 35-6, which was one of the solid pins in a vented cladding.

Metallographic examinations showed that all pins had filled the initial annular clearance between the fuel and the cladding. The axial holes were also closed except near the top end of the pins. There was evidence of intergranular corrosion of the cladding, which evidently resulted in premature failure of the specimens.

The tests showed no apparent advantage in introducing an axial hole in the fuel, as compared to an increased annular void. These tests did show that decreasing the effective density of the fuel increased the achievable burnup of the fuel by a factor of 2 (as compared with previous results conducted with Nb-1 w/o Zr cladding, described in Ref. 4).

(3) Irradiation of Specimens Having Preselected Volume Limitations. This experiment was designed to determine the fission-gas release from metallic fuel elements under conditions where the fuel was

permitted to swell within preselected volume limitations. The experiment also evaluated the comparative restraint capability of the cladding where there was a variable annular clearance between the fuel and the cladding.

The fuel was U-15 w/o Pu-10 w/o Fz alloy pins contained in Nb-1 w/o Zr cladding. The tubing ID of the six claddings varied from 0.160 to 0.216 in. Axial fuel swelling was prevented by restrainers attached to the top seal-plugs. Table XI summarizes the information for these specimens.

TABLE XI. Summary of Information for Specimens Having Controlled Volume Limitations

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Fission-gas Release, %
									a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a		
39-1	U-15Pu-10Fz	79.6	Nb-1Zr	0.202	0.021	28	630	740	2.7	8.2	1	b
39-2	U-15Pu-10Fz	74.1	Nb-1Zr	0.204	0.019	39	630	740	2.7	7.7	0	33
39-3	U-15Pu-10Fz	67.8	Nb-1Zr	0.209	0.018	74	630	740	2.7	7.0	-3	-
39-4	U-15Pu-10Fz	61.1	Nb-1Zr	0.220	0.019	89	630	740	3.0	7.0	-7	63
39-5	U-15Pu-10Fz	52.4	Nb-1Zr	0.237	0.021	124	630	740	3.0	6.1	1.3	76
39-6	U-15Pu-10Fz	44.2	Nb-1Zr	0.260	0.022	168	630	740	3.0	5.1	-2	83

^aBased on effective density.

^bLongitudinal split in cladding.

The volumes of the NaK bond* and the upper gas plenum were calculated so that at operating temperature the plenum pressure of each specimen would be at 10 atm whenever the fuel had increased in volume sufficiently to make complete contact with the cladding.

The specimens were irradiated to 3.0 a/o burnup before the capsule was destructively examined. The postirradiation examination showed that all specimens were intact except 39-1, which had a longitudinal split in the cladding. This specimen had been allowed a radial fuel-volume expansion of only 26%. All the other specimens had allowable radial-volume expansions of 30% or greater and were intact. Transverse and longitudinal sectioning of the specimens showed that all fuel pins had swelled so that they were in intimate contact with the cladding. The fuel-length change varied from 1.3% for specimen 39-5 (effective density, 52.4%) to -7.4% for specimen 39-4 (effective density, 61.1%).

The most important results were the fission-gas recovery data; namely, specimens that had shown volume increases in excess of 30% released significant amounts of fission gas. These results supported the theoretical model by Barnes,²¹ which indicated that interconnection of bubbles should occur after fuel volume increases of $33\frac{1}{3}\%$. The interlinked bubbles provide paths for fission-gas release to the surface of the fuel. Examination of polished sections of the fuel at 500X magnification showed evidence of bubble interconnection, as shown in Fig. 12. Figure 13 shows the effect of fuel-volume increase on fission-gas release and shows results from U-Fs and U-Pu-Zr alloys (Section IV.B.2 of this report).

*The heat-transfer medium for these specimens was changed from sodium to NaK to facilitate assembly and loading of the specimens.

EI-5767

250X

Fig. 12. Interconnected Porosity in Metal Fuel Having a Measured Volume Expansion of 69%

106-9681

Fig. 13. Fission-gas Release vs. Fuel-volume Increase

The results from this experiment support the view that fission-gas release from metal fuels is largely independent of burn-up and irradiation temperature, but depends principally on the amount of fuel swelling. The data points in Fig. 13 represent burnups of 2.7-12.5 a/o and maximum fuel temperatures of 450-840°C.

B. Performance of Improved High-melting U-Pu-Zr and U-Pu-Ti Alloy Systems

The development of fuel-reprocessing schemes for the remote removal of all fission products from uranium and plutonium could result in other uranium-plutonium alloys being considered for fast breeder reactors. Alloying elements investigated at Argonne National Laboratory included Se, Ti, Y, Zr, Nb, Mo, Pd, and Th. Of these, the elements that were the most effective

in raising the solidus temperature of the U-Pu alloy systems were zirconium and titanium. The resultant two alloys (U-Pu-Zr and U-Pu-Ti) also happened to be the most desirable from reactor-physics considerations.

1. Properties of U-Pu-Zr and U-Pu-Ti Alloys

Studies by Kelman and his colleagues²² on the properties of U-Pu-Zr and U-Pu-Ti alloys disclosed that U-Pu-Zr alloys of interest were

106-8519

Fig. 14. Melting Temperature of Reaction Products of Type 304 Stainless Steel with Several Fuels

compatible with Type 304 stainless steel up to 800°C, whereas U-Pu-Ti interacted readily at 650°C. The use of austenitic stainless steel as a cladding material is attractive for sodium-cooled fast reactors because of neutronic properties, compatibility with flowing sodium, and commercial availability as small-size tubing. Figure 14 (Ref. 23) compares the melting temperatures of the reaction products of Type 304 stainless steel and U-Pu-Zr alloys and includes similar information on other alloys.

Repeated thermal cycling of U-Pu-Zr and U-Pu-Ti alloys through their phase changes showed no significant effects.²⁴ Length-change increases varied from 0 to 5%, but the errors in measuring slightly distorted rods were believed to be the cause of the length increase. The density change, which varied from -0.6 to +0.8%, was probably due to homogenization of the cast structure rather than thermal-cycling damage.

The U-Pu-Zr and U-Pu-Ti alloy systems were similar in that both contained appreciable amounts of alpha-uranium and zeta-uranium-plutonium phases at room temperature, both showed extensive solubility for the alloy addition, and both will first form a body-centered-cubic gamma-solid solution on solidification.²⁵ Although initially both alloy systems were studied extensively, emphasis was eventually placed on the U-Pu-Zr alloy because of its superior compatibility characteristics. Table XII lists the pertinent properties of the U-Pu-Zr and U-Pu-Ti alloys investigated.

Phase-transformation studies by O'Boyle²⁶ on the U-Pu-Zr alloy system having nominal concentrations of 15 w/o Pu and 10-14 w/o Zr showed that the alloy contains alpha-uranium, zeta-uranium-plutonium, and delta-UZr₂ up to 595°C. Between 595 and 655°C, the fuel contains alpha-uranium, zeta-uranium-plutonium, and gamma. Above 655°C, the fuel is in the gamma phase. Dilatometry tests on U-Pu-Zr alloys showed that these transformations proceeded rapidly. In the performance of irradiations

on fuel pins of the alloy, where the surface of the fuel might be below 595°C and the center of the fuel above 655°C, the three different phase fields would exist. The central zone would be in the gamma phase, surrounded by two concentric zones of the lower temperature phases.

TABLE XII. Properties of U-Pu-Zr and U-Pu-Ti Alloys²³

Alloy Composition	U		U		U		U		U		U	
	Pu	Zr	Pu	Zr	Pu	Zr	Pu	Zr	Pu	Ti	Pu	Ti
Nominal Composition												
w/o	11.1	6.3	15	10	18.4	14.1	17.1	3.4	15	6.5	15	10
a/o	10	15	12.9	22.5	15	30	15	15	11.9	25.7	10.7	35.6
Approx Liquidus Temp, °C	1200		1250		1290		1200		1300		1340	
Approx Solidus Temp, °C	1120		1155		1170		1075		1160		1200	
Solid Transformation												
Temp Range, °C	595-680		595-665		595-660		570-780		570-850		680-850	
Density (at 25°C As-cast), g/cm ³	17.0		16.0		15.1		17.0		15.6		14.4	
Thermal Expansion												
Avg Coef x 10 ⁶ , °C ⁻¹												
25°C to first trans.	18.3		17.6		17.5		21.2		18.9		19.2	
End of trans. to 950°C	18.1		20.1		20.0		16.4 ^a		20.0		19.2	
In Trans. Range $\Delta l/l$, x 10 ³	5.1		5.2		5.0		9.3		17		13.7	
Thermal Cycling												
Density Change, %	0		-0.1		0		+0.1		-		-	
Hardness (at 25°C As-cast), DPH	470		540		410		430		-		400	
Thermal Conductivity, W/(cm)(°C)												
200°C	-		0.12		0.11		0.13		0.13		0.12	
400°C	-		0.18		0.17		0.20		0.17		0.16	
600°C	-		0.22		0.21		0.25		0.21		0.20	
800°C	-		0.25		0.24		0.30		0.24		0.23	
Tensile Strength, kg/mm ²												
25°C	18.1		6.7		7.7		-		4.5		6.5	
675°C	1.2		2.1		2.9		3.6		2.6		7	
Compressive Strength, kg/mm ²												
25°C	164		129		116		96		130		90	
675°C	4		5.6		5.6		3.8		6.3		10	

^aTo 860°C.

Preirradiation metallographic examination of the structure of the U-Pu-Zr and U-Pu-Ti alloys were performed by Lukasik and Savage.²⁷ Samples were taken from both the top and bottom of the injection castings. The gamma grain size and the laminar spacing were finer at the top of the casting, which had a more rapid cooling rate; however, homogenizing the alloy for 1 week at 950°C provided a more uniform structure.

The irradiated U-Pu-Zr alloys had a plutonium concentration of 15-18.5 w/o and zirconium additions of 12-14 w/o. Within the range of compositions, the alloys were primarily in an $\alpha(\text{U,Pu})\text{Zr}_2 + \alpha\text{-U} + \zeta(\text{U,Pu})$ three-phase region. The electrolytically polished and etched surfaces showed oxygen-stabilized alpha-zirconium particles that were cored in the as-cast structure but became spheroidized in the homogenized structure. Figure 15 is a photomicrograph of a homogenized injection casting showing the alpha-zirconium particles.

The U-15 w/o Pu-10 w/o Ti alloy, the only composition irradiated, is primarily in a two-phase region, $(\text{U,Pu})_2\text{Ti}$ and oxygen-stabilized alpha-titanium. Figure 16 is a photomicrograph of the homogenized structure.

42326

500X

Fig. 15. Injection-cast U-18.5 w/o Pu-14.1 w/o Zr Alloy Homogenized at 950°C for One Week and Oil Quenched. The light particles are oxygen-stabilized alpha-zirconium.

42254

500X

Fig. 16. Injection-cast U-15 w/o Pu-10 w/o Ti Alloy Homogenized at 950°C for One Week and Oil Quenched. The light particles are oxygen-stabilized alpha-titanium.

2. Irradiation Results on U-Pu-Zr Alloy Specimens

The first irradiation of a U-Pu-Zr alloy was on six U-15 w/o Pu-12 w/o Zr fuel-alloy specimens; two specimens were clad in Type 304 stainless steel, two in Type 316 stainless steel, and two in Hastelloy-X. The specimens are described in Table XIII. The effective density of the fuel was 75%, and the plenum volumes were an average of 96% of the fuel volume.

TABLE XIII. Summary of Information for U-Pu-Zr and U-Pu-Ti Alloy Prototype Elements

Spec. No.	Fuel Composition, w/o	Effective Density, %	Cladding Composition, w/o	Cladding OD, in.	Cladding Thickness, in.	Plenum Volume, %	Max Cladding Temp, °C	Max Fuel Temp, °C	Burnup		Fuel-length Change, %	Fission-gas Release, %	Condition of Specimens ^b
									a/o (U + Pu)	fiss/cc x 10 ⁻²⁰ ^a			
48-2	U-15Pu-12Zr	75	304 SS	0.196	0.015	96	610	700	2.4	6.2	13	60	A
48-5	U-15Pu-12Zr	75	304 SS	0.196	0.015	92	-	-	2.4	6.2	-	-	B
48-3	U-15Pu-12Zr	75	316 SS	0.196	0.015	97	610	700	2.4	6.2	12	53	A
48-6	U-15Pu-12Zr	75	316 SS	0.196	0.015	97	-	-	2.4	6.2	-	-	B
48-1	U-15Pu-12Zr	75	Hast.-X	0.196	0.015	95	610	700	2.4	6.2	13	52	A
48-4	U-15Pu-12Zr	75	Hast.-X	0.196	0.015	98	-	-	2.4	6.2	-	-	B
45-1	U-18.5Pu-14.1Zr	63	V-20Ti	0.208	0.016	73	655	840	12.5	28.0	3	72	A
44-1	U-15Pu-10Ti	69	V-20Ti	0.203	0.015	64	630	810	10.7	25.0	14	-	A

^aBased on effective density.

^bA = Specimen intact and straight; and B = fuel formed eutectic with cladding.

When the specimens were first brought to temperature in the reactor, thermocouples on the three lower tier specimens showed unusual temperature fluctuations. When the specimens were examined at 2.4 a/o burnup, the fuel had penetrated the cladding of the three specimens on the lower tier. The most probable explanation for these failures is inadequate sodium bonding, resulting in a localized overtemperature condition.

The metallographic examination of the three intact upper-tier specimens, which operated at maximum cladding temperatures of 610°C, showed excellent compatibility between the fuel and the cladding. Figure 17 shows photomicrographs of these interfaces. The transverse sections of the fuel showed three distinct bands believed to be associated with the phase-transformation temperatures discussed previously (see Fig. 18). The outer zone is believed to correspond to the alpha-uranium + delta + zeta phase, the second band was alpha-uranium + zeta + gamma phase, and the central area was principally the gamma phase. As anticipated, the fuel had swelled to the internal diameter of the cladding, and the sodium bond was entirely displaced into the gas plenum. Figure 18 also shows that the original void volume, which had existed as a sodium-filled annulus around the solid-metal fuel pin at the start of irradiation, became redistributed throughout the fuel volume in the form of finely divided porosity.

A high-burnup experiment was performed on a U-18.5 w/o Pu-14.1 w/o Zr alloy pin contained in V-20 w/o Ti cladding. The effective density of the fuel was 63%, and the plenum volume was 73% of the fuel volume.

Fig. 17. Fuel-cladding Interface of U-15 w/o Pu-12 w/o Zr and Hastelloy-X, Type 304 Stainless Steel, and Type 316 Stainless Steel Irradiated at Maximum Cladding Temperature of 610°C for 2.4 a/o Burnup

Fig. 18

Transverse Section of U-15 w/o Pu-12 w/o Zr Alloy (Specimen 48-1) Clad in Hastelloy-X. The concentric bands in the fuel were correlated to phase-transformation temperatures (16X).

This intact specimen achieved an analyzed 12.5 a/o burnup (28×10^{20} fiss/cm³) at a maximum cladding temperature of 655°C. As far as is known, this is the highest fission density to be achieved without cladding failure in a metal-fuel specimen designed as a prototype fuel element. The irradiation was discontinued after it was certain that the specimen had exceeded 10 a/o burnup. Figure 19 is a photograph of the specimen after irradiation. Periodic nondestructive examination of the capsule during irradiation showed that the pin attained (at 4.2 a/o burnup) a maximum elongation of 3%. **This dimension did not change throughout the remainder of the irradiation.**

EI-5629

Fig. 19. U-18.5 w/o Pu-14.1 w/o Zr Alloy (Specimen 45-1) Contained in V-20 w/o Ti Alloy Cladding after Irradiation to 12.5 a/o Burnup at a Maximum Cladding Temperature of 655°C. The fission density is 28×10^{20} fiss/cc.

Diameter changes in the clad specimen were calculated from volume changes measured by immersion. An average increase of 0.0002 in. in diameter was observed. **No measurable length increase was noted.** The cladding was punctured, and the recovered fission gas was analyzed for fission-product isotopes. It was determined that 74% of the fission gas had been released to the plenum.

The metallographic examination showed no significant penetration of the fuel into the cladding. Figure 20 is a transverse section of the fuel pin. As for the U-Pu-Zr specimens described previously, the concentric

Fig. 20

Transverse Section of U-18.5 w/o Pu-14.1 w/o Zr Alloy (Specimen 45-1) Contained in V-20 w/o Ti Cladding and Irradiated to 12.5 a/o Burnup at a Maximum Cladding Temperature of 655°C

zones in the fuel are believed to be associated with the phase-transformation temperatures in the alloy. Figure 21 shows photographs of the fuel-cladding interface, as well as typical center-fuel porosity. Figure 22 shows typical postirradiation structures of the U-Pu-Zr alloy.

The condition of the fuel and the calculated final operating gas pressure of the plenum (~2200 psi) indicate that the specimen could have attained higher burnups without cladding failure.

Additional successful irradiations have been conducted on 16 full-length fuel elements of U-Pu-Zr alloy in the EBR-II reactor.²⁸ The pins were clad in either Type 304 stainless steel, Type 316 stainless steel, Hastelloy-X, or Hastelloy-X-280. Burnups of 4.6 a/o were attained without cladding failure or distortion at maximum cladding temperatures of 630°C.

Fig. 21. U-18.5 w/o Pu-14.1 w/o Zr Alloy (Specimen 45-1) Contained in V-20 w/o Ti Cladding and Irradiated to 12.5 a/o Burnup at Maximum Cladding Temperature of 655°C. The photograph at left shows the fuel-cladding interface; the photograph at right shows the size and distribution of center-fuel porosity.

EI-5442

250X

Fig. 22. Typical Porosity (dark areas) in the Center-fuel Section of Irradiated U-Pu-Zr Alloy (Specimen 48-1). The light particles are analogous to the alpha-zirconium phase observed in the preirradiated structure.

3. Irradiation Results of U-Pu-Ti Alloys

The U-Pu-Ti alloy, as mentioned previously, was regarded as secondary in importance, and, as such, only one irradiation was performed on a U-15 w/o Pu-10 w/o Ti pin contained in V-20 w/o Ti cladding. Table XIII includes a summary of information on this specimen.

The specimen reached an analyzed 10.7 a/o burnup (25×10^{20} fiss/cm³) at a maximum cladding temperature of 630°C. Figure 23 is a photograph of the intact specimen after irradiation. Periodic nondestructive examination of the capsule by neutron radiography showed a maximum fuel-pin elongation of 14% at 4.6 a/o burnup. This dimension remained the same throughout the rest of the irradiation.

EI-5628

3/4X

Fig. 23. U-15 w/o Pu-10 w/o Ti Alloy (Specimen 44-1) Contained in V-20 w/o Ti Alloy Cladding after Irradiation to 10.7 a/o Burnup at a Maximum Cladding Temperature of 630°C

The cessation of lengthwise growth in the U-Pu-Zr and U-Pu-Ti specimens is not unusual. As stated previously, similar behavior has been

observed in U-Pu-Fz irradiation specimens where the fission gases had been released in the fuel.

An average cladding diameter increase of 0.006 in. was calculated from volume changes measured by immersion. No measurable length change was noted. The cladding was punctured for fission-gas recovery, but because of equipment failure the gas sample was lost in the hot cell.

The metallographic examination of the specimen showed no significant penetration of the fuel into the cladding. But in isolated areas of the fuel-cladding interface, there was evidence of surface reaction penetrating into the fuel to a depth of ~ 0.0005 in. Figure 24 is a transverse section of the specimen. The central area of the specimen shows evidence of the existence of the $(U,Pu)_2Ti + \gamma$ phase, which is stable above $710^\circ C$. Figures 25 and 26 show photomacrographs of the fuel-cladding interface, as well as typical central porosity in the specimen.

EI-5887

20X

Fig. 24. Transverse Section of U-15 w/o Pu-10 w/o Ti Alloy (Specimen 44-1) Contained in V-20 w/o Ti Cladding and Irradiated to 10.7 a/o Burnup at Maximum Cladding Temperature of $630^\circ C$

Fig. 25. U-15 w/o Pu-10 w/o Ti Alloy (Specimen 44-1) Contained in V-20 w/o Ti Cladding and Irradiated to 10.7 a/o Burnup at Maximum Cladding Temperature of 630°C. The photograph at left shows the fuel-cladding interface; the photograph at right shows the size and distribution of center-fuel porosity.

Fig. 26. Typical Irradiated Structure of U-Pu-Ti Alloy. The dark areas show interconnected porosity. The light gray particles are believed to be remnants of the alpha-titanium phase observed in the preirradiated structure.

V. CONCLUSIONS

1. Of all the alloys studied, the U-Pu-Zr alloy system is the most promising for use in fast breeder reactors operating at cladding temperatures of 600-650°C. At these temperatures and irradiation conditions, alloys with zirconium concentrations of 10-12 w/o are compatible with Type 304 stainless steel cladding.

2. Metal fuels release substantial amounts of fission gas when fuel swelling exceeds ~30%. Fuel rods designed to take advantage of this characteristic can be successfully taken to burnups beyond 10 a/o.

3. The swelling of metal fuel during irradiation (at a given temperature and pressure) appears to be relatively uniform until fission-gas bubbles interlink and the gas is released from the fuel. After this occurs, swelling rates are reduced to relatively low values.

4. If the initial allowable radial fuel expansion is reduced to less than 30%, a proportionate increase in the final fuel length results. Under these conditions, where insufficient volume expansion has taken place to permit interlink of gas bubbles, the stresses on that portion of the cladding adjacent to the fuel become very high; reliance must then be placed on the restraint characteristics and structural integrity of the cladding.

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