ANL-76-28

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**U-Pu-Zr METAL ALLOY:
A POTENTIAL FUEL FOR LMFBR's**

by

**C. M. Walter, G. H. Golden,
and N. J. Olson**

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Printed in the United States of America
 At 1141 from
 U. S. Energy Research and Development Administration
 Technical Information Center
 P. O. Box 616
 Oak Ridge, Tennessee 37830
 Price: \$1.00 plus \$0.10

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EBR-II Project

November 1975

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ABSTRACT

This report critically reviews the available information pertinent to the potential use of U-Pu-Zr alloy fuels clad with stainless steel in LMFBR's. The areas considered include breeding potential, burnup potential, thermal performance, fuel fabricability, fuel reprocessing, and safety considerations. Because information on U-Pu-Zr alloys is limited, wide use is made of experience with EBR-II metallic driver fuel to infer advantages and limitations of U-Pu-Zr fuels. It is concluded that sufficient potential exists for the applicability of U-Pu-Zr fuels to LMFBR's to warrant additional analytical and experimental studies.

1. SUMMARY AND CONCLUSIONS

The principal advantages, uncertainties, and recommendations for the U-Pu-Zr metallic fuel system are summarized below.

A. Potential Advantages

1. The principal advantage of U-Pu-Zr fuels is their potential for high breeding ratio due to their high fertile/fissile atom densities. Earlier studies showed that U-Pu-10 wt % Zr has similar or perhaps slightly higher breeding ratios than (U, Pu)C fuels.

2. U-Pu-Zr fuels clad with stainless steel have a potential for high burnup (in excess of 10 at. %) at peak cladding temperatures of 1200-1300°F. Although U-Pu-Zr fuels clad in Type 316 stainless steel have been irradiated only to 4.5 at. % burnup, metallic EBR-II driver fuel (U-5 wt % Fs, clad with stainless steel that has a number of similarities in behavior) has shown high reliability up to 10.5 at. % burnup.

3. U-Pu-Zr fuels clad with stainless steel have the potential for fairly high-temperature operation by virtue of their high fuel-cladding eutectic temperatures (~1500°F) and high solidus temperatures (~2100°F). Low fuel-cladding eutectic temperatures and low solidus temperatures for metallic fuels containing plutonium have previously been a major limitation.

4. It appears that the fuel-fabrication processes (based on injection casting) used to fabricate over 100,000 irradiated and unirradiated EBR-II driver-fuel elements can be extended without significant difficulty to the fabrication of irradiated and unirradiated U-Pu-Zr fuel elements. Small numbers of U-Pu-Zr fuel elements have been successfully made by these processes.

5. Although a direct extension of the simple melt-refining reprocessing cycle used for EBR-II driver fuel is not possible for U-Pu-Zr alloys, direct application of segments of that cycle is feasible. Specifically, melting of the irradiated fuel to remove gaseous and volatile fission products can be applied to U-Pu-Zr. Much of the EBR-II experience with a close-coupled reprocessing facility that returns fuel to the reactor within 45 days of discharge from the reactor is applicable. The melt-refining process has the additional advantage that it can be conveniently scaled to fit reactor size, in contrast to the aqueous process.

6. Metallic fuels have potential advantages over oxide fuel for run-beyond-cladding-breach operation. These advantages primarily result from their better compatibility with the sodium coolant.

7. Scoping calculations indicate that, in LMFBR's of equivalent size, the inherent prompt negative feedback for U-Pu-Zr fuel is similar to that for oxide fuel.

B. Major Uncertainties

1. Because calculations of breeding ratio, inherent prompt negative feedback, and reactor response under off-normal conditions are very dependent on the details of a specific reactor design, the calculations performed to date should be considered tentative.

2. The irradiation performance at high burnups (>4.5 at. %) of U-Pu-Zr fuels clad with stainless steel has not been experimentally determined. Although the irradiation behavior of U-Pu-Zr fuels is similar to that of U-5 wt % Fe EBR-II driver fuel in many respects, there are important differences, the effects of which are unknown at high burnup.

3. The high fuel-cladding eutectic temperatures and the high fuel/cladding compatibility found in the U-Pu-Zr/stainless steel system are anomalous when compared to other plutonium-containing metallic fuels. Reliance on this high eutectic temperature and fuel/cladding compatibility must be accompanied by a good understanding of their cause. Although oxygen levels in the fuel and/or cladding appear to be associated with the high eutectic temperature, a fundamental understanding of the cause of this high eutectic temperature, both in and out of the reactor, is not available from studies conducted to date.

4. Although the basic injection-casting fuel-fabrication process has been adequately demonstrated for production of U-5 wt % Fs EBR-II driver fuel, it has only been used to produce small experimental quantities of U-Pu-Zr fuel elements.

5. Complete schemes for reprocessing of irradiated U-Pu-Zr alloys have been proposed. Significant portions of these schemes have been tested on EBR-II U-5 wt % Fs driver fuels and are applicable to U-Pu-Zr alloys, but major portions of the schemes are as yet untested on high-burnup metallic fuels.

C. Recommendations for Further Studies

1. Detailed calculations of breeding ratios, doubling times, and safety-related aspects of U-Pu-Zr-fueled LMFBF systems are required to provide the assurance that such systems have adequate potential to justify further development. These calculations are also required to provide guidance on fuel-element design and operating conditions for experimental irradiations.

2. The irradiation of experimental fuel elements (specifically, U-15 wt % Pu-10 wt % Zr clad with cold-worked Type 316 stainless steel and advanced alloys) to high burnup is required to determine the reliability of this fuel system as well as to characterize its irradiation behavior.

3. The cause of the high eutectic temperature and high fuel/cladding compatibility in the U-Pu-Zr/stainless steel system requires additional experimental as well as analytical effort, using irradiated and unirradiated alloys. For irradiated materials, careful pre- and postirradiation characterization and analysis of experimental fuel-element irradiations (see preceding paragraph) should be adequate. However, the fundamental understanding of the cause will require substantial out-of-reactor effort, particularly on determining the possible role played by oxygen.

4. Additional effort is required to optimize fabrication of U-Pu-Zr fuel pins. Of particular importance is work on the preparation of homogeneous alloys and on the development of the injection-casting process.

5. Development and demonstration are required for the proposed fast-turnaround fuel-reprocessing steps to convert, in a close-coupled fuel-recycle facility, irradiated U-Pu-Zr into new fuel elements for reirradiation.

II. INTRODUCTION

A. Scope and Objectives

This report objectively summarizes and evaluates the current state of knowledge on U-Pu-Zr fuel alloys that is germane to their potential use in LMFBR's. We will examine the major advantages and disadvantages of U-Pu-Zr fuels and identify principal areas of uncertainty in their behavior. Recommendations will be made on the value of additional studies for these alloys, as well as on specific areas which require further study if U-Pu-Zr fuels are to be pursued.

The following topics will be considered in the evaluation of U-Pu-Zr fuels:

1. Potential for breeding.
2. Irradiation performance in fast reactors.
3. Thermal performance, with emphasis on fuel/cladding compatibility.
4. Fuel fabricability.
5. Fuel reprocessing.
6. Safety-related considerations.

This report primarily considers ternary U-Pu-Zr alloys in the composition range of 12-18 wt % Pu and 6-14 wt % Zr; however, the emphasis is placed on U-15 wt % Pu-10 wt % Zr as a reference. Cladding materials include Types 304 and 316 stainless steel and a number of advanced alloys high in nickel, chromium, and, in some cases, cobalt; but the emphasis is on Type 316 stainless steel as a reference. Uranium-15 wt % Pu-10 wt % Zr and Type 316 stainless steel were chosen as reference materials because of the relative amount of data and analyses available on them, and because they represent a fuel system that could probably be sufficiently developed in the minimum time.

Although this report is not intended to provide an exhaustive review of all data available on alloy fuels (the data for plutonium-bearing alloys were well summarized by Kittel et al.¹ in 1971), information on other alloy fuels will be introduced where appropriate. Of particular value is the experience with U-5 wt % Fs.* Uranium-5 wt % fissionium clad with Types 304L and 316 stainless steel has been used as the exclusive driver fuel in the EBR-II reactor for more than 10 years. The information gained with U-5 wt % Fs in EBR-II therefore constitutes the largest single body of experience available

*Fissionium (Fs) is an equilibrium concentration of fission-product elements left by the pyrometallurgical reprocessing cycle designed for EBR-II and consists of 2.4 wt % molybdenum, 1.9 wt % ruthenium, 0.3 wt % rhodium, 0.2 wt % palladium, 0.1 wt % zirconium, and 0.01 wt % niobium.

on the application of metallic fuels to fast reactors. Furthermore, there are a number of similarities between U-Fs and U-Pu-Zr fabricability, reprocessing, safety properties, and irradiation behavior. Because of these two factors, it is appropriate to include somewhat detailed information on U-Fs in this report.

B. Design Concepts

Early engineering development of metal fuels for EBR-II centered on attempting to contain the fuel swelling with a strong cladding material; alternatively, a fuel-alloy development program was directed toward minimizing the fuel swelling. It was found that selective alloy additions greatly reduced swelling with respect to pure uranium, but a relative optimum was reached with the U-5 wt % Fs alloy currently used as the driver fuel in EBR-II. The initial fuel swelling due to the accumulation of solid fission products could not be readily restrained, and some room for this initial swelling was incorporated into a design to reliably achieve moderate burnups. This concept led to the Mark-IA design, currently used only in the EBR-II control and safety rods, which allows for 17% fuel swelling before fuel/cladding contact, accompanied by fission-gas releases from fuel to plenum of $\leq 2\%$ of total inventory. Even with such low gas release, the life expectancy of the design is burnup-limited due to (1) a small plenum volume of about 0.012 in.³ at 3 at. % BU_{max} (maximum fuel-pin burnup) at normal operating temperatures, and (2) a thin cladding-wall thickness of 0.009 in. The Mark-IA design is currently qualified at 1.8 at. % BU_{max} to obtain essentially a zero probability of failure. Efforts are underway to increase this burnup limit to a value nearer 3.0 at. %. About 80,000 Mark-IA driver-fuel elements have been irradiated in EBR-II, and considerable experimental effort has been expended to learn as much as possible about the cladding failure modes.^{2,3}

Further research at Argonne on the relationship between fuel swelling and gas release led to the engineering development of the Mark-II design currently used as the EBR-II driver fuel. This design concept allows for 33% fuel swelling prior to fuel/cladding contact, the plenum is much larger than that of the Mark-IA to accommodate the increased gas release, and the cladding is thicker (0.012 in.). It has been experimentally verified that interconnection of porosity occurs just prior to the swelling value of 33% theorized by Barnes.⁴ If large gas releases can be achieved prior to fuel/cladding contact, the contact pressures are dramatically reduced, as experimentally evidenced by the observed magnitudes of mechanical strain at a given burnup for the Mark-IA and -II designs.⁵ Therefore, there is considerable improvement of burnup capability of the Mark-II design over the Mark-IA. All EBR-II driver-fuel designs have used sodium as a thermal bond between the fuel pin and cladding. Therefore, changing the smear densities has been straightforward (i.e., changing the pin diameter, the fuel enrichment, and, correspondingly, the size of the sodium annulus between the pin and jacket).

To optimize performance of the metal fuel, a compromise between gas release and smear density (i.e., a large gas release requires a low smear density) is required. For the 33% fuel swelling that must be allowed to ensure

large gas release, the smear density must be 75%. Because of the sodium pin-to-cladding thermal bond, the smear density can be easily changed without significant effects on fuel or cladding temperatures. With the large gas releases, the plenum volume used is also a major design consideration. The Mark-II design concept thus far has shown a burnup capability of up to 10.5 at. % in EBR-II,⁶ even though the design parameters such as choice of cladding (Type 304L stainless steel) and plenum-volume/fuel-volume ratio (~0.8-1.0) may not be optimum for future fast-reactor application.

Since fission-gas release and fuel swelling are major variables in the design of a metal fuel element, it is instructive at this point to consider them in more detail. Fuel swelling as a function of burnup and operating temperature varies widely, even for fuel from the same batch.⁷ However, it has been shown that fission-gas release is a function of fuel swelling. At fuel temperatures above 950°F (see Fig. 1), the initial gas release rises sharply from ~2 to ~60% because of the interconnection of porosity, and is independent of the particular fuel-alloy composition.^{8,9} Therefore, the experimentally determined fuel swelling required for interconnection of the porosity (25-30%) is between that allowed by the Mark-IA and the Mark-II designs. This is an important point to consider with respect to smear density. It is also important to note that the relationship between gas release and fuel swelling is nearly the same for all metal fuels; i.e., the gas release is large when interconnection of porosity occurs. Therefore, the optimum fuel-element designs are similar, for example, for both the U-5 wt % metal EBR-II driver fuel and the more advanced U-Pu-Zr alloy.

Fig. 1

Effect of Swelling in Metallic Fuels on Release of Fission Gas,⁸ ANL Neg. No. 106-9681.

III. BREEDING POTENTIAL

A. Comparison with Other Fuel Types

The breeding ratio of a fast reactor is approximately proportional to the ratio of its ^{238}U atom density to its ^{239}Pu atom density. That is, the more ^{238}U the reactor contains relative to its ^{239}Pu content, the better it will breed. The ^{239}Pu critical mass of a large power LMFBR of a given height and diameter is only weakly dependent upon the detailed composition of the reactor. For any fuel used in such a reactor, a given amount must be ^{239}Pu ; the remainder is ^{238}U and oxygen, carbon, or metal diluent, depending upon the fuel type. It thus follows that the greater the fuel density, the more ^{238}U the reactor will contain, and the better its breeding potential will be.

Table I shows theoretical densities and theoretical heavy-atom (HA) densities for mixed-oxide, mixed-carbide, mixed-nitride, and U-Pu-10 wt % Zr metal-alloy fuels. On the basis of HA density, the metal fuel has the greatest breeding potential of the four fuel types.

TABLE I. Densities of Mixed-oxide, Mixed-carbide, Mixed-nitride, and U-Pu-10 wt % Zr Fuels

	Mixed Oxide ¹⁰	Mixed Carbide ¹⁰	Mixed Nitride ¹⁰	U-Pu-10 wt % Zr ¹¹
Theoretical fuel density, g cm ⁻³	10.9	13.6	14.3	16.0
Theoretical HA density, g cm ⁻³	9.8	12.9	13.5	14.4

Many studies have been made to calculate breeding ratios for LMFBR's using different fuels. Most such studies have been for mixed-oxide fuel, some have been for mixed-carbide fuel, and a few have been for mixed-nitride and metal-alloy fuels. Unfortunately, it is difficult to use these studies to make consistent comparisons of the breeding potential of the various fuel types. This is due to differences in design assumptions such as:

1. Core H/D (ratio of height to diameter) as based upon consideration of sodium-void effect versus economic optimization.
2. Core volume.
3. Thermal power level and spatial distribution (radial zoning).
4. Blanket size and composition.
5. Smeared fuel density.
6. Peak allowable linear power.
7. Peak allowable burnup.
8. Refueling scheme.

Other differences are in the cross-section sets used for physics analyses, in the physical, chemical, and mechanical properties, etc.

With such differences, it is not surprising that rather wide ranges of breeding ratios have been reported for the different fuel types. These ranges are about 1.15-1.35 for mixed-oxide fuel and 1.30-1.50 for mixed-carbide, mixed-nitride, and metal fuels. Thus, comparisons of various fuel types are meaningful only to the extent that there is consistency of design parameters for the reactors using these fuel types.

A recent design study by Caspersson et al.¹⁰ gives breeding ratios for mixed-oxide-, mixed-carbide-, and mixed-nitride-fueled LMFBR's sized to deliver about 500 MWe. The driver-region height was set at 3 ft for all three reactors; other design parameters are shown in Table II. The reactor breeding ratios for the oxide, carbide, and nitride LMFBR's are 1.20, 1.42, and 1.35, respectively. Note that these cores have volumes of about 2400-3100 liters, fuel volume fractions of 33.3-36.0%, and aspect ratios (H/D's) of 0.44-0.50.

TABLE II. Design Parameters for Oxide-, Carbide-, and Nitride-fueled LMFBR's with a Rating of about 500 MWe.¹⁰

	Mixed Oxide	Mixed Carbide	Mixed Nitride
Fuel form	(U, Pu)O _{1.98}	(U, Pu)C _{1.94}	(U, Pu)N _{1.90}
Fuel bond material	Helium	Sodium	Sodium
Heavy-metal smear densities			
g/cm ³	7.66	10.28	11.14
% of theoretical	80.0	79.1	82.2
Cladding OD-ID	0.250-0.220	0.354-0.324	0.339-0.309
Core-region heavy metal content, kg	7837	9490	9629
Average linear power, kW/ft	8.8	18.8	18.8
Average specific power, kW/kgHM	1.19	11.5	11.5
Core-region equivalent OD, ft	6.79	6.23	6.00
Core H/D ^a	0.44	0.48	0.50
Core volume, l	1076	2590	2402
Driver-fuel volume fraction, %	33.3	35.6	36.0
Total core power, MWt	1068	1090	1107
Average power density, kW/l	354	421	461

^aCore height 3 ft for all three reactors.

An unpublished parametric study of 1000-MWe LMFBR's using mixed-oxide, mixed-carbide, and metal-alloy (U-Pu-10 wt % Zr) fuel was done at Argonne National Laboratory in 1967. All such reactors had a power output of 2500 MWt, a core-power/reactor-power ratio of 0.9, average core power densities ranging parametrically from 300 to 700 kW/liter, and core aspect ratios ranging parametrically from 0.2-1.0. Design parameters for the reactors considered are shown in Table III.

TABLE III Design Parameters for Oxide-, Carbide-, and Metal-fueled LMFBR's with a Rating of about 1000 MWe

	Mixed Oxide	Mixed Carbide-1	Mixed Carbide-2	U-Pu-10 wt % Zr
Fuel burnup, MWd/ton	100,000	100,000	100,000	90,000
Full-power fuel lifetime, years	1.75	1.75	2.50	1.75
Smearcd fuel density, % of theoretical	80	80	80	70
Average linear power, kW/ft	10.5	16	16	8
Fuel-element OD, in	0.2545	0.271	0.318	0.212
Cladding-wall thickness, in	0.015	0.015	0.015	0.015

Two different sets of cases of the carbide-fueled reactor were considered. In the first, the full-power lifetime of the fuel was taken as 1.75 years, the same as that used for all the reactors fueled with mixed oxides and U-Pu-10 wt % Zr. For a given average core power density, burnup, and fractional fuel density, this led to unrealistically lower fuel volume fractions than were obtained for either the mixed-oxide or metal-alloy reactors. The lower fuel volume fractions resulted in correspondingly lower coolant velocities and core pressure drops and unrealistically low breeding ratios. It was thus decided to study another set of carbide-reactor cases in which flow characteristics were more like those of the mixed-oxide and metal-alloy reactors. This was done by increasing the full-power fuel lifetime to 2.5 years, leading to fuel volume fractions closer to those in the other systems. Results for the mixed-oxide fuel are given in Table IV, for the two mixed-carbide cases in Tables V and VI, and for the metal-alloy fuel in Table VII. The cores are all single-zone; i.e., all fuel has the same enrichment.

Interpolation of the results in Table IV indicates that, for a mixed-oxide reactor having an H/D of 0.44 and an average core power density of 350 kW/liter, the fuel volume fraction is 26.0% and the breeding ratio (BR) is 1.19. For a mixed-carbide core with the same full-power fuel lifetime of 1.75 years, an H/D of 0.48, and an average core power density of 420 kW/liter, the fuel volume fraction (see Table V) is 23.5% and the breeding ratio is 1.32. For the corresponding case with a full-power fuel lifetime of 2.50 years (Table VI), the fuel volume fraction is 33.6% and the breeding ratio is 1.48.

The metal-alloy reactors considered in Table VII (1.75-year full-power fuel lifetime) have hydraulic characteristics nearer to those of the 1.75-year carbide reactors than to those of the 2.50-year carbide reactors. That is, the metal reactors have conservatively low fuel volume fractions, and this has a strongly adverse effect on their breeding ratios. Nevertheless, the breeding ratio of a metal reactor having an H/D of 0.5 and an average core power density of 420 kW/liter is 1.36. The corresponding fuel volume fraction is only 26.9%. If the fuel volume fraction were increased to about 34%, the breeding ratio would increase to about 1.48.

TABLE IV Parameter Study of Single-zone Cores: Oxide Fuel with Full-power Lifetime of 1.75 Years at 100,000-MWd/Ton Burnup

H/D	Core Power Density, kW/liter	Core Volume, liters	Core Height, cm	Core Diameter, cm	Core Volume Fractions, %			M _{crit.} , kg	$\left(\frac{P_{\max}}{P_{\text{avg}}}\right)_{\text{ax}}$	$\left(\frac{P_{\max}}{P_{\text{avg}}}\right)_{\text{rad}}$	BR
					Fuel	Structure	Coolant				
1.0	300	7500	212.14	212.14	22.25	14.47	63.28	1878	1.3285	1.6423	1.157
	400	5625	192.63	192.63	29.66	18.14	52.20	1678	1.3393	1.6647	1.238
0.8	300	7500	182.57	228.30	22.25	13.87	63.88	1877	1.3014	1.6676	1.154
	400	5625	165.81	207.57	29.66	17.30	53.04	1675	1.3129	1.6902	1.239
0.6	300	7500	150.88	251.46	22.25	13.30	64.45	1900	1.2658	1.7018	1.148
	400	5625	136.86	228.30	29.66	16.50	53.84	1694	1.2779	1.7239	1.240
	500	4500	127.10	212.14	37.08	20.07	42.85	1556	1.2892	1.7485	1.307
0.4	300	7500	114.91	287.73	22.25	12.69	65.06	1990	1.2140	1.7499	1.141
	400	5625	104.55	261.51	29.66	15.67	54.67	1767	1.2276	1.7731	1.241
	500	4500	96.93	242.62	37.08	18.90	44.02	1618	1.2394	1.7972	1.316
0.2	300	7500	72.54	362.71	22.25	11.93	65.82	2330	1.1331	1.8327	1.130
	400	5625	65.84	329.49	29.66	14.66	55.68	2052	1.1466	1.8576	1.248
	500	4500	60.96	305.71	37.08	17.48	45.44	1865	1.1581	1.8829	1.337
	600	3750	75.30	287.73	44.50	20.75	34.75	1727	1.1688	1.9060	1.408

**TABLE V Parameter Study of Single-zone Cores Carbide Fuel with Full-power Lifetime of
1.75 Years at 100,000-MWd/Ton Burnup**

H/D	Core Power Density, kW/liter	Core Volume, liters	Core Height, cm	Core Diameter, cm	Core Volume Fractions, %			M _{crit} , kg	$\left(\frac{P_{max}}{P_{avg}}\right)_{ax}$	$\left(\frac{P_{max}}{P_{avg}}\right)_{rad}$	BR
					Fuel	Structure	Coolant				
1.0	300	7500	212.2	212.2	16.82	12.04	71.14	1868	1.317	1.616	1.228
	400	5625	192.8	192.8	22.42	13.91	63.67	1652	1.325	1.626	1.322
0.8	300	7500	182.8	228.5	16.82	11.34	71.84	1866	1.289	1.640	1.220
	400	5625	166.1	207.6	22.42	13.53	64.05	1653	1.298	1.653	1.316
0.6	300	7500	150.9	251.5	16.82	10.8	72.38	1892	1.253	1.674	1.208
	400	5625	137.1	228.5	22.42	13.03	64.55	1676	1.263	1.687	1.308
	500	4500	127.3	212.2	28.03	15.29	56.68	1528	1.272	1.704	1.385
0.4	300	7500	115.2	287.9	16.82	10.72	72.46	1990	1.203	1.724	1.186
	400	5625	104.6	261.6	22.42	12.36	65.22	1756	1.213	1.736	1.300
	500	4500	97.1	242.9	28.03	14.52	57.45	1598	1.223	1.754	1.383
	600	3700	91.4	228.5	33.64	17.54	48.82	1485	1.233	1.775	1.443
0.2	300	7500	72.6	362.8	16.82	9.26	73.93	2347	1.122	1.800	1.164
	400	5625	65.9	329.6	22.42	11.23	66.35	2061	1.134	1.818	1.291
	500	4500	61.2	306.0	28.03	13.14	58.83	1863	1.144	1.837	1.390
	600	3750	57.6	287.9	33.64	15.28	51.09	1717	1.153	1.858	1.466
	700	3214.3	54.7	273.5	39.24	17.84	42.92	1605	1.162	1.878	1.524

TABLE VI. Parameter Study of Single-zone Cores. Carbide Fuel with Full-power Lifetime of 2.50 Years at 100,000-MWd, Ton Burnup

H/D	Core Power Density, kW/liter	Core Volume, liters	Core Height, cm	Core Diameter, cm	Core Volume Fractions, %			M_{crit} , kg	$\left(\frac{P_{max}}{P_{avg}}\right)_{ax}$	$\left(\frac{P_{max}}{P_{avg}}\right)_{rad}$	BR
					Fuel	Structure	Coolant				
1.0	300	7500	212.2	212.2	24.03	12.80	63.18	2150	1.342	1.665	1.361
0.8	300	7500	182.8	229.5	24.03	12.01	63.97	2147	1.316	1.689	1.361
	400	5625	166.1	207.6	32.03	15.12	52.84	1922	1.327	1.712	1.456
0.6	300	7500	150.9	251.5	24.03	11.41	64.56	2171	1.281	1.723	1.358
	400	5625	137.1	228.5	32.03	14.27	53.69	1941	1.294	1.746	1.460
0.4	300	7500	115.2	287.9	24.03	10.57	65.41	2262	1.231	1.769	1.357
	400	5625	104.6	261.6	32.03	13.38	54.59	2017	1.245	1.794	1.465
	500	4500	97.1	242.9	40.04	16.57	43.39	1852	1.257	1.819	1.542
0.2	300	7500	72.6	362.8	24.03	9.90	66.08	2624	1.151	1.851	1.351
	400	5625	65.9	329.6	32.03	12.26	55.71	2316	1.165	1.876	1.479
	500	4500	61.2	306.0	40.04	14.86	45.10	2107	1.177	1.901	1.571
	600	3750	57.6	287.9	48.05	18.22	33.73	1958	1.188	1.927	1.634

TABLE VII. Parameter Study of Single-zone Cores: Metal Fuel with Full-power Lifetime of 1.75 Years at 90,000-MWd/Ton Burnup

H/D	Core Power Density, kW/liter	Core Volume, liters	Core Height, cm	Core Diameter, cm	Core Volume Fractions, %			M _{crit} , kg	$\left(\frac{P_{\max}}{P_{\text{avg}}}\right)_{\text{ax}}$	$\left(\frac{P_{\max}}{P_{\text{avg}}}\right)_{\text{rad}}$	BR
					Fuel	Structure	Coolant				
1.0	300	7500	212.2	212.2	19.20	16.07	64.73	1926	1.321	1.616	1.257
0.8	300	7500	182.8	228.5	19.20	15.67	65.13	1928	1.294	1.642	1.248
	400	5625	166.1	207.6	25.60	18.81	55.59	1686	1.301	1.652	1.355
0.6	300	7500	150.9	251.5	19.20	14.93	65.86	1955	1.259	1.675	1.237
	400	5625	137.1	228.5	25.60	17.95	56.45	1708	1.266	1.686	1.350
0.4	300	7500	115.2	287.9	19.20	13.96	66.83	2050	1.209	1.722	1.221
	400	5625	104.6	261.6	25.60	17.12	57.28	1790	1.218	1.736	1.339
	500	4500	97.1	242.9	32.00	20.45	47.55	1609	1.225	1.754	1.429
0.2	300	7500	72.6	362.8	19.20	13.74	67.07	2418	1.134	1.810	1.182
	400	5625	65.9	329.6	25.60	15.86	58.54	2095	1.143	1.824	1.320
	500	4500	61.2	306.0	32.00	19.00	49.00	1872	1.151	1.844	1.423
	600	3750	57.6	287.9	38.40	22.17	39.42	1706	1.158	1.864	1.505

These calculations confirm that metal-alloy fuels have breeding potential that is similar to that of mixed-carbide fuel.

B. Conclusions on Breeding Potential

1. Breeding ratios of large metal-fueled LMFBR's are estimated to be about 1.5 compared with about 1.25 for corresponding oxide-fueled LMFBR's. Since reactor doubling times vary approximately as $1/(BR - 1)$, doubling times for the metal-fueled reactors would be expected to be about half the doubling times for corresponding oxide-fueled reactors.

2. Detailed calculations of breeding ratios and doubling times for U-Pu-Zr-fueled systems are required to provide assurance that such systems have adequate potential to justify further development. These calculations are also required to provide guidance on fuel-element design and operating conditions for experimental irradiations.

IV. BURNUP POTENTIAL

The design concept for metal fuels that has evolved--allowing for optimum fission-gas release--has demonstrated high burnup potential. Burnup potential is a key consideration in assessing the desirability of metallic fuels for use in advanced LMFBR's. The largest body of data on the irradiation behavior of fast-reactor fuels has been generated on the EBR-II driver fuel (U-5 wt % Fs clad with solution-annealed (SA) 304L and SA 316 stainless steel). However, significant, though limited, irradiation results have also been obtained on the U-Pu-Zr and U-Pu-Ti fuel systems.

A. Irradiation Experience with U-5 wt % Fs System (EBR-II Driver Fuel)

The EBR-II driver-fuel element is composed of a U-5 wt % Fs metallic fuel pin sodium-bonded to a stainless steel cladding. Two principal driver-fuel-element designs (Mark IA and Mark II) have been employed. These designs are described in Table VIII.¹² Some variations of the Mark II design have been

TABLE VIII Comparison of Mark-IA and -II Fuel-element Designs Irradiated in EBR-II¹²

	Mark IA	Reference Core ^a	Mark II	
			Experimental Elements	
			Encapsulated	Unencapsulated and Encapsulated
Enrichment, at % ²³⁵ U	52.5	67.0	93 and 80	64.0
Fuel-pin weight, g	64.0	51.7	54.5	54.5
Fuel-pin length, in	13.5	13.5	14.2	14.2
Fuel-pin diameter, in	0.144	0.130	0.130	0.130
Fuel volume, in ³	0.220	0.177	0.188	0.188
Fuel Cladding radial gap, in	0.006	0.010	0.010	0.010
Cladding wall thickness, in	0.009	0.012	0.012	0.012
Cladding OD, in.	0.174	0.174	0.174	0.174
Cladding material	SA 304L	SA 316	SA 304L	SA 304L and SA 316
Element length, in	18.1	24.1	24.1 and 26.1	26.1
Plenum volume, in ³	0.041	0.145	0.145-0.184	0.164
Plenum gas	Argon	Argon	Helium	Argon
Sodium level above fuel, in	0.65	1.25	0.3-1.0	1.5
Restrainer height above fuel, in	0.40	0.50	0.2-0.8	0.5, 1.0, and none
Restrainer type	Solid	Dimple	Ring, split ring, or dimple	Dimple
Spacer-wire diameter, in	0.049	0.049	None on element	0.049 (Unencapsulated only)
Spacer-wire material	SA 304L	SA 316	None	SA 304L (Unencapsulated only)

^aCurrent driver fuel for EBR-II

experimentally irradiated in EBR-II, and these are also shown in Table VIII. Since the Mark-II design, in concept, is the closest to optimum for metal fuels with respect to smear density, ratio of plenum volume to fuel volume, etc., only the irradiation results for Mark II will be discussed. The irradiation results for Mark IA are discussed in detail in Ref. 3.

The experimental irradiation programs for Mark-II fuel have been carried out in two simultaneous parts: encapsulated and unencapsulated irradiations. A rather large number of design variations were considered, such as: 64, 80, and 93% enrichments of ^{235}U ; two types of cladding (SA 304L and SA 316); different types of restrainers to restrain axial fuel growth during irradiation; and different plenum/fuel volume ratios, which were achieved by using elements of different lengths (24 and 26 in.) and slight pre-irradiation adjustments of the sodium levels above the fuel (see Table VIII).

One of the principal objectives of these experiments is to obtain information on the final cladding-failure mode(s) associated with the Mark-II driver-fuel design. This information not only provides necessary input to the driver-fuel programs, but may also provide design information for more advanced metal fuels (e.g., U-Pu-Zr systems). Because a reasonably large number of design variations were to be tested, a large number of failures were required. Most of the run-to-failure program was carried out in encapsulated elements in EBR-II (subassembly X071) in order to minimize the effect of the experiment on reactor plant factor. However, one subassembly (X180) containing unencapsulated elements was run to first failure, and a number of run-to-failure experiments with reference-core-design Mark-II fuel are in progress, but have yet (as of June 1975) to achieve burnups at which failures are expected.

The experiment with encapsulated Mark-II driver fuel (designated X071H as of June 1975) having the design variations shown in Table VIII has contributed most of the information for establishing the burnup potential of metal fuels. This experiment has been ongoing in EBR-II since 1967 (initially as subassembly X029) and has had 11 reconstitutions.¹³ All reconstitutions and reactor positions for this experiment have been planned so as to maintain a constant mean power-to-flow ratio such that the operating temperatures and power ratings of individual fuel elements were kept as constant as possible. The experiment has produced 20 cladding failures for the 80%- and 93%-enriched elements with SA 304L cladding. Data on these failures are listed in Tables IX-XIV. (The statistical parameters given in the tables are described in Sec. IV.A.1 below.) One 93%-enriched element has run to 16.1 at. % burnup without failure. In addition, a number of encapsulated 64%-enriched, 26-in.-long elements with both SA 304L and SA 316 claddings have been irradiated to ~9.3 at. % maximum burnup in X071H without failure (see Tables XV and XVI).

TABLE IX. Cladding Failures of Encapsulated Experimental Mark-II Elements
with SA 304L Cladding: 93%-enriched Fuel, 24-in.-long Elements

Element No.	Restrainer Type ^a	Na Level, in.	Plenum Volume, in. ³	T _{clad} ^b _{max} , °F	Axial Failure Location, ^c in.	BU _{max} , at. %	Failure Rank	F(BU _{max})	Remarks
7 total	-	-	-	-	-	≤8.1	Unfailed	-	Suspended
205	A	0.36	0.149	1120	7.5	9.2	1.875	-	From X071A
230	C	0.32	0.150	1130	7.5	9.2	3.750	0.240	From X071A
203	A	0.46	0.147	1040	10.0	9.4	5.625	0.370	From X071A
234	B	0.52	0.146	d	-	9.4	Unfailed	-	Suspended after X071A
237	C	0.53	0.146	1070	7.7	10.0	7.969	0.533	From X071B
224	C	0.57	0.145	d	d	10.4	10.313	0.695	From X071H
232	A	0.48	0.147	1100	-	16.4	Unfailed	-	Still running after X071H

BU₀ = 8.9 at. %; β = 0.854; η = 1.33 at. %; R² = 98.2% (explanation of variance).

^aA ≡ split ring, B ≡ ring, C ≡ dimple.

^bMaximum time-averaged cladding temperature.

^cDistance from bottom of fuel column.

^dTo be determined.

TABLE X. Cladding Failures of Encapsulated Experimental Mark-II Elements
with SA 304L Cladding: 93%-enriched Fuel, 26-in.-long Elements

Element No.	Restrainer Type ^a	Na Level, in.	Plenum Volume, in. ³	$T_{\text{clad max}}^{\text{b}}$, °F	Axial Failure Location, ^c in.	BU_{max} , at. %	Failure Rank	$F(BU_{\text{max}})$	Remarks
7 total	-	-	-	-	-	≤7.9	Unfailed	-	Suspended
256	A	0.83	0.176	d	-	10.4	Unfailed	-	From X071H
263	A	0.50	0.182	1110	9.7	10.6	2.143	0.128	From X071C
266	B	0.86	0.176	1070	9.5	10.8	4.286	0.277	From X071C
256	A	0.88	0.175	d	d	11.0	6.429	0.426	From X071H
267	C	0.84	0.176	1090	8.5	12.1	8.571	0.574	From X071E
265	A	0.91	0.175	1060	12.5	12.6	10.714	0.723	From X071E
288	C	0.36	0.181	1060	-	13.3	Unfailed	-	From X071G
285	C	0.36	0.184	1060	d	(14.9) ^e	(12.857) ^e	(0.872) ^e	Failed in X071H, which had 16.2 at. % BU_{max}

$BU_0 = 10.5$ at. %; $\beta = 0.691$; $\eta = 1.57$; $R^2 = 97.4\%$ (explanation of variance).

^aA ≡ split ring, B ≡ ring, C ≡ dimple.

^bMaximum time-averaged cladding temperature.

^cDistance from bottom of fuel column.

^dTo be determined.

^eBest estimates. Exact burnup at time of element failure is not known.

TABLE XI. Cladding Failures of Encapsulated Experimental Mark-II Elements with SA 304L Cladding: 80%-enriched Fuel, 24-in.-long Elements

Element No.	Restrainer Type ^a	Na Level, in.	Plenum Volume, in. ³	$T_{\text{clad}}^{\text{max}}$ ^b , °F	Axial Failure Location, ^c in.	BU_{max} , at. %	Failure Rank	$F(BU_{\text{max}})$	Remarks
11 total	-	-	-	-	-	≤8.2	Unfailed	-	Suspended
200	C	0.49	0.147	1050	9.5	10.2	3.75	0.240	From X071E
209	A	0.39	0.149	1030	8.5	10.4	7.50	0.500	From X071E
202	C	0.31	0.150	1050	9.0	10.5	11.25	0.760	From X071F

^aA ≡ split ring, C ≡ dimple.

^bMaximum time-averaged cladding temperature.

^cDistance from bottom of fuel column.

TABLE XII. Failure Data Used in Analysis of 80%-enriched, 24-in.-long Encapsulated Experimental Mark-II Elements with SA 304L Cladding

BU_{max} , at. %	Failure Fraction	$F(BU_{\text{max}})$	Remarks
9.4	0/3	0.017 ^a	X071D results
10.2	2/3	0.500	X071E results
10.4	3/3	0.794	X071E and X071F results

$BU_0 = 8.9$ at. %; $\beta = 4.03$; $\eta = 1.38$; $R^2 = 99.7\%$ (explanation of variance).

^aReliability of three elements assumed to be 0.95.

TABLE XIII. Cladding Failures of Encapsulated Experimental Mark-II Elements
with SA 304L Cladding: 80%-enriched Fuel, 26-in.-long Elements

Element No.	Restrainer Type ^a	Na Level, in.	Plenum Volume, in. ³	$T_{\text{clad}}^{\text{max}}$ ^b °F	Axial Failure Location, ^c in.	BU_{max} at. %	Failure Rank	$F(BU_{\text{max}})$	Remarks
5 total	-	-	-	-	-	≤8.4	Unfailed	-	Suspended
275	A	0.41	0.184	1070	12.4	11.3	1.625	-	From X071G
284	A	1.00	0.173	1070	9.0	11.3	3.250	0.238	From X071G
269	C	0.40	0.184	d	d	13.2	4.875	0.369	From X071H
276	A	0.92	0.175	d	d	13.3	6.500	-	From X071H
277	B	0.40	0.184	d	d	13.3	8.125	0.631	From X071H
287	B	0.77	0.177	d	d	13.5	Unfailed	-	From X071H
270	C	0.97	0.174	d	d	13.7	10.563	0.828	From X071H

^aA ≡ split ring, B ≡ ring, C ≡ dimple.

^bMaximum time-averaged cladding temperature.

^cDistance from bottom of fuel column.

^dTo be determined.

TABLE XIV. Failure Data Used in Analysis of 80%-enriched, 26-in.-long Encapsulated Experimental Mark-II Elements with SA 304L Cladding

BU_{max} , at. %	Failure Fraction	$F(BU_{max})$	Remarks
10.4	0/7	0.0073 ^a	X071F results
11.1	2/7	0.230	X071G results
13.5	6/7	0.770	X071G and X071H results

$BU_0 = 10.3$ at. %; $\beta = 1.54$; $\eta = 2.26$ at. %; $R^2 = 99.3\%$ (explanation of variance).

^aReliability of seven elements assumed to be 0.95.

TABLE XV. Status of 64%-enriched, 26-in.-long Encapsulated Experimental Mark-II Elements with SA 304L Cladding

Element No. ^a	Na Level, in.	Plenum Volume, in. ³	T_{clad}^{max} ^b , °F	BU_{max} , at. %	Status
11	1.5	0.164	1024	2.8	Unfailed
3	1.5	0.164	954	8.5	Unfailed
10	1.5	0.164	1048	8.6	Unfailed
6	1.5	0.164	1028	8.8	Unfailed
1	1.5	0.164	1062	9.3	Unfailed

^aFuel restrainers all dimple type.

^bMaximum time-averaged cladding temperature.

TABLE XVI. Status of 64%-enriched, 26-in.-long Encapsulated Experimental Mark-II Elements with SA 316 Cladding

Element No. ^a	Na Level, in.	Plenum Volume, in. ³	T_{clad}^{max} ^b , °F	BU_{max} , at. %	Status
12	1.5	0.164	1066	7.5	Unfailed
7	1.5	0.164	1020	8.6	Unfailed
17	1.5	0.164	986	9.2	Unfailed
15	1.5	0.164	986	9.4	Unfailed

^aFuel restrainers all dimple type.

^bMaximum time-averaged cladding temperature.

A large number of unencapsulated 64%-enriched, 26-in.-long elements have been irradiated to 6 at. % maximum burnup in six standard 91-element driver-fuel subassemblies. About two-thirds of the initial 556 elements achieved 6 at. % maximum burnup; about half of these elements were clad with SA 304L, and the other half with SA 316. No cladding failures occurred in this portion of the experimental program (up to 6 at. % BU_{max}); the irradiation results are reported in Ref. 14.

The unencapsulated-irradiations program with 64%-enriched, 26-in.-long elements was continued from the 6 at. % BU_{max} level with 37 elements clad only with SA 316. This final experiment, X180, produced a single failure at 9.3 at. % BU_{max} , as shown in Table XVII.* The peak cladding temperatures (at the inside cladding wall at the top of the fuel column) in X180 are calculated to range from 970 to 1070°F (~1060°F for the failed element).

TABLE XVII Failure Statistics of 64%-enriched, 26-in.-long Unencapsulated (X180) Experimental Mark-II Elements with SA 316 Cladding

Number of Elements ^a	Na Level, in	Plenum Volume, in ³	Axial Failure Location, ^b in	BU_{max} at %	Failure Rank	F- BU_{max}	Remarks
2	1.5	0.164		9.0			Suspended
4	1.5	0.164		9.1			Suspended
7	1.5	0.164		9.2			Suspended
12	1.5	0.164	10.8	9.3	1/520	0.033	Failed (No. 556) ^c
7	1.5	0.164		9.4			
5	1.5	0.164		9.5			

^aFuel restrainers all dimple type

^bDistance from bottom of fuel column

In addition to the previously described encapsulated and unencapsulated experiments, there is a large ongoing run-to-failure program associated with the 67%-enriched, SA-316-clad, 24-in.-long reference Mark-II design (see Table VIII). At present (June 1975), 91 elements have been irradiated up to 6 at. % BU_{max} , and 4459 elements have been irradiated to the current 4.7 at. % burnup limit without a single element failure.¹⁵

1. Statistical Analysis of Cladding Failures

Since end-of-design-life failures have occurred only with the U-5 wt % metal driver-fuel alloy with stainless steel cladding, the statistical analysis of failures must by necessity be limited to this system. Only the results for the encapsulated 80%- and 93%-enriched fuel with SA 304L cladding will be analyzed. Before statistical analysis of failures may be performed, a suitable statistical failure model must be chosen or determined.

*It has been subsequently found that this failed element was clad with SA 304, and is therefore not typical of Mark-II clad with SA 316.

Since its publication,¹⁶ the Weibull failure model has received considerable attention because of its versatility in producing good agreement with data in a number of reliability-oriented industries.¹⁶⁻²¹ Application of the Weibull model to failures of reactor fuel elements was first made in 1954 at the Hanford production reactors and never failed to provide an adequate description of the fuel-element failure data, a prime requirement in empirical model building.²² Recently, the model has provided a very good statistical description of failures of Mark-IA driver fuel in EBR-II.³ Theoretically, the Weibull distribution is applicable when one is concerned with the failure of an item in which a large number of flaws exist and the failure depends upon the severest flaw in the system. Hence, the theoretical basis for the Weibull model is that it is a reasonable description of what leads to failure. The basic Weibull distribution function may be written as

$$F(x) = 1 - \exp\left[-\left(\frac{x - x_0}{\eta}\right)^\beta\right], \quad (1)$$

where

x = time-dependent variable,

$F(x)$ = cumulative failure probability at x , or the fraction of the population accounted for,

x_0 = origin of the distribution [$F(x \leq x_0) = 0$],

η = scaling parameter (>0),

$\eta + x_0$ = characteristic life [$F(\eta + x_0) = 63\%$],

and

β = shape parameter, or Weibull slope (>0).

Next, one requires a method to determine the best estimates for the observed failure probabilities. All the failure data that will be analyzed here came from the X071 experiment with encapsulated fuel (Tables IX-XIV); hence, we do not know the exact burnup or time of failure. But we do know the cumulative probability of failure at any time the experiment was examined. Thus we choose to work with the cumulative failure probability (Eq. 1) for the analysis.

The binomial correction employed by Johnson²³ was used in assessing the failure probabilities from the data (Tables IX-XIV). This correction is necessary because, for small sample size, there is a better than even chance that a larger failure fraction will be observed than dictated by the actual median failure probability. Instead of using the actual observed failure fraction, we used the simplified binomial interpretation of the cumulative failure probability.²³

$$F(x) = \frac{j - 0.3}{N + 0.4}, \quad (2)$$

where j is the number of failures observed out of N observations, or the rank order number. The correction is relatively small for $N > 10$.

Another correction is required when suspended items are involved. This approach is covered in detail by Johnson in Ref. 23; the expression for the increment in failure rank (Δn) is

$$\Delta n = \frac{(N + 1) - (\text{previous rank order number})}{1 + (\text{number of items beyond present suspended item})} \quad (3)$$

The relationship between Eqs. 2 and 3 is

$$j = \sum_{i=1}^j \Delta n_i.$$

Equations 2 and 3 were used to assess the failure probabilities in Tables IX, X, XI, and XIII. Some of the data points for 80%-enriched fuel in Tables XI and XIII have burnup intervals too close together to achieve valid analysis for the entire data sets. Therefore, we used the last known burnup at which there were no failures (assuming 95% reliability at such burnups), and applied averaging techniques to obtain at least three well-conditioned data points from Tables XI and XIII; those points are listed in Tables XII and XIV. The best-estimate Weibull parameters for the respective data sets are shown in Tables IX, X, XII, and XIV.

Although there were not enough data points to statistically test all design parameters, there is a definite difference between 24- and 26-in.-long SA-304L-clad elements. The difference is basically an additional 1.5 at. % BU_{max} capability for the longer elements, which may be indicative of the larger plenums of those elements. There are no failures for the encapsulated 64%-enriched elements with SA 304L or SA 316 cladding (Tables XV and XVI). The only failure for the unencapsulated 64%-enriched fuel is with SA 304L cladding (Table XVII). Further conclusions will have to await more irradiation results. The experimental failure statistics for data accumulated thus far are graphically illustrated in Fig. 2, along with the lines describing the results calculated from the respective Weibull parameters.

2. Cladding Fracture Modes

The failure mode(s) for encapsulated, SA-304L-clad Mark-II elements has not been fully established. Many of the early-burnup failures associated with the 93%-enriched 24-in. elements (see Table IX) exhibited small

Fig. 2. Cumulative Failure Probability vs Maximum Burnup for Variations in Design of Mark-II Driver Fuel. ANL Neg. No. 103-T5320

longitudinal cladding cracks, but later (in time) failures with higher neutron doses (e.g., Table XIII) and larger cladding swelling tend to exhibit "pin-holes" and are rather difficult to locate. All the cladding failures located thus far have been 7.5-12.5 in. above the bottom of the fuel pin, as noted in Tables IX-XIV.

To date (June 1975), only one failure of a Mark-II element has been examined in detail,²⁴ but the results are worth re-

viewing. Scanning-electron-microscopy (SEM) evaluation of the fracture surface from the SA 304L cladding of encapsulated element 230 (see Table IX) from experiment X071A was completed at LASL. This cladding was observed to have failed 7.5 in. from the bottom of the fuel column when the element was examined at 9.2 at. % BU_{max} . Some of the irradiation parameters of interest at the failure location were: 16.1% cladding volume swelling, 2.4% mechanical hoop strain, $5.8 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$) fluence, and peak calculated cladding temperature of 1078°F at the inner wall.

A typical region of the fracture surface is shown in Fig. 3. The fuel/cladding interaction zone (20% of the 0.012-in. cladding thickness) is to the left of the arrow. The region to the right of the arrow (toward the outer cladding surface) exhibits a very ductile region, with the dimples indicating the direction of fracture propagation from the outside to the inside of the cladding wall. There was considerable evidence of stress corrosion, as is apparent in Fig. 4, with the attack shown on the outer surface of the cladding adjacent to the fracture surface. The fracture surface observed in this examination was far more ductile in appearance than that observed for the Mark-IA design (see Ref. 3), even though both designs of driver fuel have similar uniform hoop strains at the fracture locations. (Mechanical hoop strains averaging 2% are readily achievable before failure of SA 304L-clad Mark-II elements.⁵) Peak SA 304L cladding volume swellings of up to 23% before failure have been measured by immersion density.

3. Cladding-diameter Change

A considerable amount of data on cladding-diameter change as a function of irradiation exposure has been generated for both SA-304L-clad

Fig. 3 Typical Region of Cladding Fracture Surface of Element 23 from Subassembly X-71A. Mag. 300X. AML Neg. No. 1-B-R7-22.

Fig. 4 Attack of Cladding Surface Adjacent to Fracture of Element 23 from Subassembly X-71A. Section A - the outer cladding surface, and section B - the fracture surface. Mag. 300X. AML Neg. No. 1-B-R7-21.

and SA-316-clad Mark-II elements. These data are summarized in Fig. 5 in terms of maximum burnup. The data points have been omitted from the figure to improve clarity; the lines represent mean values for the maximum diameter changes of several individual elements. The results are plotted two ways: (1) the maximum diameter change due to cladding swelling ($\Delta V/V_0$) and (2) the total measured maximum diameter change (total strain). The swelling is measured by immersion density, and $\Delta D_{\max}^{\text{swell}}/D_0$ is assumed equal to $(\Delta V/V_0)/3$. The $\Delta D_{\max}/D_0$ for an individual element normally occurs above the core midplane. The difference between the total diameter change and the contribution of swelling to it is an estimate of the mechanical hoop strain. Two fluence scales are given to encompass the range of fluence-to-burnup ratios caused by the different ^{235}U enrichments.

Fig. 5. Maximum Diametral Cladding Strain--Total and That Contributed by Swelling--for Mark-II Fuel Elements Irradiated in EBR-II. ANL Neg. No. 103-T5335.

It appears possible to achieve very large diameter changes with the SA-304L-clad elements prior to failure. These large $\Delta D_{\max}/D_0$ values at high burnups result from large contributions from both swelling and mechanical hoop strain. The synergistic effect of temperature, neutron dose, and stress on $\Delta D_{\max}/D_0$ are not known, but the difference between the performance of SA 316 and SA 304L cladding is illustrated reasonably well in Fig. 5. The figure also shows what appears to be a nearly constant ratio between the contributions of swelling and mechanical strain to $\Delta D_{\max}/D_0$.

B. U-Pu-Zr Fuel System

For a metal fuel to be considered in the LMFBR program, it must be a plutonium-bearing alloy (containing 10-20 wt % Pu) that is compatible with

commercially available stainless steel and advanced alloy claddings. Some research and development work to find such a fuel has been carried out in the past at Argonne National Laboratory. This work resulted in a promising advanced fuel-alloy system, U-Pu-Zr, that showed a fuel-alloy-stainless-steel eutectic temperature of 810 C (1490 F) as compared to the U-5 wt % Fe alloy-stainless steel eutectic temperature of 705 C (1301 F).²⁵ Since the eutectic temperature is the assumed limiting variable for the application of metal fuels to advanced LMFBR designs, the higher temperature for the plutonium-bearing metal fuel alloy is significant.

A number of in- and out-of-pile experiments have been completed with the following U-Pu-Zr compositions (in wt %):¹

1. U-15 Pu-10 Zr.
2. U-18.5 Pu-14.1 Zr.
3. U-16.6 Pu-6.3 Zr.
4. U-15 Pu-9 Zr.
5. U-14 Pu-12 Zr.
6. U-15 Pu-12 Zr.

The first irradiations of U-Pu-Zr alloys were performed on prototype EBR-II fuel elements of U-15 Pu-12 Zr alloy clad in SA 304 stainless steel, SA 316 stainless steel, Hastelloy-X, and V-20 Ti alloy. The elements were irradiated in instrumented, temperature-controlled capsules in the CP-5 reactor to 2.4 at. % burnup at a maximum cladding temperature of 1130 F (610 C).²

Metallographic examinations of the irradiated specimens showed excellent compatibility between the fuel and the different cladding compositions. The transverse macrosections of the fuel showed three distinct concentric bands which are related to phase distributions existing at the temperature of irradiation (see Fig. 6). The outer zone is believed to correspond to the

Fig. 6

Transverse section of U-15 wt % Pu-12 wt % Zr Alloy Jacketed in Hastelloy-X, after 2.5 at. % Burnup. The concentric bands in the fuel are related to phase distributions in the alloy during irradiation. (ANL Neg No. 176-5913)

alpha-uranium + delta + zeta phase, the second band to the alpha-uranium + zeta + gamma phase, and the central area to the gamma phase. The fuel had swelled to the internal diameter of the cladding, and the bond sodium was entirely displaced into the gas plenum. Figure 6 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.

Irradiation of the U-Pu-Zr element clad in V-20 Ti alloy was continued to 12.5 at. % burnup at a maximum cladding temperature of 1210°F (654°C). Periodic neutron radiography of the capsule during irradiation showed that the fuel attained a maximum elongation of 3% within the cladding, which occurred at 4.2 at. % burnup. This dimension did not change throughout the remainder of the irradiation.

An average 0.0002-in. increase of the original cladding diameter of 0.174 in. was measured. No measurable length increase of the cladding was noted. It was determined that 74% of the fission gas had been released to the plenum. The cladding was intact.

Because of the promising results obtained on small prototype fuel elements, irradiations were made on full-length elements of U-Pu-Zr alloy in EBR-II subassemblies XG05, XG06, XA07, and XC28.^{8,26} Table XVIII lists the design and operating parameters of those fuel elements and two others containing U-15 Pu-10 Ti fuel.

Fuel compositions in the EBR-II irradiations were U-15 Pu-9 Zr, U-15 Pu-10 Zr, and U-15 Pu-12 Zr, with smeared densities ranging from 66 to 75%. Cladding included SA 304 and SA 316 stainless steel, Hastelloy-X, Hastelloy-X-280, and V-20 Ti. All cladding was in the annealed state. Fuel-element diameters were 0.196 or 0.208 in., with wall thicknesses of 0.015 or 0.019 in. The fuel pins were nominally 13 in. long, with 0.144-in. diameters. Linear heat ratings ranged from 9.1 to 10.5 kW/ft, and maximum cladding temperatures ranged from 1021 to 1206°F (549 to 652°C).

The most thoroughly evaluated EBR-II irradiation data for the U-Pu-Zr fuel system are the results from the 16 encapsulated elements contained in subassembly XA07.⁸ These elements were irradiated to 4.0-4.6 at. % burnup without cladding failure.

External dimensional measurements of the irradiated elements showed maximum diameter changes of 0.05-0.5% at the 4.5 at. % burnup level. These changes are somewhat lower than would be expected with SA-316-stainless-steel-clad U-5 wt % Fs (see Fig. 5); however, the U-Pu-Zr elements had somewhat larger plenums, so their ratio of hoop stress to plenum pressure was lower. No immersion-density measurements were taken to determine the cladding swelling. Fuel-pin lengths within the cladding were determined by

TABLE XVIII. Design and Operating Parameters for U-Pu-Zr and U-Pu-Ti Fuel Irradiated in EBR-II

Fuel Composition, wt %	Element No.	EBR-II S A No	Fuel Smeared Density, % TD	Cladding			Max Linear Heat, kW/ft	Max Cladding Temp, °F	Burnup, ^a at %
				Material	OD, in	Thick, in.			
U-15 Pu-9 Zr	ND-28	XA07	73	304 SS	0.205	0.019	10.9	1206	4.6
	ND-32		74	316 SS	0.196	0.015	9.9	1164	4.2
	ND-41		74	304 SS	0.205	0.019	10.4	1197	4.4
	ND-43		75	Hast -X	0.196	0.015	10.4	1180	4.4
U-15 Pu-10 Zr	BA-01	X028	75	304L SS	0.196	0.015	9.4	1182	0.77
	BA-02		75	304L SS	0.196	0.015	9.4	1182	0.77
	BA-03		75	304L SS	0.196	0.015	9.4	1182	0.77
	BA-04		75	304L SS	0.196	0.015	9.5	1184	0.78
	BA-05		75	304L SS	0.196	0.015	9.4	1182	0.77
	BB-01		75	304L SS	0.205	0.020	9.5	1184	0.78
	BC-01		75	316 SS	0.196	0.015	9.4	1182	0.77
	BC-02		75	316 SS	0.196	0.015	9.5	1184	0.78
	BC-03		75	316 SS	0.196	0.015	9.4	1182	0.77
	BD-03		76	V-20 Ti	0.195	0.015	9.4	1182	0.77
	BD-04		76	V-20 Ti	0.195	0.015	9.4	1182	0.77
	BD-05		76	V-20 Ti	0.194	0.015	9.4	1182	0.77
	BE-01		76	V-20 Ti	0.195	0.015	9.4	1182	0.77
	BE-02		76	V-20 Ti	0.195	0.015	9.4	1182	0.77
	BE-03		76	V-20 Ti	0.195	0.015	9.4	1182	0.77
U-15 Pu-12 Zr	ND-23	XG06	65	V-20 Ti	0.209	0.016	10.8	1184	4.2
	ND-24	XG05	65	V-20 Ti	0.209	0.016	10.8	1184	5.6
	ND-25	XA07	74	304 SS	0.205	0.019	9.5	1144	4.0
	ND-26		74	316 SS	0.196	0.015	9.5	1121	4.0
	ND-27		73	304 SS	0.205	0.019	9.7	1154	4.1
	ND-29		72	316 SS	0.196	0.015	9.5	1132	4.0
	ND-30		72	316 SS	0.196	0.015	10.4	1168	4.4
	ND-31		74	316 SS	0.196	0.015	9.9	1155	4.2
	ND-33		73	Hast -X	0.196	0.015	9.9	1155	4.2
	ND-34		73	Hast -X	0.196	0.015	9.9	1161	4.2
	ND-35		75	Hast -X	0.197	0.015	10.4	1173	4.4
	ND-37		65	Hast -X-280	0.207	0.015	10.4	1170	4.4
	ND-39		65	Hast -X-280	0.207	0.015	10.2	1164	4.3
	ND-44		66	Hast -X-280	0.208	0.015	9.9	1152	4.2
U-15 Pu-10 Ti	NC-17	XG05	64	V-20 Ti	0.204	0.016	10.8	1184	5.6
	NC-23	XG06	68	V-20 Ti	0.204	0.016	10.8	1184	4.2

^aBased on analyses of neodymium or technetium

neutron radiography, which disclosed an average length increase of 1.9%. The irradiated fuel pins were not seated on the bottom closures of the cladding, but were elevated an average of 0.20 in. The elements had been intentionally assembled without fuel restrainers to observe possible axial fuel movement. The neutron radiographs revealed one or two partial axial separations, which varied from 0.02 to 0.06 in. in width, in most of the fuel pins within the cladding.

Measurements of fission-gas recovery showed that 60-71% of the fission gas had been released from the high-burnup fuel alloy to the plenum above the bond sodium. Detailed metallographic and microprobe examinations of the fuel/cladding interfaces showed reaction zones that had a maximum depth of 0.005 in. in the Hastelloy-X and the SA 304 stainless steel, and 0.001 in. in the SA 316 stainless steel.

Irradiation of a group of encapsulated U-15 Pu-10 Zr fuel elements clad with SA 304 and SA 316 stainless steel in EBR-II subassembly X028 (see Table XVIII) was terminated at 0.77 at. % burnup when capsule BC-02 and its contained element failed. It appeared that the capsule failed first, so the capsule bond sodium was removed and the fuel element overheated. Irradiation of this group of fuel elements was discontinued primarily because of concern regarding the weld quality of the remaining capsules. Postirradiation examination showed that in all elements the fuel had swelled into contact with the cladding.²⁶

Two U-15 Pu-10 Zr fuel elements clad with 0.016-in.-thick V-20 Ti alloy were irradiated in EBR-II subassemblies XG05 and XG06 to burnups of 5.6 and 4.2 at. %, respectively.²⁶ The peak linear heat rating was 10.8 kW/ft, and the maximum cladding temperature was 1184°F (640°C). Element ND-24, irradiated in subassembly XG05, released 57% of its fission gas into the element plenum. Microprobe analysis showed infiltration of sodium into the porous fuel, with local concentrations of 1-6 wt %.

A number of other metallurgical features of the U-Pu-Zr system are considered in Sec. V below. That discussion will consider the influence of fuel-cladding eutectic and solidus temperatures on high-temperature irradiation performance.

C. U-Pu-Ti Alloys

The U-Pu-Ti alloy system, like the U-Pu-Zr system, is potentially attractive because of the high melting points of alloys of interest. However, U-Pu-Ti alloys are not as compatible with stainless steel as U-Pu-Zr alloys. Only a limited number of irradiations were performed; all the fuel was clad in V-20 Ti alloy.¹ A prototype element was irradiated to 10.7 at. % burnup in a thermal-reactor irradiation,⁹ and two full-length elements were irradiated to burnups up to 5.6 at. % in EBR-II.²⁶

D. Conclusions on Burnup Potential

1. The high-burnup potential of properly designed LMFBR metal fuel elements is shown by the demonstrated capability of EBR-II Mark-II driver fuel to reliably perform at burnups greater than 10 at. %. This Mark-II design concept could be used with an LMFBR metal fuel such as U-Pu-Zr.

2. Successful EBR-II irradiations were made with stainless steel-clad U-Pu-Zr alloy fuel elements to burnups of ~4.5 at. %, which demonstrated the feasibility of this fuel system with peak cladding temperatures of 1200°F (649°C).

3. Information on irradiation performance is needed at high burnups (>4.5 at. %) to establish the burnup potential of the U-Pu-Zr metal fuels clad with 20%-coldworked Type 316 stainless steel and with advanced alloys such as PE16. The design and operating parameters for these irradiations should reflect guidance from the systems studies recommended in paragraph 2 of Sec. III.B.

V. THERMAL PERFORMANCE

A major consideration for evaluating applicability of fuels for LMFBR's is the maximum coolant temperature at which the fuel design will satisfactorily perform. Many early metal-alloy fuels have suffered from substantial limitations in this critical area. Of particular current interest are stainless steel-clad U-Pu-Zr alloy fuels, which are less thermally limited than any other stainless steel-clad plutonium-containing metallic fuel studied to date.

When thermal-performance limits for U-Pu-Zr fuels are considered, the following three areas must be addressed; these are discussed in detail later in this section.

1. Fuel/Cladding Compatibility. Of greatest importance is the temperature at which a fuel-cladding eutectic forms, resulting in an accelerated rate of cladding deterioration. Another important consideration is the rate of fuel/cladding interdiffusion that occurs below the eutectic temperature. This interdiffusion results in a gradual loss of effective cladding thickness with time, and may limit the burnup potential.

2. Fuel Melting. Because fuel melting may result in undesirable rapid changes in reactivity or in greatly accelerated cladding-penetration rates, it should be avoided during normal operation.

3. Fuel Slumping. The fuel should not slump significantly (shorten by creep under its own weight) during normal reactor operating conditions. The effect of fuel slumping on reactor safety is considered in Sec.VII. The U-Pu-Zr alloys, like most metallic fuel alloys, are compatible with sodium, and sodium is commonly used as a thermal bond between the fuel and the cladding. While this compatibility is taken for granted in metallic fuels, it is an important consideration in the use of other fuel types (e.g., oxides and carbides), particularly when running beyond cladding failure.

Two thermal considerations, not unique to metallic fuels, are the cladding mechanical strength and ductility and the cladding corrosion in sodium coolant. These are not discussed in detail in this report, but a brief statement on them is in order.

First, the strength and ductility of cladding must be adequate to withstand the stress/time history imposed by thermal, gas-pressure, and fuel-pressure stresses. This consideration is not unique to metallic fuels, but both the relative contributions of these three stresses and the cladding stress state may be unique.

Second, excessive wastage or detrimental change in important properties of the cladding by the flowing primary-sodium coolant must be avoided. Sodium corrosion can compromise cladding integrity as well as result

in excessive deposition of activated chemical elements from the cladding in the reactor regions outside the core (e.g., pumps, heat exchangers, etc.). This deposition can greatly increase difficulties in maintaining these components; but, again, it is not unique to metallic fuels.

A. Fuel/Cladding Compatibility

1. Fuel-Cladding Eutectic Temperature

One critical parameter for uranium-base alloy fuels clad with iron- or nickel-base alloys is the fuel-cladding eutectic temperature. Table XIX summarizes the available compatibility data for unirradiated U-Pu-Zr fuel alloys and compares them with data for other metallic fuel systems.* The important point to note is the high eutectic temperature ($1515 \pm 45^\circ\text{F}$) found for the combination of SA 304 stainless steel and U-Pu-Zr fuel containing 10 wt % or more of zirconium. This is well above any other metallic fuel-stainless steel eutectic temperature we are aware of (e.g., it is about 195°F above the eutectic temperature of the U-5 wt % F₂ and SA 316 stainless steel of the EBR-II Mark-II driver fuel). It is 180°F above the U/Fe eutectic temperature and 740°F above the Pu/Fe eutectic temperature. It is expected that the eutectic temperature for the U-Pu-Zr/Type 316 stainless steel system would be similar to that obtained with the U-Pu-Zr/SA 304 stainless steel system.

Future LMFBR designs may require low-swelling and stronger cladding materials if burnup limits are not to be restricted. Hence, the fuel-cladding eutectic temperatures of U-Pu-Zr fuel and a number of advanced cladding alloys are also presented in Table XIX. The compositions of these alloys are listed in Table XX. Although the alloys are not ones currently being considered in the advanced-cladding-alloys programs, for the purpose of compatibility studies their compositions can be considered representative of advanced cladding alloys. Except for the high-nickel-content Incoloy 800 and Hastelloy-X alloys, the listed alloys exhibit essentially the same fuel-cladding eutectic temperatures as those with SA 304 stainless steel.

The irradiation in EBR-II (in subassembly XA07) of U-15 wt % Pu-10 wt % Zr fuel clad with SA 304L stainless steel, with SA 316 stainless steel, with Hastelloy-X, and with Hastelloy-X-280 to maximum burnups of 4.5 at. % and at maximum cladding temperatures of 1200°F showed no formation of the fuel-cladding eutectic.⁸ This provides some confirmation of the out-of-reactor results of Table XIX, but does not give the required information on actual eutectic temperatures for irradiated fuel and cladding.

It is reasonable to ask what rates of cladding penetration would occur if the fuel/cladding interface temperatures exceed the fuel-cladding eutectic temperatures. Exceeding the eutectic temperature is not acceptable under normal operating conditions, because the rates are too rapid. For off-normal conditions (mild overpower or reduced flow), however, the eutectic temperatures can be exceeded for short times without cladding failure. Because

*The alloy designated F₂ is identical to the alloy F₁, except that it has a slightly higher zirconium content.

TABLE XIX. Fuel-Cladding Eutectic Temperatures (°F)

Fuel Alloy, wt %	Cladding Material								
	304 SS	316 SS	Fe	Timken 16-15-6 ^a	Timken 16-25-6 ^a	Haynes 56 ^a	N-155 ^a	Incoloy 800 ^a	Hastelloy-X ^a
C-15 Pu-10 Zr ²⁷	1515 ± 45	-	-	1515 ± 45	<1560	1515 ± 45	<1560	1430 ± 45	1335 ± 45
U-16.6 Pu-6.3 Zr ²⁷	1335 ± 45	-	-	-	-	-	-	-	1335 ± 45
U-18.5 Pu-14.1 Zr ²⁷	1515 ± 45	-	-	1535 ± 30	<1560	1515 ± 45	1515 ± 45	1515 ± 35	1335 ± 45
U-15 Pu-10 Ti ²⁷	<1290	-	-	-	-	-	-	-	-
U-5 Fe ²⁸	1300 ± 10	1320 ± 10	-	-	-	-	-	-	-
U-15 Pu-10 Fe ²⁹	1220 ± 25	-	-	-	-	-	-	-	-
Uranium (unalloyed) ³⁰	-	-	1335 ± 5	-	-	-	-	-	-
Plutonium (unalloyed) ³¹	-	-	775 ± 5	-	-	-	-	-	-

^aCompositions are given in Table XX.

TABLE XX. Composition of Advanced Cladding Alloys

Alloy	Composition, wt %						
	Fe	Ni	Cr	Mo	Co	W	Mn
Timken 16-15-6	55	15	16	6	-	-	7.5
Timken 16-25-6	50	25	16	6	-	-	1.3
Haynes 56	46	13	21	4.5	11.5	1.5	1.5
N-155	30	20	21	3	20	3	1.5
Incoloy 800	46	32	21	-	-	-	0.75
Hastelloy-X	18	48	22	9	1.5	0.6	1.0

no information has been generated on cladding penetration above the eutectic temperature for stainless steel-clad U-Pu-Zr fuel, we will attempt to infer its behavior from results obtained on the U-5 wt % Fe/SA 304 stainless steel system.³²⁻³⁴ These studies have shown that, at temperatures slightly above the melting point of uranium or the liquidus of uranium-fissium, U-Fe alloys will penetrate stainless steel or iron at the rate of about 10 mils/sec. Reference 34 also explored the maximum penetration rates of uranium through iron at temperatures between the uranium-iron eutectic temperature and the uranium melting point. The following penetration rates were found: 0.01 mil/min at 1365°F, 0.76 mil/min at 1565°F, 3.1 mils/min at 1745°F, and 8.5 mils/min at 1925°F. It is not unreasonable to assume that the penetration rates of U-Pu-Zr alloys through Type 316 stainless steel would be of the same order when their eutectic temperature of ~1515°F is exceeded. Therefore, exceeding the eutectic temperature of stainless steel-clad U-Pu-Zr fuels by as much as 200°F for short times (of the order of 1 min) does not imply gross fuel-element failures due to eutectic formation.

2. Solid-state Interdiffusion between Fuel and Cladding

Extensive penetration of fuel components into the cladding or loss of cladding components into the fuel below the eutectic temperature could compromise the ability of the cladding to contain the fuel at high burnup. Table XXI summarizes available results of out-of-pile fuel-penetration tests below the fuel-cladding eutectic temperature for U-Pu-Zr fuel alloys and compares them to results obtained with other metallic fuel systems. The data presented in this table are only for stainless steel. Much more limited penetration data (usually single points at short times) were generated on the advanced alloys listed in Table XX. These data generally showed penetration rates similar to those obtained for SA 304 stainless steel, except that the high-nickel alloys showed higher penetration rates. Although the effect of fuel penetration on the mechanical properties of the stainless steels in Table XXI is not really known, a relative comparison is possible. For example, the penetration at 1300°F of the U-Pu-Zr alloys into SA 304 stainless steel ranges from 15 to 20% of that found for the U-5 Fe system. Hence, there is some improvement with the

TABLE XXI Out-of-pile Solid-state Penetrations of Fuels into Stainless Steel Cladding in One Year (mils)^a

Fuel-Cladding Combination, wt %	Temperature, °F				
	1100	1200	1300	1400	1500
U-15 Pu-10 Zr-304 SS ²⁷	-	-	0.4	0.9	4.6 ^b
U-18.5 Pu-14.1 Zr-304 SS ²⁷	-	-	0.3	0.6	1.4
U-5 Fe-304 SS ²⁸	0.4	0.6	2.0	Eutectic	Eutectic
U-5 Fe-316 SS ²⁸	0.4	0.9	3.0	Eutectic	Eutectic
U-15 Pu-10 Fe-304 SS ²⁹	-	2.6 ^b	Eutectic	Eutectic	Eutectic
U-10 Pu-10 Fe-304 SS ²⁹	0.7	1.5	Eutectic	Eutectic	Eutectic

^aAll penetrations followed an $x^2 = kt$ law, where x is the penetration distance, k is penetration coefficient, and t is the time. From this relationship, penetrations for times other than one year can be calculated from the penetrations given in the table.

^bCalculated from only one data point, assuming $x^2 = kt$.

ternary alloy over a fuel system used as the EBR-II driver fuel, which has shown good burnup potential (see Sec. IV). The significant conclusion that can be drawn from Table XXI is that, in the absence of irradiation, fuel/cladding interdiffusion in the U-Pu-Zr/stainless steel system is not a significant problem at anticipated LMFBR temperatures.

Of additional interest in the metal-fuels program is the diffusion of stainless steel cladding components into the uranium-base fuel. Nickel and, to some extent, iron diffuse out of the austenitic cladding into the fuel, thereby increasing the chromium content in the interdiffusing region of the cladding. With the loss of the austenitic stabilizer, nickel, the austenitic structure transforms to ferrite and sigma phases. This embrittling structure can lead to cracking upon thermal cycling. Interdiffusion bands of ~3 mils are not uncommon for SA-304L-stainless-steel-clad experimental Mark-II driver fuel (12-mil cladding thickness) prior to failure in EBR-II. This observation may be compared with the following review.

Some data on the effect of irradiation on interdiffusion between U-15 wt % Pu-10 wt % Zr fuel and stainless steel are available from the irradiation of fuel elements in EBR-II experiment XA07.⁶ These elements were operated at maximum fuel/cladding-interface temperatures of 1200°F to a burnup of 4.5 at. %. This burnup was equivalent to six months of full-power operation. Table XXII summarizes the cladding penetrations observed during postirradiation examination of these elements. In addition, the table includes in-reactor penetrations extrapolated to one year using both $x^2 = kt$ (the usual penetration-rate law) and $x = kt$ (the law for the most rapid penetration rate seen in- or out-of-reactor), and extrapolated out-of-reactor penetrations extrapolated using $x^2 = kt$.

TABLE XXII Comparison of In-reactor and Out-of-reactor Penetrations of Stainless Steel Cladding at 1200°F (mils)

Fuel-Cladding Combination wt %	In-reactor			Out-of-reactor
	Actual in 6 mo	Extrapolated (x ² = kt) to 1 yr	Extrapolated (x = kt) to 1 yr	Extrapolated (x ² = kt) to 1 yr
U-15 Pu-10 Zr 304 SS	5.5	7.8	11.0	0.2
U-15 Pu-10 Zr 316 SS	1.0	1.4	2.0	-
U-18 Pu-14 Zr 304 SS	-	-	-	0.1
U-5 Fs 316 SS ^a	-	0.1 ^b	0.1 ^b	0.4
U-5 Fs 304 SS ^a	-	1.0 ^b	1.0 ^b	0.4

^aPenetrations at 1100°F

^bActual measured 1-year penetration

Two important points are noted from the data. First, the in-reactor penetration rate for the SA 316 stainless steel is much less than that for the SA 304 stainless steel. Irradiation results for EBR-II driver fuel indicate that SA 316 stainless steel is also more compatible than SA 304 stainless steel with U-5 wt % Fs at temperatures up to 1100°F.¹⁴ Although the difference in compatibility is not as great as seen in the irradiation results for U-Pu-Zr, it is significant. A difference in SA 316 stainless steel and SA 304 stainless steel was not seen for out-of-pile results on U-5% Fs (see Table XXII). A second and even more significant observation to be made for the U-Pu-Zr fuel is the much greater cladding penetration in-reactor than out-of-reactor. For the U-5 wt % Fs/SA 304 stainless steel combination, on the other hand, the cladding penetration (at ~1100°F) was only slightly greater in-reactor than out-of-reactor. Additional irradiation data at higher cladding temperatures for longer times are essential if U-Pu-Zr clad with stainless steel is to be a viable LMFBR fuel element. Nevertheless, the modest in-reactor penetration (~1 mil in six months) of SA 316 stainless steel cladding indicates that U-15 wt % Pu-10 wt % Zr clad with SA 316 is a satisfactory high-burnup element, from the compatibility viewpoint, at maximum cladding temperatures up to ~1200°F.

3. Compatibility Variables

The high eutectic temperatures and low interdiffusion rates exhibited by U-Pu-Zr alloys with SA 304 stainless steel may be influenced by the oxygen levels in the fuel and cladding. Electron-microprobe analyses made on certain annealed diffusion couples indicated that compatibility may be improved by the formation in the fuel of diffusion layers consisting of one or more zirconium-rich phases stabilized by oxygen.²⁷ These phases may restrict the transport of iron and nickel into the fuel as well as diffusion of uranium and plutonium into the stainless steel. The presence of oxygen at some level in the cladding and fuel alloys may therefore be beneficial, if not essential, for obtaining good compatibility in U-Pu-Zr/stainless steel systems. It is important that this point be pursued further.

A most interesting feature of this fuel alloy system is that, except for oxygen, the starting composition may not be as important for cladding compatibility as the out-of-pile studies indicate.

Fig. 7. Irradiation Zones and Diffusion Couples for U-Pu-Zr System. ANL Neg. No. 103-89099.

During irradiation, when the fuel-centerline temperatures reach $\sim 1380^{\circ}\text{F}$, the fuel exhibits three distinct metallurgical zones caused by compositional (phase) changes⁸ that are due to the temperature gradient from the center to the outside of the fuel pin and to irradiation. These zones, schematically represented in Fig. 7, may be physically separated.

Table XXIII lists the composition ranges along the fuel-pin lengths for these zones after the EBR-II irradiation of U-Pu-Zr alloys 4 and 5 (see Sec. IV.B) to 4.5 at. % burnup.⁸ Comparison of these

compositions with the solidus curves of Fig. 8 shows that the high zirconium content of the outer

zone, adjacent to the cladding, would lead to an outer-zone solidus temperature higher than 2190°F ; this may result in beneficial compatibility effects. The middle zone has a solidus temperature below 1830°F , while the center zone, with its higher zirconium content, again exceeds the 2190°F solidus temperature.

Beta-gamma autoradiographs⁸ show that active fission products diffuse into the outer and center zones in a similar manner to the natural and stable isotopes of zirconium. There is very little net radial movement of the

TABLE XXIII. Mean and (Range of) Atomic Percentages of Plutonium, Zirconium, and Uranium in Irradiation Zones of Alloys 4 and 5 after 4.5 at. % Burnup⁸

	Outer Zone	Middle Zone	Center Zone
Pu	12.5 (10-15)	10 (8-12)	12.5 (11-14)
Zr	37.5 (25-50)	6 (2-10)	48.5 (42-55)
U	50.0 (35-65)	84 (78-90)	39.0 (31-47)

Starting Composition of Alloys 4 and 5

Pu	12.3	(11.6-12.9)
Zr	23.0	(20.4-26.1)
U	64.7	(58.2-66.7)

Fig. 8. Solidus Lines for U-Pu-Zr Alloys 1-5 as a Function of Atomic Fractions of Zirconium and Plutonium. (Small circles indicate starting compositions.) ANL Neg. No. 103-B9097.

plutonium (see Table XXIII). At lower fuel-centerline temperatures (i.e., at the bottom of the fuel column), the center zone, which is all gamma phase, is not produced, and the outer and middle zones are wider. Other investigators³⁵ have noted the same diffusion phenomenon out-of-pile in an axial temperature gradient.

B. Fuel Melting

Table XXIV summarizes the available information on U-Pu-Zr melting points (solidus and liquidus) and compares them with melting points of other metallic fuels. The solidus temperatures for the U-Pu-Zr alloys are well above those for previously considered U-Pu alloys such as U-Pu-Fs and even exceed the melting point of uranium. The U-15 wt % Pu-10 wt % Zr alloy has a solidus temperature of $\sim 2110^{\circ}\text{F}$, more than 250°F above the solidus temperature of U-5 wt % Fs (the EBR-II driver fuel). The U-Pu-Ti alloys also have high solidus temperatures, but are not compatible with stainless steels above $\sim 1300^{\circ}\text{F}$.

TABLE XXIV. Melting Points of Metallic Fuels

Fuel Alloy, wt %	Melting Point, °F	
	Solidus	Liquidus
U-11.1 Pu-6.3 Zr ²⁵	~2050	~2190
U-15 Pu-6.8 Zr ²⁵	2020 ± 20	2260 ± 40
U-15 Pu-10 Zr ²⁵	~2110	~2280
U-15 Pu-13.5 Zr ²⁵	2180 ± 20	2600 ± 40
U-18.5 Pu-14.1 Zr ²⁵	~2140	~2350
U-15 Pu-10 Ti ²⁵	~2190	~2440
U-5 Fe ¹⁶	1836 ± 10	1978 ± 10
U-15 Pu-10 Fe ²⁵	~1590	~1830
Uranium (unalloyed) ¹⁷	2070 ± 2	2070 ± 2
Plutonium (unalloyed) ¹⁸	1185 ± 5	1185 ± 5

As was shown in Fig. 8, the solidus temperature of the U-Pu-Zr system increases with increasing zirconium content for the ternary compositions considered in this report. As can be seen, the weight percents involved translate into rather high atomic percents. Therefore, some work should be done to optimize the initial zirconium content of the alloy (i.e., minimizing zirconium content while retaining acceptably high solidus temperatures and good compatibility with stainless steel).

The effect of fission-product buildup during irradiation on the U-Pu-Zr solidus temperature has not been investigated, but on the basis of data on U-Pu-Fe alloys in Ref. 25, we might expect it to be lowered somewhat. Additional effort is required with regard to this aspect of melting.

C. Fuel Slumping

Shortening of fuel by creep under its own weight (slumping) has been of traditional interest in metallic fuels. Some out-of-reactor information has been generated on compressive creep of U-18.5 wt % Pu-14.1 wt % Zr in the 1100-1300°F temperature range.²⁵ Based upon solidus temperature, the creep of U-15 wt % Pu-10 wt % Zr would probably be slightly higher. Figure 9 uses this data to extrapolate the time for the former fuel composition to compressively creep (slump) 1% at 1300, 1500, 1700, and 1900°F. For the assumed 36-in.-long fuel pin, the stress is 20 psi at the bottom of the pin (where the temperature is lowest), 10 psi at the core midplane, and 5 psi one-quarter of the way down from the top of the pin (where the fuel temperature approaches its highest value)

For an assumed conservatively high mean fuel temperature [i.e., 0.5 (fuel-surface temperature + fuel-centerline temperature at high burnup)] of about 1700°F at the top of the pin, 1500°F at the core midplane, and 1300°F at

Fig. 9. Compressive Creep of U-19.5 wt % Pu-14.1 wt % Zr (36-in.-long fuel column). ANL Neg. No. 103-B9009.

the bottom of the pin, the time to compressively creep 1% at the various locations in the pin is represented by the small circles in Fig. 9. Overall pin shortening is an integral of these values, so that 1% slumping might be expected to occur in about 6000 hr (250 days). This is about a factor of 10 less than the normal lengthening expected due to fuel swelling during this irradiation period. In other words, although irradiation may increase the slumping rate somewhat, fuel slumping does not appear to be of any significant concern. It has been shown experimentally that, at the fuel-swelling values obtained at high burnup, interconnection of swelling porosity occurs accompanied by sodium infiltration and a significant increase in thermal conductivity.¹⁹ Therefore, the temperatures used here certainly represent an overestimate of fuel-centerline temperature.

A note is in order on the method¹⁵ used to generate Fig. 9. The data were generated at stress levels more than a factor of 10 greater than those that interest us, for short times (<100 min), and over a narrow temperature range (1250-1300°F). Therefore, the extrapolations to generate Fig. 9 are large.

These extrapolations were performed using conventional high-temperature creep relationships $[(\text{time to 1\% creep})^{-1} = (\text{constant}) \sigma^n \exp(-Q/RT)]$ and should be appropriate for the temperatures of interest.

D. Conclusions on Thermal Performance

1. Out-of-reactor results indicate that U-15 wt % Pu-10 wt % Zr is compatible (i.e., has an acceptable cladding-penetration rate and no eutectic formation) with SA 304 stainless steel and a number of high-strength advanced base alloys to temperatures approaching 1500°F. Information needs to be generated on use of U-15 wt % Pu-10 wt % Zr with Type 316 stainless steel. Oxygen content of the fuel and cladding may have some effect on compatibility and requires further investigation.

2. In-reactor results to 4.5 at. % burnup indicate that U-15 wt % Pu-10 wt % Zr is compatible with SA 316 stainless steel (i.e., has an acceptable cladding-penetration rate and no eutectic formation) at 1200°F. Irradiations at higher temperature and to greater burnups need to be conducted to define time-temperature limits.

3. In-reactor results to 4.5 at. % burnup indicate that, at 1200°F, U-15 wt % Pu-10 wt % Zr penetrates SA 304 stainless steel at about five times the rate it penetrates SA 316 stainless steel. This observation is inconsistent with out-of-reactor results on other fuel systems and requires further investigation via irradiation tests. The difference may be due to factors other than the inherent differences between the two stainless steels (e.g., oxygen content of the cladding).

4. Future compatibility tests and irradiations should factor in the presence of fission products consistent with the levels expected with high burnup and fuel reprocessing.

5. The solidus of U-15 wt % Pu-10 wt % Zr alloy is sufficiently high (~2100°F) that fuel melting can be easily avoided during normal reactor operation, even assuming rather high sodium outlet temperatures. The effect of fission-product buildup (particularly fissium elements) on the fuel solidus temperature should be determined.

6. Although data on fuel slumping are scarce, slumping under normal operating conditions (and probably even severe transients to near the fuel solidus temperature) appears to be of no significant concern.

7. The adequacy of Type 316 stainless steel cladding (cold-worked or annealed) and advanced alloys to contain U-Pu-Zr fuel with acceptable cladding distortions at high burnup (~10 at. %) can be demonstrated only by additional fuel-element irradiations.

VI. FUEL FABRICABILITY

The ease of fabrication of a given fuel design is an important factor in determining the practical applicability of that design to future LMFBR's. Fuel-fabrication processes must be available for both initial core loadings and re-cycled fuel. This section of our report summarizes the fabrication processes used on U-Fs EBR-II fuels and includes the results obtained with these processes. It also summarizes the available information on U-Pu-Zr fabrication.

For U-Fs fuel, the same basic processes were used for fabrication of unirradiated fuel and for remote refabrication of irradiated fuel. These same basic processes can probably be used on unirradiated and irradiated U-Pu-Zr alloys; however, some alternative approaches will also be discussed for U-Pu-Zr.

A. Fabrication of U-5 wt % Fs EBR-II Fuels

A flowsheet for the fabrication of irradiated or unirradiated U-Fs EBR-II fuel is shown in Fig. 10. An inert-atmosphere, shielded alpha-gamma cell is used only for processing and fabrication of irradiated fuels that have experienced a low fission-product-decontamination factor (i.e., fuels from which fission products have not been completely removed). Although the processes used for fabrication of unirradiated^{40,41} and irradiated⁴² EBR-II fuels are well documented, we will briefly summarize them.

For unirradiated fuel, an alloy billet is prepared by direct melting of uranium with molybdenum, ruthenium, rhodium, palladium, niobium, zirconium, and silicon alloying elements in a stabilized zirconia crucible at ~2500°F for about 2 hr and casting in a graphite mold coated with zirconia, yttria, or thoria. For irradiated fuel, the alloy billet results from the melt-refining step (see Sec. VII.A). For either process, the billet is then charged into a graphite crucible coated with zirconia, yttria, or thoria in the injection-casting furnace. The injection-casting furnace consists of a vacuum-pressure chamber in which a number of precision-bore mold tubes are suspended vertically, open end down, above the crucible in which the fuel is melted. The furnace and the molds are evacuated during the melting cycle. The molds are lowered to immerse the mold ends, and the furnace is rapidly pressurized with inert gas. The pressure on the surface of the metal in the crucible causes the evacuated molds to fill. As soon as the metal in the molds solidifies, the molds are raised.

High-silica glass is used as the mold material because it can be formed to satisfactory tolerances and it withstands the thermal shock of casting at ~2550°F. One end of the mold tube is flattened and fused closed; the opposite end is cut at a 60° angle to the axis to aid in penetrating the melt surface and to allow excess metal to drip from the end of the mold. Reaction between the silica and the fuel alloy is prevented by a thin coating of zirconia, thoria, or yttria. Experience with the EBR-II injection-casting furnaces, each of which melts 10-17 kg of alloy and casts as many as 160 fuel pins at a time, indicates

Fig. 10. Fabrication Scheme for Metallic Fuel. ANL Neg. No. 103-B9096.

that the equipment can be scaled up to melt 70-kg charges to cast about 350 pins at a time. Pressurized inert gas is circulated between the molds for rapid cooling.

A residual heel amounting to about 20% of the charge weight is left behind after casting. This heel is usually removed from the crucible after solidification, broken, and recharged into succeeding melts without significant loss.

Demolding is accomplished by crushing the glass away from the fuel pin. The fuel pins are deliberately cast overlength, and segments are cropped from the top and bottom by shearing. The end shards are collected and returned either to the melt-refining or the injection-casting operation. No further mechanical work is required to finish the fuel pins. The pins are inspected for weight, length, diameter, and porosity using automated inspection equipment.

High-purity sodium to thermally bond the pin to the jacket is loaded as a solid wire segment into the previously fabricated jackets. After sodium-loading, the fuel pins are inserted into the jackets, and an end plug is welded in place using capacitive-discharge techniques. The plug-to-jacket weld is leak-tested by a pressure-decay method, and the elements are then ready for the sodium-bonding operation. Sodium-bonding is effected by heating the fuel elements to $\sim 950^{\circ}\text{F}$, holding for about 1 hr, and impacting ~ 1000 times.

Final element inspection includes eddy-current interrogation for sodium-bond quality and sodium level, visual examination for handling damage, and dimensional checks.

Table XXV summarizes the types and quantities of EBR-II U-Fs fuel elements produced as of June 1975 by the basic processes described above. It also differentiates between fabrication of unirradiated fuel and refabrication of irradiated fuel. Well over 100,000 metallic EBR-II fuel elements have been manufactured from both irradiated and unirradiated material in essentially pilot-plant manufacturing facilities. Only one failure of a U-Fs fuel element during the 10 yr of EBR-II operating experience was traceable to a fuel-element manufacturing defect. This one failure was due to a faulty lower weld in a Mark-IA element.

TABLE XXV. EBR-II U-5 wt % Fs Fuel Elements
Produced as of June 1975

Fuel Design	Unirradiated	Irradiated
Mark I and Mark IA	59,000	35,000
Mark II	22,000	0

B. Fabrication of U-Pu-Zr Fuels

Injection casting appears to be a feasible approach to fabricating U-Pu-Zr alloys, although other processes (e.g., extrusion) should also be considered. Small numbers (~50) of U-Pu-Zr fuel pins have been manufactured by injection casting for the experimental irradiations described previously. If the injection-casting process is assumed applicable, then the flowsheet used for U-Fs alloys (Fig. 10) can be used without change. It is appropriate, however, to examine fabrication processes within this flowsheet that would have to be modified from those used for U-Fs. Although not much work has been done on U-Pu-Zr, the principal problems appear to be associated with obtaining homogeneous alloy billets and cast fuel pins.

1. Preparation of Homogeneous Alloy Billets

Workers at LASL have had considerable success in preparing homogeneous cast billets by mechanical stirring of the melt and bottom-pour chill casting.³⁵ Two ceramic crucible coatings that could be applied to standard bottom-pour graphite crucibles were developed so that conventional alloying and casting techniques could be used. The coating more inert to the U-Pu-Zr alloys was NbC applied directly to the graphite surface. The amount of niobium dissolved into the melt was reported to be less than 25 ppm, and the oxygen pickup was negligible. The second coating, a Y_2O_3 coating codeveloped by LASL and Acheson Colloids Company, consisted of a water suspension of Y_2O_3 with a K_2SiO_3 binder added. When properly applied, this coating was unwetted by the U-Pu-Zr melt, and the graphite crucible could be reused. Alloy billets prepared in Y_2O_3 -coated crucibles picked up about 100 ppm of both silicon and oxygen.

Alloying was accomplished by inductively co-melting the metals (uranium, plutonium, and zirconium) under vacuum in the coated graphite crucible. The molten metals were heated to 2550-2700°F and mechanically stirred for 30 min to ensure dissolution of zirconium into the melt. The stopper rod was raised, and the melt was chill-cast into an aluminum mold at ambient temperature. Table XXVI summarizes the chemical analysis of some of these cast alloys. This approach was effective in minimizing axial variations in zirconium content; however, as is typical for inverse segregation, the variations in radial composition were as great as 0.8 wt % Zr.

TABLE XXVI. Chemical Analysis of Chill-cast U-Pu-Zr Alloys³¹

Element	Composition, wt %					
	Top	Bottom	Top	Bottom	Top	Bottom
Zr	6.8-7.4	6.6-7.0	10.1-10.4	9.9-10.2	13.3-14.1	13.4-13.6
Pu	14.8	14.9	15.2	15.1	14.6	14.6
U	78.1	77.9	75.4	74.7	72.3	72.0

Alloy preparation at ANL was less successful without mechanical stirring and required extensive use of master alloys to get zirconium into solution at temperatures low enough to prevent unacceptable reaction of the melt with the crucibles.⁴³

2. Injection Casting

ANL workers experienced some difficulty in injection-casting U-Pu-Zr alloys.⁴³ This difficulty was in large part due to the rather high injection-casting melt temperatures required, e.g., ~2800° F for U-15 wt % Pu-14 wt % Zr. This temperature approaches the softening temperature of the Vycor casting molds. Because U-15 wt % Pu-10 wt % Zr has a lower liquidus temperature and some adjustment in casting parameters is feasible, it should be possible to injection cast at somewhat lower melt temperatures. Nevertheless, the ANL workers were able to successfully cast 0.144- and 0.166-in.-dia fuel pins with acceptable homogeneity in up to 13-in. lengths, using U-15 wt % Pu-14 wt % Zr and U-15 wt % Pu-10 wt % Zr. It appears that, with some additional fabrication development work (perhaps using the LASL billet-preparation technique, together with a refinement of the ANL injection-casting technique), the process outlined in Fig. 10 could be applied to both irradiated and unirradiated U-Pu-Zr alloys.

3. Alternative Processes for Fuel-pin Fabrication

Even though injection casting of U-Pu-Zr has been demonstrated on a small scale, other pin-forming processes should not be overlooked. LASL has done some development on extrusion and swaging of U-Pu-Zr into fuel pins.³⁵ Of these two techniques, extrusion showed by far the greatest promise. LASL's studies indicated that bare extrusion at ~1000° F of 0.75-in.-dia chill-cast billets down to 0.144-in.-dia fuel pins with a 100- to 150-ton press should present no particular difficulty. The problem with this approach is twofold, however.

a. The remote operation and maintenance of an extrusion press in a hot-cell environment for refabricating irradiated fuels will be difficult at best.

b. Considerable effort would be required to determine whether such fuel could be properly heat-treated to remove fabrication-induced texture (preferred orientation). Very small degrees of preferred orientation have been shown to result in rather substantial irradiation growth in metallic fast-reactor fuels.⁴⁴

One final obvious difference exists between fabricating U-Fs and U-Pu-Zr alloy (irradiated or unirradiated): the requirement for adequate containment of plutonium to protect personnel.

C. Conclusions on Fabricability

1. Based upon experience with EBR-II U-Fa fuels, an extension of existing technology to the fabrication of irradiated and unirradiated U-Pu-Zr alloys does appear feasible.

2. Such an extension in technology would require additional development studies on producing homogeneous U-Pu-Zr fuel alloys and acceptable injection-casting yields in quantities consistent with the fuel needs of a reactor.

3. The feasibility of extrusion or other alternative pin-fabrication methods for producing large quantities in a remote-refabrication facility should be evaluated. If one or more of the methods seem feasible, development studies on the fabrication and the heat treatment to eliminate texture would be required.

VII. FUEL REPROCESSING

Before a fuel becomes a serious contender for the LMFBR program, a high degree of confidence must exist that it can be reprocessed in a practical and economical manner. Without a good fuel-reprocessing scheme, much of the justification for breeders is lost.

This section of our report briefly (1) describes past ANL experience with reprocessing irradiated stainless steel-clad U-5 wt % F_s EBR-II driver fuel and (2) considers the feasibility of future reprocessing of irradiated U-Pu-Zr fuels clad in stainless steel. Our discussions consider remote pyrochemical (pyrometallurgical) and solvent-extraction reprocessing as well as remote refabrication of irradiated fuels in a facility closely coupled with a reactor(s). The advantages of this close coupling are: (1) short cooling periods that allow for rapid return of fuel to the reactor (an especially important consideration in breeder reactors), and (2) greatly reduced need for off-site shipping of special nuclear materials and radioactive waste products.

A. Reprocessing of U-5 wt % F_s EBR-II Driver Fuel

The development and application of remote pyrochemical (pyrometallurgical) reprocessing and remote refabrication of U-5 wt % F_s EBR-II driver fuel are well-documented.^{42,45,46} The reprocessing begins about 14 days following a reactor shutdown and consists of the following steps: (1) transfer of the 91-element subassemblies to the reprocessing facility, (2) disassembly of the subassemblies into individual elements, (3) mechanical decladding of the elements, and (4) chopping of the declad fuel pins for charging into the lime-stabilized zirconia melt-refining crucible.

Melt refining (see Fig. 11) is conducted at ~2550°F for 1-3 hr, after which the refined melt is chill-cast into a mold. During the melt-refining step, electropositive fission products (mainly alkaline-earth and rare-earth fission products) react with the zirconia crucible to form oxides, which deposit along the wetted crucible surfaces. The fission-product gases (xenon and krypton) are released to the furnace, from which they are pumped (through a charcoal trap that removes traces of iodine) to a large tank where they are stored. Condensable fission products (mainly cesium and iodine) are collected within the furnace on a Fiberfrax fume trap above the crucible.

The refined ingot, containing molybdenum, ruthenium, rhodium, palladium, niobium, technetium, and zirconium fission products (fissium), is removed from the mold and formed into new fuel pins by injection casting, a process in which the molten fuel is forced by gas pressure into precision-bore Vycor tubes. Following injection casting, the new fuel pins are cut to length, inspected, and inserted into stainless steel jackets together with a sufficient amount of sodium to bond the pins to the cladding. The fuel elements (jackets plus pins) are then seal-welded closed, leak-checked, bonded, inspected, and reassembled into 91-element subassemblies. The new subassemblies are then transferred back to the reactor for irradiation.

APPROXIMATE DISTRIBUTION OF ELEMENTS IN MELT-REFINING PROCESS

Fig. 11. Fission-product Removal in Melt Refining of EBR-II U-Fs Fuel. ANL Neg. No. 103-B9071.

The entire reprocessing and refabrication process is performed remotely and has been routinely accomplished in 45 days or less in the EBR-II Fuel Cycle Facility. This facility is close-coupled to the EBR-II reactor, and during 1964-1969 produced 35,000 reprocessed and refabricated fuel elements for the reactor. In 1969, the mission of the EBR-II reactor was changed from a prototype demonstration reactor with closed-cycle fuel reprocessing to a fast-neutron irradiation test facility. This change in mission for the reactor dictated a mission change for the Fuel Cycle Facility to an examination facility for experiments irradiated in EBR-II.

Molybdenum, ruthenium, rhodium, palladium, niobium,

and technetium are not removed during melt refining, and zirconium is only partially removed. The aggregate of these elements (fissium) therefore increases in the refined material at the rate of about 0.35 wt % Fs per at. % burnup. This buildup in fissium content is controlled by a side stream called "dragout." Suppose a fuel initially containing 5 wt % Fs is irradiated to 1 at. % burnup. The fissium content would then increase to 5.35 wt % Fs, about a 7% increase. Simply stated for this example, dragout is the intentional side streaming of 7% of the refined material to another process for quantitative removal of fissium elements. Uranium with the proper enrichment is then added to the remaining 93% to bring the refined ingot up to 100% of its original weight, resulting in an adjusted fissium composition of 5 wt % Fs.

In actual practice, the melt-refining step results in an ingot plus a skull (oxidized dross and some unpoured metal) which remains in the zirconia crucible. This skull accounted for 5-10% of the melt-refining charge, so no additional dragout was required for fuels having 1-2 at. % burnup.

Before discussing the process used for reclamation of skulls, we need to consider the effects of burnups above 2 at. % on the dragout requirements. First, assume a 0.35 wt % Fs increase per at. % burnup and a desirable 5 wt % Fs content at the beginning of each burnup cycle. Further assume a burnup limit of 10 at. %, which seems to be a reasonable goal based upon results of irradiating EBR-II Mark-II fuel. At 10 at. % burnup, the fissium content will increase

from 5 to 8.5 wt % (i.e., a 70% increase in fission content). Therefore, as a first approximation, 70% dragout is required. The implications of this are important; even if the desired starting fission content is increased to 10 wt % F_s, 35% dragout is required. Therefore, either two major processes (melt refining and dragout refining) are required to reprocess high-burnup U-Fs fuels, or a single process must be developed to remove the fission elements.

A skull-reclamation process for EBR-II U-Fs fuel (see Fig. 12) was developed and tested on a pilot scale,⁴⁷ but was never installed and made operational in the EBR-II Fuel Cycle Facility. In the process, liquid-metal and molten-salt media are used, and solubility differences and selective oxidation-reduction reactions are applied to make possible the necessary separations of uranium from the fission elements. The solvent-metal phases are zinc-magnesium alloys, and the salt phase is a mixture of alkaline-earth halides (47.5 mol % MgCl₂, 47.5 mol % CaCl₂, and 5.0 mol % MgF₂ or CaF₂). This approach was established to process skulls from low-burnup fuel, which accounts for 5-10% of the melt-refining charges. Its applicability to 35 or 70% dragout on high-burnup fuels appears impractical, because the equipment and the quantity of process wastes become prohibitively large. In addition, it is limited to uranium fuels, since it is incapable of separating plutonium from rare earths. This separation would be required for plutonium-bearing fuels such as U-Pu-Zr.

As an alternative, a combination of melt-refining and solvent-extraction processes could have significant advantages. The melt-refining step would then serve as a head-end step for the subsequent solvent-extraction process. Thus, about half the spent fuel would be available for immediate recycling (after suitable alloy adjustment). The melting operation would also serve to remove fission-product gases (xenon, krypton, and tritium) and volatile fission products (cesium and iodine) from the half of the fuel that goes to solvent extraction. (These fission products are a major problem in the application of solvent extraction to reprocessing of any kind of high-burnup, short-cooled fuel.) An additional benefit would be a reduction in gross beta-gamma activity, which contributes to solvent damage.

B. Reprocessing of U-Pu-Zr Fuels

We are not aware of any information on the reprocessing and refabrication of U-Pu-Zr alloys. However, preliminary studies have been conducted on pyrochemical reprocessing of U-Pu-Ti fuels.⁴⁸ A process similar to that suggested for U-Pu-Ti would be appropriate for U-Pu-Zr. Figure 13 presents a flowsheet for reprocessing U-Pu-Ti fuel together with a uranium blanket. This process resembles that used successfully for EBR-II U-Fs fuel in that it refines irradiated fuel by use of melting, molten-salt media, and solvent-metal phases. The flowsheet does not represent the only selection of pyrochemical steps that would satisfy the processing requirements. Alternative process steps require study, however, but some of them are expected to lead to more economical processing. The flowsheet of Fig. 13 was chosen for U-Pu-Ti

*THE WASTE FLUX CAN BE REMOVED AFTER EITHER THE REDUCTION OF URANIUM OXIDES OR DECOMPOSITION OF THE INTERMETALLIC COMPOUND.

Fig. 12. EBR-II Liquid-metal Process for Reclamation of Melt-refining Skulls. ANL Neg. No. 103-B9075.

* The removal of FPN II is not a routine operation. Titanium, if a component of the core alloy, is eliminated with the FPN II

** The residual amount of fission-product elements in the products is based on 0.324 w/o fission products in discharged blanket fuel.

Fig. 13. Principal Steps in a Possible Pyrochemical Process for U-Pu-Ti Core and Uranium Blanket. ANL Neg. No. 103-89076.

because it consists of an assemblage of pyrochemical steps for which (1) sufficient support is available from laboratory and pilot-plant studies to provide some degree of confidence in the technical feasibility of the steps, and (2) sufficient information is available from component-development studies to develop conceptual equipment designs.

Although differences in fabricability exist between U-Fs and U-Pu-Zr (see Sec. VI), the same basic processes (injection casting, etc.) can be used. Therefore, the information already presented on refabrication of U-Fs fuels is also applicable to U-Pu-Zr fuels.

An alternative approach to reprocessing U-Pu-Zr fuels is the Purex solvent-extraction process. That process has a long and extensive history in reprocessing a broad range of reactor fuels^{49,50} and is currently being studied with the objective of adapting it to fast-reactor fuels. It is anticipated that some unique problems may be associated with dissolution of the alloy to prepare feed solution. For example, there is evidence that uranium-zirconium alloys may react explosively with nitric acid.⁴⁹ The same hazard may exist with U-Pu-Zr-Fs alloys. Oxidizing the alloy before dissolution may eliminate that hazard. However, dissolution of solid-solution (U-Pu-Zr) oxide may be difficult and may require the use of hydrofluoric acid. The problem of alloy dissolution would require considerable investigation. Criticality hazards with respect to the dissolver, feed-adjustment tanks, and contacting equipment would have to be resolved.

The Purex process produces uranium and plutonium nitrates essentially free of all fission products. Reconversion to the metals is a well-established procedure.⁴⁹ Such clean metals would be well suited to adjustment of the fission content of the blended metal alloy that is to be returned to fuel fabrication.

A further potential benefit may be derived from the combined system of melt-refining and Purex reprocessing. Since about half the fuel is completely purified, it may not be necessary to remove the alkaline- and rare-earth fission products from the portion that is recycled directly to refabrication. It would, then, be possible to eliminate the 1-3-hr liquation at 2550°F. It may also be possible to substitute another crucible material for the costly zirconia crucible, which cannot be reused. Thus, perhaps a graphite crucible could be used for effecting a rapid melt of the chopped fuel, followed by pouring the double ingot.

If these modifications of the melt-refining step are practical, it should be possible to significantly reduce the operating time for this step. Thus, two or three melting operations could be performed in the same furnace during a 24-hr day. A single 1000-MWe LMFBR would require a fuel-reprocessing rate of about 70 kg of fuel per day. If a critically safe furnace can handle 20 kg in a single melt, two furnaces should be sufficient for reprocessing the fuel from a 1000-MWe reactor. Increasing the size of the facility to handle fuel from three close-coupled plants should be feasible.

Since a Purex plant can be designed to handle several tonnes of reactor fuel per day, a single Purex plant could handle the dragout from several melt-refining facilities. These facilities would include equipment for production of uranium and plutonium metals from the Purex plant products and for remote refabrication of fuel elements and fuel subassemblies. Accordingly, the overall system would be uniquely adaptable to a large reactor park. Such a park would include a large number of reactors (perhaps a variety of reactors) and the associated facilities for reprocessing and refabricating fuel subassemblies.

Clearly, additional studies are required on processes for refining irradiated U-Pu-Zr fuels. However, the basic technology associated with all the remote steps of subassembly transfer, disassembly, pin decladding, melt refining, pin casting, fuel-element refabrication, and subassembly refabrication have been demonstrated in the EBR-II Fuel Cycle Facility for about 35,000 fuel elements. The following demonstrations of feasibility already achieved at EBR-II are particularly important.

1. Removal of gaseous and volatile fission products by simply melting the irradiated fuel. This is very practical for U-Pu-Zr alloys, which melt at about 2300° F (1260°C).
2. Quick turnaround of refined and refabricated fuel to the reactor, because only minimal cooling time (~14 days following reactor shutdown) is required before beginning reprocessing.
3. Ability to remotely refine, refabricate, and inspect metallic fuels.
4. Ability to remotely maintain reprocessing and refabricating equipment.
5. Ability to design and operate an efficient, quick-turnaround, on-site reprocessing plant that is scaled in size to match the LMFBR it supports.
6. Demonstration of high reprocessing- and refabricating-facility availability (~85%) over an extended period of time (six years).

The larger subassemblies designed for commercial breeder reactors will require redesign of equipment and operating facilities. The higher burnup anticipated will probably require a longer cooling period (say 30 days). Additional studies will be necessary to establish optimum conditions for fuel dissolution and other steps, but the fundamental background for each step justifies an optimistic projection for the overall process. One problem, which applies to all fast-reactor fuel processes, is the ultimate control and storage of fission-product wastes, including iodine and the fission-product gases.

C. Conclusions on Reprocessing

1. A simple extension of refining processes developed and successfully applied on EBR-II U-Fs fuel to the refining of U-Pu-Zr fuels does not appear

feasible. The reasons for this are: (a) the higher burnups and the accompanying high fissium-element content expected in U-Pu-Zr cannot be controlled with only melt refining, and (b) the higher burnups and the presence of plutonium in U-Pu-Zr cannot be accommodated by the EBR-II skull-reclamation process. However, parts of the EBR-II reprocessing scheme are applicable (e.g., melting to remove gaseous and volatile fission products).

2. A simple extension of remote refabrication processes developed and applied on EBR-II U-Fs fuel to the remote refabrication of U-Pu-Zr fuels does appear feasible.

3. The experience gained with remote reprocessing and refabrication of U-Fs fuel elements in the EBR-II Fuel Cycle Facility is invaluable in demonstrating for metallic fuels that (a) alloy fuel can be routinely refined, refabricated, and returned to the reactor in a relatively short time (~45 days) following reactor discharge, (b) complete remote refining and refabrication is a practical approach to fuel recycle, (c) remote maintenance of refining and refabrication equipment can be performed routinely, and (d) a fuel-reprocessing facility using remote techniques can operate at a high facility availability (~85%) over an extended period of time (~six years).

4. A combination of melt refining and solvent extraction of highly radioactive fuel in conjunction with remote refabrication appears to offer great advantages to a metallic fuel cycle, including (a) initiation of reprocessing as early as 14 days following reactor shutdown, (b) quick turnaround of external fuel inventory during recycle (i.e., reduced overall inventory), (c) reprocessing at the reactor site in a plant scaled to fit the adjoining LMFBR, and (d) reduction in the quantity of fuel that must be subjected to complete aqueous processing and subsequent reduction to metal form.

VIII. SAFETY CONSIDERATIONS

This section compares safety-related characteristics of large metal-fueled LMFBF's with those of corresponding oxide-fueled LMFBF's and then considers how these characteristics affect off-normal reactor operation. The most important safety-related characteristics are fuel-element design, time constants for heat transfer, modes of failure, and mechanisms for feedback reactivity. Off-normal operation includes the two general categories of transient overpower and loss-of-flow malfunctions.

A. Safety-related Characteristics

Table XXVII lists safety-related parameters for a 1000-MWe metal-fueled LMFBF and a corresponding oxide-fueled LMFBF. Design information on the metal-fueled reactor was taken from the unpublished parametric study done at Argonne National Laboratory in 1967 and discussed in Sec. III.A. This reactor had a core height of 38.2 in., a core equivalent diameter of 95.6 in. ($H/D = 0.40$), a core volume of 4500 liters, and an average core power density of 500 kW/liter. Information on the oxide-fueled reactor was taken largely from Ref. 51. The core height of this reactor was 36.0 in., its equivalent diameter was 81.8 in. ($H/D = 0.44$), its volume was 3100 liters, and its average power density was 657 kW/liter. In both reactors, the sodium-coolant inlet temperature was 855°F, and the mixed mean core ΔT was 295°F.

TABLE XXVII Safety-related Characteristics of Large Metal- and Oxide-fueled LMFBF's

	Metal-fueled	Oxide-fueled
Fuel material	U-15 wt % Pu-10 wt % Zr	UO ₂ -PuO ₂
Cladding material	316 SS	316 SS
Cladding OD x wall thickness, in	0.212 x 0.015	0.230 x 0.014 ^a
Fuel-pin diameter (as fabricated), in	0.152	0.195 ^a
Thermal-bond material (as fabricated)	Na	Re ^c
Fuel mp, solidus liquidus, °F	2110 2280	5010 5050 ^b
Fuel-cladding eutectic temp, °F	1515	None
Peak linear power, ^c kW/ft	12.1	14.4 ^d
Normal max cladding temp, ^d °F	1520	1300 ^a
Normal max fuel temp, ^d °F	1474	4480
Fuel-element time constant for heat transfer, sec	0.4	3
Doppler coefficient [- $\beta(dk/dT)$], Na in (i.e., no sodium loss from core)	-0.003 ^e	0.008 ^a
Axial expansion coefficient [- (dk/dT)], ^f 1/°F	2.4×10^{-6}	0 to 1.8×10^{-6}

^aFrom Ref. 51.

^bFor UO₂-15 at % PuO₂.

^c(Core average linear power) x (radial peaking factor) x (axial peaking factor).

^dWithout uncertainty factors, sodium inlet temperature = 855°F.

^eFrom Ref. 48.

^fValue depends upon amount of fuel-cladding mechanical interaction, see text.

As shown in Table XXVII, the ternary metal fuel alloy has a solidus temperature of about 2110°F and a liquidus temperature of about 2280°F. This alloy, however, forms a eutectic with SA 316 stainless steel at about 1515°F. For the U-5 Fs/SA 316 stainless steel fuel element used in EBR-II, the cladding penetration rate becomes significant with respect to reactor-accident

time scales (i.e., about 5% cladding-wall penetration/min) at about 1565°F. On the basis of discussions in Secs. V.A.1 and V.A.3, it is assumed here that this same temperature holds for the ternary alloy. That is, a cladding-inner-wall temperature of 1565°F represents a condition of incipient failure for a fuel element of the ternary alloy. This is about 245°F above the normal maximum cladding-inner-wall temperature given in Table XXVII. The corresponding increase in average fuel temperature is from about 1196 to 1411°F.

Another important temperature is that at which sodium boiling occurs; such boiling could lead to overheating of elements and their subsequent failure. Sodium boils at 1640°F at an absolute pressure of 1.1 atm. If sodium superheating is neglected and a ΔT of about 20°F across the sodium film and of about 30°F across the cladding wall at the core top is assumed, sodium boiling would occur when the maximum cladding-inner-wall temperature is about 1690°F. This is about 125°F above the cladding-inner-wall temperature at which cladding penetration by fuel becomes a safety concern. Thus, for the metal-fuel element, cladding penetration by fuel is taken as the controlling failure mechanism.

The solidus and liquidus temperatures of the mixed-oxide fuel depend upon its composition; for UO_2 -15 at. % PuO_2 , these temperatures are about 5010 and 5050°F, respectively. The mixed-oxide fuel does not form a eutectic with the cladding. Here, as with the metal-fuel element, overheating due to sodium boiling would be expected at a cladding-inner-wall temperature of about 1690°F, which is about 390°F above the normal maximum temperature. The corresponding increase in average oxide-fuel temperature is from about 2560 to about 3000°F.

Inherent negative feedback reactivity acts to increase the time to reach a severe failure condition. It also might act to mitigate the consequences of such failure in the extremely unlikely event of total malfunctioning of the plant protection system. Inherent negative feedback reactivity is provided by material thermal expansion and by the Doppler effect. Material expansion increases neutron leakage from the reactor and moves fuel to positions of lower reactivity worth. Such movement may be direct (i.e., by axial expansion of fuel) or indirect (i.e., by longer-time-scale heating of the grid plate, resulting in radial expansion of the core). A negative Doppler effect is due to reduction of self-shielding in ^{238}U capture resonances with increasing temperature, causing an increase in neutron captures in the ^{238}U . These capture resonances in ^{238}U are almost entirely below an energy level of about 25 keV. Thus the softer the spectrum in an LMFBR, the greater will be its Doppler effect.

In a small LMFBR such as EBR-II, the fuel enrichment is relatively high (i.e., the ^{238}U content is relatively small) and the spectrum is hard. The Doppler effect in such a reactor is small (and may even be positive) unless considerable effort is made to degrade the neutron spectrum. For example, a mixed-oxide-fuel core in EBR-II was calculated⁵² to have a Doppler coefficient $T(dk/dT) \approx -2 \times 10^{-4}$, and such a core with 17 vol % BeO added for spectral softening had a Doppler coefficient $\approx -2 \times 10^{-3}$.

Inherent negative feedback reactivity in EBR-II is provided by expansion of fuel, sodium, and structural material, all having different time constants. The total isothermal temperature coefficient for EBR-II was calculated to be $-2.0 \times 10^{-5} (\Delta k/k)/^{\circ}\text{F}$.⁵³ The prompt component (time constant < -0.4 sec) of the isothermal coefficient was calculated to be about $-7.8 \times 10^{-6} (\Delta k/k)/^{\circ}\text{F}$, of which about 29% is due to axial expansion of fuel, and 64% to reduction in density of core sodium. In a large LMFBR, the sodium-density effect would be expected to be small (and in the case of an oxide-fueled reactor to have a time constant of several seconds).

The Doppler coefficient in a large metal-fueled LMFBR is also sensitive to the amount, if any, of moderating material added. In the previously cited design study⁴⁸ of a 1000-MWe metal-fueled reactor, calculations were done for beryllium additions at the core/radial-blanket interface and radial-blanket/radial-reflector interface. Doppler coefficients $[-T(dk/dT)]$ were found to range from 0.002 for no beryllium to 0.004 with beryllium at both interfaces. An average value of 0.003 is used in Table XXVII. The Doppler coefficient in large oxide-fueled LMFBR's is calculated to be typically in the range of 0.007-0.009; an average value of 0.008 is used in Table XXVII. Note that all the foregoing are "sodium-in" values, i.e., for no sodium loss from the core. "Sodium-out" values would be lower.

The amount of axial expansion of oxide fuel in an LMFBR element under transient conditions depends in a complex manner upon the initial fuel density, fuel diameter, and gap between fuel and cladding. It also depends upon the linear power, burnup, cladding thickness, and temperature distribution of the sodium coolant. At low burnups, before the fuel contacts the cladding, the fuel may expand freely if it has not undergone extensive cracking. Although the oxide fuel has a lower linear-expansion coefficient than its cladding by about a factor of two, it would heat up faster than the cladding during a rapid reactivity or flow transient, because it has a lower thermal conductivity. Thus, the maximum axial expansion in an oxide element would occur if the oxide fuel expanded freely and coherently. Cracking, however, resulting from rapid shutdowns, may lead to the formation of spaces between pieces of fuel.

When the pieces of fuel heat up, they expand into the free spaces rather than expanding coherently. Such effects were observed in RAPSODIE.⁵⁴ Once the fuel contacts the cladding, it may be constrained to expand with the cladding. Since cladding temperature rises are not expected to be large in a credible reactivity or flow transient, the reactivity feedback due to cladding-controlled axial expansion of fuel is not expected to be large. For the foregoing reasons, there has been a widespread consensus that oxide fuel for an LMFBR does not have a well-characterized, reliable, axial-expansion coefficient of reactivity.⁵⁵ In Ref. 51, zero fuel-axial-expansion feedback reactivity was treated as "the worst assumptions possible; the only negative feedback is the Doppler effect." That reference also asserts that "This combination (Doppler plus axial fuel growth restrained by the cladding) probably represents a minimum amount of realistic negative feedback that can be expected during the transient." Following Ref. 55, however, zero axial-expansion feedback is assumed here for an oxide element.

With regard to metal fuel, consider first the axial-expansion characteristic at burnups below that at which contact with the cladding occurs. Take

$$\frac{dk}{dT} = \frac{dk}{dH/H} \cdot \frac{dH/H}{dT} = \alpha \frac{\Delta k}{\Delta H/H},$$

where H is the core height and α is the average linear-expansion coefficient of the metal fuel in the temperature range of interest (here taken as 572-1292°F). From thermal-expansion data given in Ref. 11, α for U-15 wt % Pu-10 wt % Zr is estimated to be $15.4 \times 10^{-6}/^{\circ}\text{F}$. This value includes expansion due to solid-state phase transformations in the interval 1103-1229°F. As a point of interest, α for the U-5 Fs EBR-II fuel alloy is calculated from dilatometric data to be $15.2 \times 10^{-6}/^{\circ}\text{F}$ for the interval 572-1292°F.⁵⁶ For this latter alloy, solid-state phase transformations occur in the interval 1026-1337°F.⁵⁶

The change in reactivity per unit change in core height [$\Delta k/(\Delta H/H)$]- - and the associated reduction in fuel density- - is taken to be the same as that used in Ref. 51 for the oxide core, namely, -0.158. This value is close to values reported elsewhere for similar reactor configurations.⁵⁵ Then, for unrestrained axial expansion of the ternary metal fuel alloy,

$$-\frac{dk}{dT} = 15.4 \times 10^{-6} \times 0.158 = 2.4 \times 10^{-6}/^{\circ}\text{F}.$$

A comparison can now be made of the inherent prompt-negative-feedback reactivity available in the interval between normal operation and incipient failure for the oxide- and metal-fueled reactors. For the oxide-fueled reactor, the feedback is taken to be due only to the Doppler effect; i.e.,

$$-\Delta k_{\text{Dop}} = 0.008 \ln \frac{3000 + 460}{2560 + 460} = 1.09 \times 10^{-3}.$$

For the metal-fueled reactor, the feedback is taken to be due to Doppler effect plus unrestrained axial expansion:

$$-\Delta k_{\text{Dop}} = 0.003 \ln \frac{1196 + 245 + 460}{1196 + 460} = 4.1 \times 10^{-4},$$

$$-\Delta k_{\text{exp}} = 2.4 \times 10^{-6} \times 245 = 6.0 \times 10^{-4},$$

and

$$-(\Delta k_{\text{Dop}} + \Delta k_{\text{exp}}) = 1.01 \times 10^{-3}.$$

Thus, if unrestrained axial expansion of all the metal fuel is assumed, the metal-fueled reactor has about the same inherent prompt-negative-feedback reactivity as the oxide-fueled reactor in the temperature range between normal operation and incipient failure. Of this, about 40% is Doppler feedback and 60%

is axial-expansion feedback. In reality, however, the metal fuel will be at various burnup levels throughout the reactor, depending upon the power distribution, peak allowable burnup, ratio of the enrichments of the inner- and outer-core zones, and refueling scheme. As a worst hypothetical case, all the metal fuel would be in complete contact with its stainless steel cladding, with axial expansion controlled by the latter. Even if the transverse fuel-pin cracking described in Sec. IV.B occurs, the axial fuel expansion should follow the cladding expansion. Taking the linear-expansion coefficient of the stainless steel to be $11.0 \times 10^{-6} \text{ } ^\circ\text{F}^{-1}$ in the temperature range of interest, we find that

$$-\Delta k_{\text{exp}} \approx 6.0 \times 10^{-4} \times \frac{11.0}{15.4} = 4.3 \times 10^{-4}$$

and

$$-(\Delta k_{\text{Dop}} + \Delta k_{\text{exp}}) \approx 0.84 \times 10^{-3}.$$

Hence, for this worst hypothetical case, the metal-fueled reactor has 77% of the inherent prompt negative feedback of the oxide-fueled reactor. These results suggest that the metal- and oxide-fueled reactors would place similar demands upon required response for a plant protection system and would provide similar feedback reactivity in a hypothetical unbounded reactivity transient.

B. Off-normal Reactor Operation

The foregoing discussion focused upon inherent prompt-negative-feedback characteristics without regard to specific malfunctions and their associated time scales. That is, effects of heat loss from the fuel were not considered. Such effects are important, however, in discussing feedback reactivity for reactor malfunctions under the two general categories of transient overpower and loss of flow. It is in this context that the order-of-magnitude difference in time constants for metal and oxide fuel elements (see Table XXVII) becomes important.

In the temperature range of interest for most postulated credible accidents, the thermal conductivity of mixed-oxide fuel is small at normal operating temperatures and decreases with increasing temperature. This inhibits heat loss to the cladding and thereby enhances the Doppler effect in the fuel and tends to reduce overheating of the cladding. However, the volumetric heat content $\rho \bar{C}_p \Delta T$ (where ρ = fuel density, C_p = specific heat, and ΔT = average temperature rise from normal operation to incipient failure) of the oxide fuel is almost twice as great as that of the metal fuel. This mitigates somewhat the Doppler effect in oxide fuel. (In other words, a given Doppler feedback in oxide fuel is obtained at the expense of about twice as much thermal energy stored in the fuel.) This thermal-conductivity effect is important for transients with time scales shorter than that of the time constant for the oxide fuel (0.3 sec), e.g., for unbounded reactivity-insertion rates greater than about 0.3 $\$/\text{sec}$. It is not important for flow-coastdown transients lasting about 1 min or longer.

At low rates of inadvertent reactivity insertion (leading to relatively slow increases in power), long-time-constant inherent negative feedback reactivity due to such effects as grid-plate expansion and subassembly bowing would be of benefit in any LMFBR, regardless of fuel type. On the other hand, short-time-constant inherent negative feedback due to the Doppler effect and fuel axial expansion would be reduced in both metal- and oxide-fueled reactors because of the relatively long time available for loss of heat from the fuel. This argument also applies to loss-of-flow malfunctions that occur in time scales of the order of 1 min.

As the reactivity-insertion rate increases, the long-time-constant feedback terms diminish in effectiveness. Under such conditions, however, the prompt feedback reactivity in the metal-fueled reactor is slower to approach its limiting adiabatic value than is the corresponding reactivity in an oxide-fueled reactor. This is simply because the time constant for heat transfer in the metal fuel is about a factor of 10 smaller. The increase in power, which is determined by the net reactivity added, would thus be expected to be more rapid in a metal-fueled reactor than in an oxide-fueled reactor for a given inadvertent unbounded reactivity-insertion rate.

Slumping of metal fuel (shortening by creep under its own weight) under normal operating conditions during long irradiation times was discussed in Sec. V.C. Slumping or compaction of such fuel under hypothetical accident conditions must also be considered. During irradiation, the metal fuel swells both radially and axially within its cladding (see Sec. IV). Assume that a metal-fueled reactor containing a large amount of swollen fuel is operating normally and then undergoes a transient leading to overheating of the fuel. The question then arises as to whether rapid compaction of the swollen fuel will take place, leading to potentially large rates of reactivity insertion.

In the EBR-II Mark-II metal-fuel design, 33% fuel swelling is allowed; at 25-30% swelling, interconnection of porosity occurs. There are two important safety aspects of the interconnected porosity. First, it allows about a 60% fission-gas release from the fuel. Second, it allows thermal-bond sodium to move into the porous fuel and log it. Thus, if high-burnup metal fuel overheats, it should not literally explode, as it might if it contained large amounts of fission gas. Nor would it be expected simply to slump rapidly into a more reactive geometry as it approached its melting point. This is because the U-15 wt % Pu-10 wt % Zr alloy begins to melt at about 2110°F, but at this temperature, the logged bond sodium would have a vapor pressure of 7.3 atm absolute. The logged sodium would thus tend to have a dispersive effect on the fuel column, at least in the early stages of a transient. Subsequent fuel motion, involving the potential for recriticality, is a complex problem that probably would require transient reactor tests and supporting analysis for solution.

A metal fuel element contains sodium as a thermal bond. Consequences of loss of the bond have been considered extensively for the metal driver fuel in EBR-II. The following summarizes the results of some tests of elements containing a partial bond or no bond.⁵⁷

An element with a partial circumferential bond has been tested at TREAT (EBR-MF-1) in a test simulating steady-state power in EBR-II. The element was fully bonded in the lower 4.5 in. of its length while the upper 9 in. had a partial circumferential bond of $180^\circ \pm 50^\circ$. The element was subjected to a flat-topped transient in TREAT with peak powers of 420 W/g and an average power of 290 W/g (equivalent to a reactor power of 85 MWt in row 1). The postirradiation results indicated no fuel-cladding eutectic formation, no cladding strain, and little change in the pin diameter. The data indicated that the fuel was heated above 642°C , the transformation temperature for α to γ phase, but was not heated to its melting temperature (1002°C for solidus).

Element EBR-MF-2, which contained a partial axial bond, was also tested in TREAT. The element was made with a complete bond in the lower 3.6 in., a partial bond for the next 4.1 in., and no bond for the upper 5.8 in. The element was subjected to a flat-topped transient in TREAT for an average power of 308 W/g (equivalent to a reactor power of 88 MWt in row 1). Posttest examination indicated that the upper section of the fuel had contracted 0.6 in. Straining of the cladding occurred in the unbonded areas, with a maximum diameter increase of 0.0005 in. ($0.3\% \Delta D/D$). Examination at the $9\frac{1}{2}$ -in. level and at the top of the pin indicated that liquid fuel had been in contact with the cladding. For most of the fuel, however, melting occurred only at the grain boundaries, the location of the lowest melting uranium alloy composition. The liquid fuel in contact with the cladding had reacted with part of the cladding to form a liquid eutectic alloy. The maximum reduction in the nominal 0.009-in. wall thickness was 0.002 in. At the center of the fuel, the grain structure indicated that the fuel had become molten only at the grain boundaries and that all of the fuel had not melted.

Tests ID-RP-1 and ID-RP-2 were conducted on elements with no sodium bond. The elements were subjected to flat-topped transients in TREAT to power densities of 170 and 270 W/g, respectively. For the former test, local fuel-cladding eutectic formations occurred, with the maximum penetrations being less than 0.001 in. For the latter tests, the maximum eutectic penetration was about 0.001 in.

In all of the above tests, the flat-topped transient allowed the elements to be at a relatively level power for about 8 sec. The power ratings were high enough so that fuel for the unbonded elements would have reached its melting temperature, slumped, and recast against the side of the element and run at equilibrium conditions at the end of the test. There was no cladding failure in any of the tests, even though there were areas of local fuel-cladding eutectic formation.

In more recent tests,⁵⁸ a special subassembly, X149, containing 12 partially bonded EBR-II Mark-IA driver-fuel elements was irradiated in EBR-II during run 59A to an exposure of 74 MWd. Eight of the elements contained

partial axial voids in their bond sodium, and four contained partial circumferential voids in theirs. Neutron radiographs of the 12 elements made subsequent to the irradiation indicated that all the elements containing axial voids had significant amounts of fuel melting in the upper 5 in. of the fuel pins. All the elements containing circumferential voids showed little change, except for small sections along the centerline of the fuel pins, where melting apparently started. No penetration of the cladding was observed in any of the 12 elements, and they were judged suitable for reinsertion into EBR-II for further irradiation. Reconstituted as experimental subassembly X149A, the subassembly was irradiated in EBR-II in runs 69A-74G, totaling 11,306 MWd (~2.1 at. % burnup). During this interval, it gave no evidence of fission-gas release or bond-sodium release. All the foregoing work supports the belief that substantial debonding of a metal-fuel element will not lead to breaching of its cladding.

In the highly unlikely event of a reactor malfunction leading to breaching of cladding by molten metal fuel, however, the potential for molten-fuel/coolant thermal interaction (MFCI) must be considered. Fauske⁵⁹ has proposed that a necessary, but not sufficient, condition for a vapor explosion to result from an MFCI is that the interface temperature between the "hot" and "cold" liquids must be greater than the homogeneous nucleation temperature of the "cold" liquid. For sodium as the cold liquid, Fauske reports that the nucleation temperature has been found experimentally to be ~2300°K. The interface temperature is defined by

$$\frac{T_H - T_I}{T_I - T_C} = \sqrt{\frac{(\rho k C_P)_C}{(\rho k C_P)_H}},$$

where

T = absolute temperature,

ρ = density,

k = thermal conductivity,

C_P = specific heat,

$H \equiv$ hot liquid,

$C \equiv$ cold liquid,

and

$I \equiv$ interface.

When we use the estimated properties for molten U-5 Fs alloy (that used for EBR-II driver fuel) and $T_I = 2300^\circ\text{K}$, the foregoing equation gives T_H as a function of T_C . For $T_C = 1000^\circ\text{K}$ (1340°F), $T_H \approx 2560^\circ\text{K}$ (4148°F). This temperature is ~2000°F above that at which the metal fuel is completely molten. Thus, before temperatures high enough to cause an MFCI with metal fuel would be reached, other potentially more serious problems would arise, e.g., rapid rearrangement of fuel.

The foregoing comparison of the behavior of large metal-fueled and oxide-fueled LMFBR's was admittedly qualitative. A quantitative comparison could only be made after doing calculations of loss-of-flow and overpower transients for each type of reactor. These calculations should be done on as consistent a basis as possible, using appropriately justified feedback-reactivity coefficients, criteria for incipient failure of fuel elements, etc. Results of the calculations would show any significant differences in plant-protection-system and containment requirements for the two reactor types.

C. Conclusions on Safety Considerations

1. Under off-normal reactor operating conditions, the controlling mechanism for element failure appears to be cladding penetration by fuel for metal-fuel elements and sodium boiling for oxide-fuel elements.
2. For the interval between normal operating conditions and incipient failure of elements, the prompt negative feedback reactivity due to both axial expansion of fuel and the Doppler effect in a large metal-fueled LMFBR is about the same magnitude as that due to the Doppler effect alone in a large oxide-fueled LMFBR.
3. Large metal- and oxide-fueled LMFBR's will have similar plant-protection-system requirements with regard to response characteristics.
4. To obtain a given amount of Doppler feedback reactivity, the thermal energy stored in the fuel of an oxide-fueled reactor must be twice as much as that stored in the fuel of a corresponding metal-fueled reactor.
5. For a given inadvertent unbounded reactivity-insertion rate, the increase in power would be expected to be more rapid in a metal-fueled reactor than in an oxide-fueled reactor.
6. Overheating of high-burnup metal fuel would not be expected to lead to rapid fuel compaction and potentially large rates of reactivity insertion; however, both out-of-pile and in-pile tests would probably be required to prove this.
7. Results of a number of in-pile (TREAT, EBR-II) tests indicate that substantial debonding of a metal fuel element will not lead to breaching of its cladding.
8. With metal fuel, molten-fuel/coolant interaction would only become explosive at temperatures at which a more serious problem would be likely to occur, namely, extensive compaction of the core.
9. Analyses should be done to provide a quantitative basis for comparing the safety-related characteristics of large metal-fueled LMFBR's with those of large oxide-fueled LMFBR's; such work should use consistent reactor configurations, operating conditions, material properties, hypothetical malfunctions, etc.

APPENDIX

Properties of U-Pu-Zr Fuel Alloys

This appendix summarizes in tabular form the available pertinent information on physical and tensile properties of U-Pu-Zr alloys. For comparison, information is also presented on other metallic fuels.

TABLE XXVIII. Physical Properties of U-Pu-Zr Alloys^{a,b,c}

Property	Alloy, wt %				
	U	U-5 Pu	U-11 Pu-6 Zr	U-15 Pu-10 Zr	U-18 Pu-14 Zr
Liquidus temp., °F	2070	2160	~2190	~2280	~2350
Solidus temp., °F	2070	1872	~2050	~2110	~2140
Transformation temp., °F					
Start	1234	1026	1103	1103	1103
Complete	1427	1337	1256	1229	1220
Density at 77 °F (as cast), g/cm ³	19.1	18.0	16.8	15.8	14.8
Average coefficient of thermal expansion, 10 ⁻⁶ /°F					
77 °F to first transformation	10.8	9.2	10.2	9.8	9.7
End of transformation to 1742 °F	12.8 ^a	b	10.1	11.2	11.1
Fractional length increase in transformation range, $\Delta l/l$	0.0045	0.0059	0.0051	0.0052	0.0050
Thermal Conductivity cal (sec)(cm)(°C)					
at 230 °F	0.069	0.048	-	0.031 ^c	-
at 1640 °F	0.127	0.120	-	0.076 ^c	-
DPH Hardness at 77 °F (as cast)	290	214	470	540	410

^aData to 1472 °F only.

^bNo data available for temperatures above transformation temperature of 1337 °F.

^cData for U-15 wt % Pu-6.8 wt % Zr.

TABLE XXIX. Tensile Properties of Extruded U-15 wt % Pu-10 wt % Zr (Ref. 35)

Temperature, °F	Ultimate Tensile Strength, 10 ³ psi	Yield Strength (at 0.2% off-set), 10 ³ psi		Elongation, %	Elasticity, 10 ⁶ psi
77	70	-	-	Nil	2.2
570	39	-	-	Nil	2.1
660	44	44	3	0.9	
750	14	10	53	0.4	

ACKNOWLEDGMENTS

The assistance of the following people is gratefully acknowledged: G. J. Bernstein, R. M. Fryer, A. G. Hins, J. H. Kittel, and M. J. Steindler for their technical review and suggestions on various sections; J. E. Flinn, G. L. Hofman, and J. F. Koenig for allowing our use of unpublished data; and K. J. Mudge for aid in preparing the report.

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