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ABSTRACT

Tests to date demonstrate that metallic fuel for advanced liquid metal reactors performs well, is easily reprocessed and refabricated and provides inherent reactor safety within an economic design. The behavior and performance of metallic fuel is key to the demonstration of the Integral Fast Reactor (IFR) concept at Argonne National Laboratory. Since 1985, more than 40 assemblies of experimental fuel in addition to the standard metallic driver fuel for Experimental Breeder Reactor II (EBR-II) have been irradiated; several more continue to be designed and fabricated. Results have characterized the influence of a wide range of fabrication, design and material variables upon irradiation behavior throughout the fuel lifetime under normal and upset conditions including operation with breached cladding. Results of tests, both in- and out-of-reactor, indicate that metallic fuel is readily and economically fabricated, capable of achieving high exposure and long reactor residence times, and possesses unique and promising safety features.

INTRODUCTION

Metallic fuels for liquid-metal-cooled fast reactors have received renewed interest as a result of Argonne National Laboratory's IFR concept.[1] This concept involves a novel approach to achieve improved safety, straightforward fuel-cycle closure, and waste advantages based on the unique features of metallic fuel.

Developments associated with the performance of metallic fuels have been fast-paced; the past 30 years of development have been adequately reviewed in several recent papers.[2-4] In February of 1985, renewed testing began when three complete assemblies of advanced metallic fuel were placed in the core of the EBR-II . The 61-pin assemblies each contained an identical complement of metallic fuel consisting of three compositions in weight percent; U-10Zr, U-8Pu-10Zr, and U-19Pu-10Zr. The pins were clad with the austenitic alloy D9, had a peak linear power rating of 48 kW/m, and achieved peak cladding temperatures of 583°C. The highest burnup achieved on these pins was 18.4 at.% with one end-of-life fuel column failure at 16.4 at.% burnup. These lead assemblies, X419-X421, have demonstrated that metallic fuels have the potential of being competitive with any existing fuel type in terms of steady-state[5] and transient[6] performance.

Subsequent to the initiation of the irradiation of the three lead assemblies, a broad-based development program was instituted to fully explore the potential of metallic fuel. To date more than 40 assemblies of experimental fuel have been irradiated with another five being fabricated. Among these were tests characterizing the influence of fabrication variables of minor impurity concentrations and defects in the fuel; cladding defects; design variables of major composition, fuel-smeared density, fuel column length and fuel-to-plenum volume ratio; and run beyond cladding breach (RBCB) of both intentionally-defected pre-irradiated elements to accelerate breach and naturally-occuring breaches. Ex-core tests and analyses were initiated to study phase relationships, fuel/cladding compatibility, zone formation, fuel plastic flow properties, thermal conductivity, and thermal expansion. Results from this work as well as the results from the postirradiation examination of the in-reactor experiments are used in a continuously evolving fuel performance code called LIFE-METAL.

IRRADIATION PERFORMANCE RESULTS

A great deal of progress has been made toward the complete understanding of metallic fuel systems. Although most of the phenomena now under investigation were initially discovered in the late 1960s, the recent irradiations have confirmed the consistency of fuel swelling, gas release, compatibility with the cladding and element redistribution within the fuel.

Performance issues important to design of advanced reactor systems include fission gas release, axial growth of the fuel and the breach characteristics demonstrated by metallic fuels. The fission gas release behavior is important as it has been found that primary loading of the cladding is produced by fission gas pressure; other loading mechanisms, such as fuel-cladding mechanical interaction, do not appear to be significant until very high burnup is achieved. The molar quantity of gas released is nearly linear with burnup while the fractional release asymptotically approaches 80% theoretical at about 18 at.% burnup, Figure 1.

In general, most of the important phenomena studied show a dependence on plutonium concentration in the U-Pu-Zr alloy. For example, as the fuel is irradiated it swells both axially and radially until contact is made with the cladding. Fuel/cladding contact is complete by 2 at.% burnup and may open again at high exposures with cladding which swells significantly. Radial fuel growth, which is freely promoted in the 73% smear density design, dominates the swelling process until cladding contact at ~2 at.% burnup, while axial growth (which depends on Pu content) behaves in a near linear fashion between 2 at.% and 18 at.% burnup, Figure 2. The extent of axial swelling is a strong function of plutonium concentration with the extent of axial elongation minimal with a plutonium content of 19 wt % at nominal operating conditions.[7] The variance in swelling behavior between U-10Zr and U-19Pu-10Zr vanishes at lower fuel operating temperatures. Microstructural examinations have shown that the swelling mechanism changes as a function of composition and temperature (tearing vs. bubble formation), and the microstructure correlates well with observed degrees of swelling anisotropy.

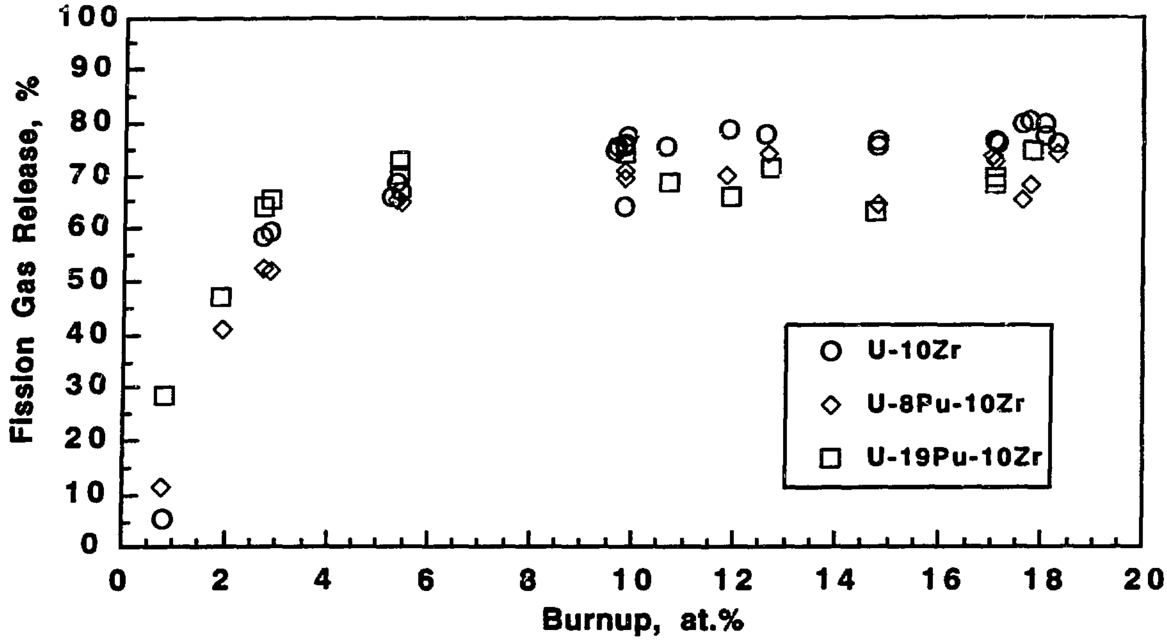

Figure 1. Gas Release from Metallic Fuel

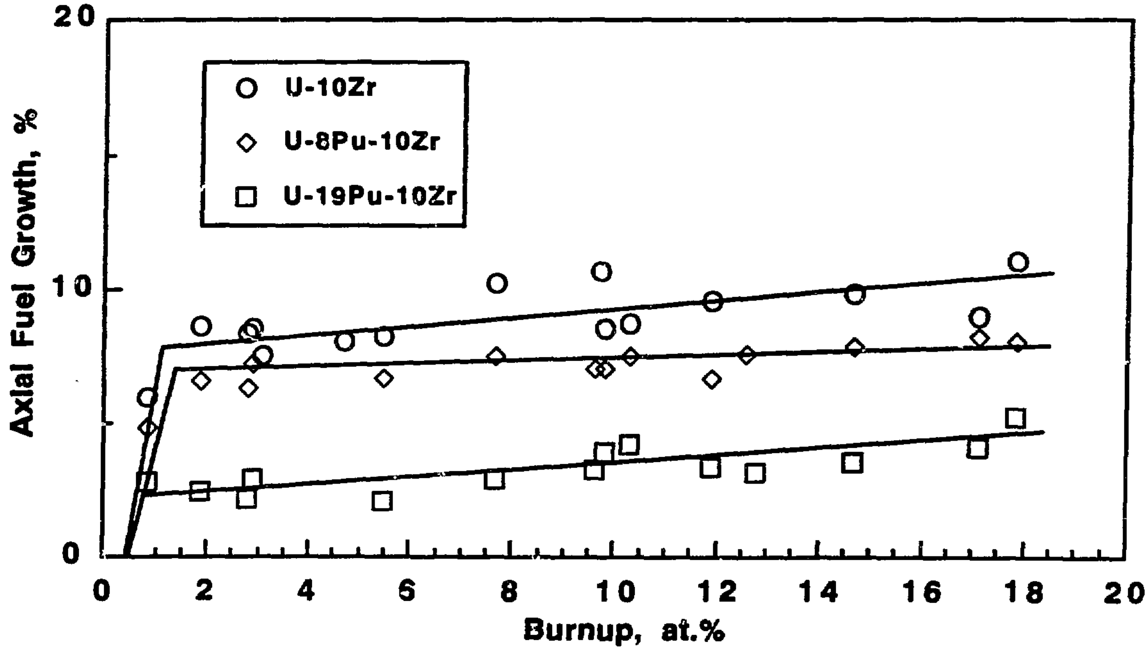

Figure 2. Axial Elongation of Metallic Fuel

One of the more interesting phenomena is the radial redistribution of the fuel alloying elements during irradiation.[8] By 2 at.% burnup, an interchange between zirconium and uranium occurs. Depending on fuel temperatures and plutonium content, this leaves either a low zirconium shell (<2 wt.%) surrounding a zirconium-rich core or a zirconium depleted zone at slug center. Pie-shaped cracks in the higher plutonium-content alloys are common at the earliest stages of redistribution but are completely "healed" by fuel growth before 10 at.% burnup. Little redistribution occurs in the other alloys. Postirradiation examination is now underway to measure composition profiles at 17-18 at.% burnup. We have found recently that the extent of radial redistribution depends on the alloy composition; as the plutonium concentration of the alloy is increased, the radial redistribution becomes more pronounced. An adequate model for this phenomenon is yet to emerge; the basis must include the results of ex-core diffusion couples and ternary diffusion analysis as well as in-core irradiation observations. To date we have found that the radial redistribution of fuel species does not limit the performance of metallic fuel.

BREACH CHARACTERISTICS

The X419 experiment was examined twice before it was terminated at 11.9 at.% burnup with no breaches observed. The X420 experiment was reconstituted once prior to first breach at 13.5 at.% burnup. The experiment was terminated at 17.1 at.% burnup with a total of eleven breaches. The X421 experiment was reconstituted at 9.8 at.% burnup and then continued on as X421A to 18.4 at.% peak burnup. Seven breeches were identified upon examination. Five breaches were associated with welds in Type 316-clad elements and two were in the TIG-closure weld on D9-clad elements. All but four of these 18 breaches were associated with weld failures.

In the Type 316 stainless steel clad elements with a capacitance-discharge closure weld, the failure site was located away from the primary weld but associated with a secondary tack weld between the lower end of the plug and the inside of the cladding tube. In the TIG-welded elements with D9 cladding, the failure site was located at a sharp notch between the end plug base metal and the fusion zone. A small, tight crack grew perpendicular to the element axis and allowed fission and tag gas to escape. The weld failures were all of a benign nature, with no fuel loss nor delayed neutron signals present. Redesign of this weld joint and fitup has eliminated the stress-riser effect. These early weld-related failures were clearly unrelated to fuel type.

Three other breaches in the X420 experiment also occurred above the core region and can be considered unrelated to the fuel. Though examination is now just beginning on these elements, it would appear from bond sodium and fission product deposits that the failure site is in the plenum region. These failures occurred ~100-150 mm above the top of the fuel. Because this is a region of low cladding temperature and strain with no adjacent fuel we can only now speculate that defects during remote reconstitution may be involved.

The only breach observed that may truly be related to the fuel occurred in element T084 from the X420 test at 16.4 at.% burnup. Exact knowledge of the breach burnup can be made since tag gas signals and delayed neutron signals were coincident. The failure site was in the fuel region of this U-19Pu-10Zr element, ~225 mm from the bottom of the fuel column ($X/L_0 = 0.67$). Expulsion of bond sodium and fission products led to a burst-like DN signal of 20 minute duration approximately four times background. The crack appeared to be very small and tight and was at the azimuthal position of maximum element-element interaction due to the excessive swelling and creep of the cladding at that burnup.

Nondestructive examination (neutron radiography and gamma spectrometry) gave no evidence of abnormal fuel restructuring at the breach site but cladding profilometry by laser scanning showed pronounced ovality of the cladding at this elevation. Weight loss analysis showed significant fission gas, bond sodium and liquid fission product venting had occurred. EBR-II operated with this natural fuel column breach for an additional 34 days (0.6 at.% burnup) to the scheduled end of the cycle without consequence.

RUN BEYOND CLADDING BREACH PERFORMANCE

The RBCB performance testing was accelerated by pre-thinning the cladding of a preirradiated test element and allowing it to breach under the stress induced by internal fission gas pressure during further irradiation. Several tests were conducted using different ternary fuel alloys of U-xPu-10Zr (x = 0-19 wt %) as well as D9 and Type-316 stainless steel cladding materials. Testing of elements with HT9 cladding is currently in progress. The test pins were previously irradiated to a range of burnups between 3 and 12 at.%. prior to thinning the cladding. Steady-state irradiation and subsequent examination of the RBCB tests have resulted in a large data base that has produced strong support of the fuel's predicted benign behavior under breach conditions. Table I provides a description of the RBCB program and its current status to date. In all of the IFR metal fuel scoping tests, the breach results appeared to have a common characteristic release signature. Typically, upon onset of the breach, an increase in reactor cover gas activity as well as a relatively short-lived delayed neutron (DN) signal was observed. In the case of tagged elements, the tag was readily identifiable. In the case of XY-24 and XY-27, a series of small peaks of activity in the reactor cover gas from fission gas released through the breach was observed. None of the breaches provided any DN activity of a prolonged nature nor a very high DN signal. Calculated time-to-rupture values using internal gas pressure alone appears to be conservative, potentially due to the bonding of the fuel to the cladding. This metallurgical interaction layer may distribute the stress at the deformation site, thereby delaying the instability that normally occurs in the final stages of creep rupture. Examination of breached elements revealed that no crack widening or fuel loss had occurred during prolonged RBCB operation. Only during the initial breach was there any evidence of significant release of activity from the fuel element. After the release of bond sodium, cesium and other liquid fission products, along with the accumulated fission gas, the reactor cover gas activity ceased, with the exception of periodic small releases of gas. Based on the results of the scoping tests, a more aggressive series of RBCB experiments were conducted on elements of IFR prototypic design. All three of the prototypic IFR fuel experiments were to be irradiated for approximately two reactor cycles each and consisted of U-10Zr and U-19Pu-10Zr fuel with D9 and HT9 cladding. The objectives to be achieved by the prototypic tests were to:

- observe general RBCB characteristics of IFR metal fuel in an open core position;
- observe dynamics of a clad breach at above midplane elevation of an IFR metal fuel slug;
- observe gas and DN signals to establish a characteristic "signature" for IFR prototypic elements at high burnup under RBCB conditions;
- characterize fission product release and possible fuel loss from elements during extended RBCB operation;
- characterize changes in thermal conditions of fuel upon breach; and
- observe changes in fuel cladding/chemical interaction after breach.

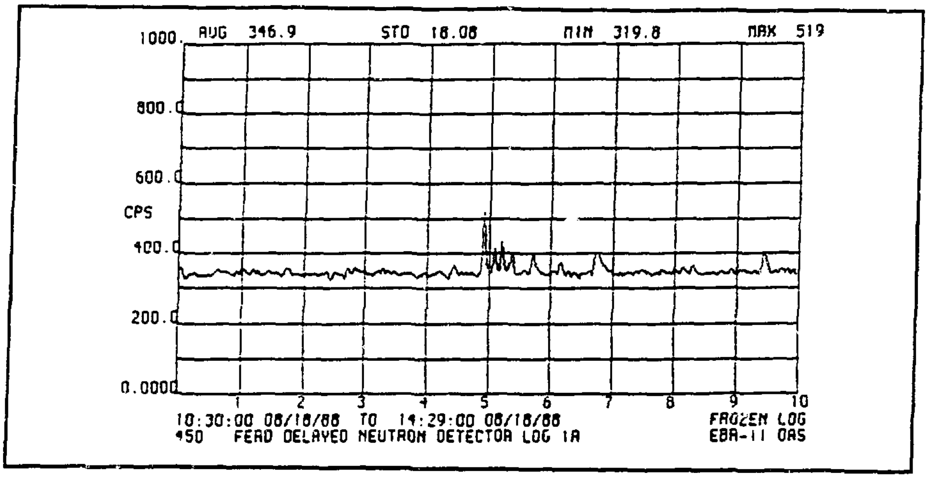

Figure 3. Delayed Neutron Signal from X482

Furthermore, *thermally-activated* cladding penetration rates are low near the temperature for first appearance of a liquid phase. We have also found that interstitial impurities in the fuel and cladding play an important role in the stabilization of a zirconium layer on the periphery of the fuel which tends to retard the interdiffusion of the fuel and cladding at the interface. At high burnup, the lanthanide fission products are found to segregate near the cladding inner surface and play a major role in compatibility at elevated temperatures. Interdiffusion of cladding components, primarily nickel and iron into the fuel and lanthanide fission products out of the fuel into the cladding during steady-state conditions promotes formation of a liquid phase at elevated temperature. After one hour at 800°C, the cladding wall was reduced as much as 26% in U-19Pu-10 Zr fuel.

In-reactor demonstration of reliability of metal fuel elements at temperatures above the eutectic formation temperature began with the EBR-II Mark II element design *consisting of* U-Fs fuel and Type 316 stainless steel cladding. Mark-II metallic fuel elements, which only exhibit liquid phase attack at temperatures above 715°C, were operated at temperatures up to 800°C in EBR-II in a 61-element subassembly, XY-22, in order to characterize high temperature breach.[9] Fuel elements with a large range of burnups were included in the test in order to compare the damage caused by the over-temperature operation at various stages in fuel element life. The subassembly operated at elevated temperature for ~42 minutes when failure of a high burnup element (7.69 at.%) occurred and the test was terminated. The failure was identified as being due to stress rupture at the fuel restrainer dimple and both mode and time-to-failure agreed very well with pre-test predictions. The unbreached elements, having sustained some limited damage to the cladding due to liquid phase formation, were removed and irradiated in another subassembly (X427) at normal operating temperatures until first

Table I. Status of RBCB Testing

	Scoping Tests			IFR Prototypic		
Experiment	XY-21/21A	XY-24	XY-27	X482	X482A	X482B
Composition, wt%	U-5Fs	U-19Pu-10Zr	U-8Pu-10Zr	U-19Pu-10Zr	U-10Zr	U-19Pu-10Zr
Cladding Material	316SS	316SS	316SS	D9	D9	HT9
Initial Burnup, at.%	~9.3	~7.5	~6.0	~11.9	~11.9	~10.6
Element Diameter, mm	4.4	4.4	4.4	5.8	5.8	5.8
Breached Condition, Days	54	233	131	168	~100	—
MWD	3348	18453	15234	9200	6200	—
RUNS	136-139	143-146	144-146	149-150	152-153	154-155
STATUS	NN41(XY21) no breach; RT95(XY21A) breached	J507 breached; J516 no breach	J432 breached; J486 breached	T139 breached	T045 breached	planned breach for element T464 from S/A X425A
DN Signal, cps*	~30-40	**	**	~600	~700	—
Weight Loss, g***	~2.0 g	2.7 g	~2.5	4.04 g	not available	—
Irradiation Facility	BFTF	FPTF	BFTF	Open Core	Open Core	Open Core

*counts above background
**unavailable due to malfunction of instrument sensitivity
***expulsion of bond sodium accounts for majority of weight loss; there was negligible fuel loss

The prototypic tests performed thus far have demonstrated the same characteristic pattern as the scoping tests (see Table I) which have also been confirmed by the characteristics of the naturally-occuring breach in X420. In experiments X482 and X482A, short duration DN signals of 600 to 700 cps above background were observed, Fig. 3. No further opening of the defect after initial breach occurred, Fig. 4, and fuel loss was negligible. Although many examinations remain to be performed on the prototypic test elements, the data obtained thus far, taken with the scoping test results, indicate an overall very benign RBCB behavior of IFR metallic fuel.

LIQUID-PHASE ATTACK AT ELEVATED TEMPERATURE

Another area of intense investigation is the determination of temperature and times for the first appearance of liquid phases in the fuel and cladding interaction zone when exposed to temperatures above nominal steady-state conditions. We have found through laboratory testing of irradiated fuel element sections and whole elements that independent of the fuel alloy composition and cladding material, ferritic or austenitic steel, the temperature for the first appearance of a liquid phase in irradiated samples is above 700°C.

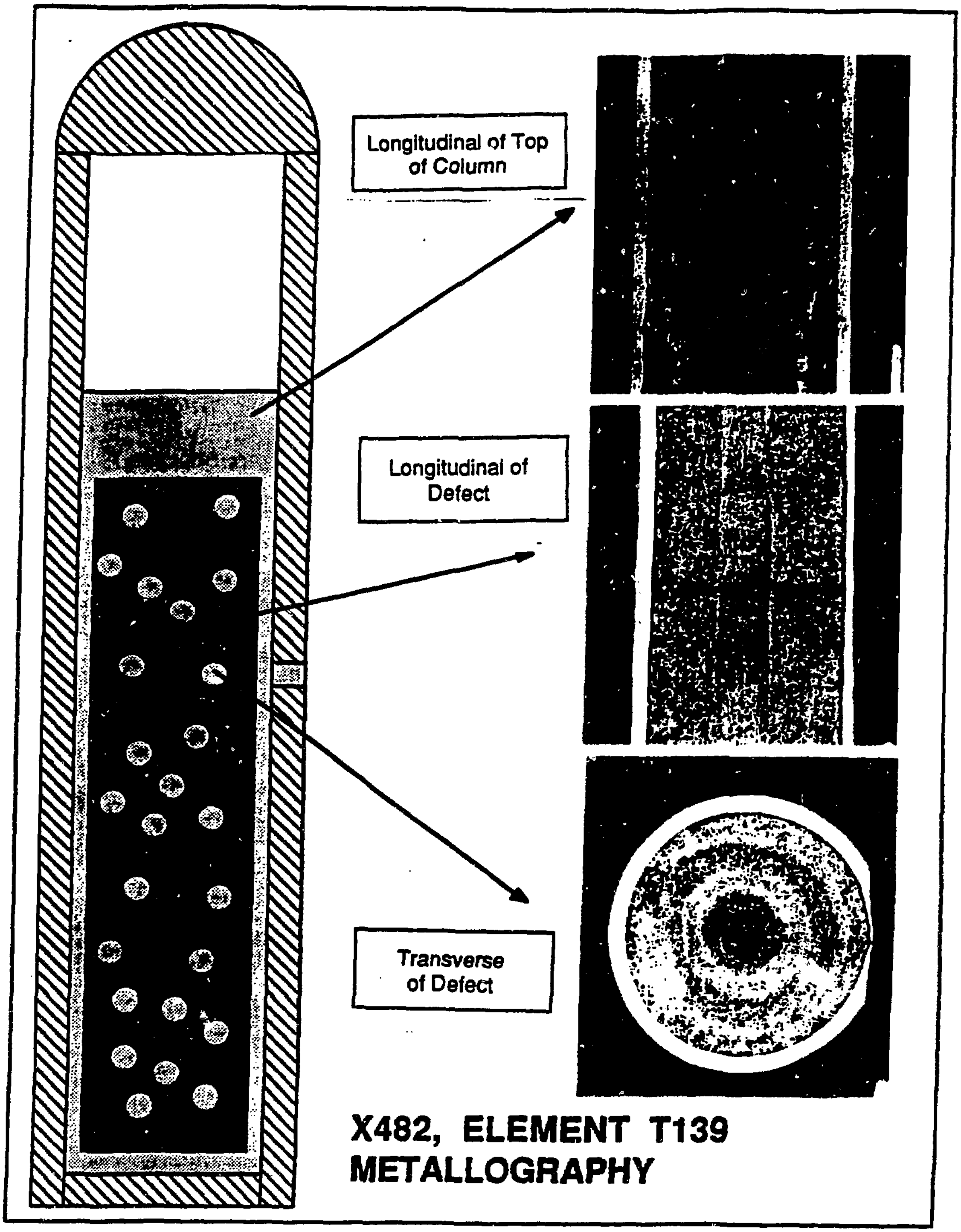

Figure 4. Microstructure of Element T139 from X482 near the Breach Site

breach. Experiment X427 contained all the remaining elements from the in-reactor eutectic penetration tests and enough fresh elements to fill a 91-element bundle.

The XY-22 test indicated that low-burnup elements had more cladding attack than the high-burnup elements. The high-burnup elements failed due to stress rupture in the restrainer dimple (a small indentation in the cladding designed to limit axial fuel growth or motion)—common to normal end-of-life failure. It was expected that the low-burnup elements could breach early due to thinning of the cladding in the fuel region prior to the normal dimple failure, so all elements were irradiated at the same time to determine which would fail first. Breach was detected after an increment of ~2 at.% burnup beyond the initial high-temperature test. Two of the high burnup elements, at 10.0 at.% and 10.2 at.% burnup, both higher than the administrative burnup limit of 8 at.% for the Mark II design, exhibited a weight loss normally associated with breach. This indicated that the normal end-of-life failure mode was to be expected for the high-burnup elements, even after an extended over-temperature event. It was also necessary to determine if the mode of failure changed as a function of burnup. All the medium to high burnup elements were then removed from the subassembly and the low burnup elements were allowed to continue irradiation as X427A until the nominal end-of-life was exceeded. The final burnup for these elements was >11 at.% when the test was terminated without breach.

The X427-X427A tests demonstrated that damage to the cladding due to short term over-temperature events would not significantly reduce the lifetime of these metallic fueled elements. The first failure of a high-burnup element occurred at 10.0 at.% burnup, which is within the 2-σ band for the expected lifetime. Post irradiation examination of the elements showed evidence of previous high temperature attack during the XY-22 irradiation, but no significant cladding degradation. Several examples of fuel restructuring due to the high temperature operation were observed. Figure 5 is an example of an element which was at ~3 at.% when exposed to the over-temperature event. When examined at 5.2 at.% burnup, the normally uniform fuel/clad interaction layer can be seen to be enhanced and much thicker on one side of the element. This reflects the operating conditions in the fuel bundle at the time of the over-temperature event.

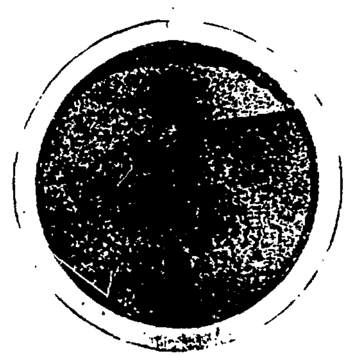

Figure 5. Fuel Structure Following Over-temperature Event and Subsequent Irradiation

The metallic fuel, which had become very porous due to accumulated fission gas, sometimes appeared to sinter and shrink away from the cladding at elevated temperatures at the top of the fuel column. This also had no observable effect on subsequent operation of the fuel for the additional ~2 at.% burnup increment required to obtain the first breach in X427. The dimples, which are under reverse bending, normally begin to crack at end-of-life burnups, and the cracks gradually penetrate from the outside of the element to the inside. Figure 6 shows that all three dimples in the 10 at.% burnup element had cracks initiated at the time of breach.

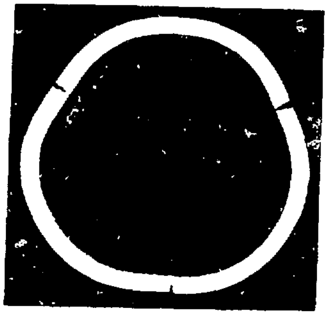

Figure 6. Microstructure at Dimple Region Exhibiting Defects

Examination of the previous low-burnup test elements at termination of the X427A test at 11 at.% burnup indicated that small cracks had initiated at the dimples, but penetration was less than 1/3 the wall thickness. These elements had fuel/clad interaction layers of limited thickness and no significant thinning of the cladding was evident.

Although no fuel failures occurred in the elements exposed to high temperature at low burnup and subsequently irradiated to high burnup, the fuel restrainer dimple in these elements was found to be the weakest point. The dimple acts as a stress riser for both steady-state and over-temperature events. Fuel elements currently used in EBR-II, and proposed for IFR-type reactors, have no fuel restrainer dimples and therefore may be ultimately limited by the cladding wastage incurred during over-temperature events. Design of advanced reactor fuel elements has indicated that predicted stress rupture failure during anticipated over-temperature events at end-of-life may be used to set exposure limits and that limited liquid phase attack may be important only at beginning-of-life when significant thinning of the cladding could occur before failure.

SUMMARY

All test results have demonstrated the robust behavior of metallic fuel and it's potential to achieve high burnup even with aggressively-designed elements. The key characteristics of behavior are now more clearly understood and no limitations to IFR applications have been identified.

The direction of the fuel performance program is now shifting toward the generation of a statistically significant data base to determine whole-core performance of the advanced metallic fuel. The core of EBR-II has been converted to U-10Zr with limited U-xPu-10Zr, where x varies between 8 and 28 wt % plutonium. This conversion will not only offer the opportunity to gain data on large numbers of assemblies, but this fuel will also be the first material introduced into the new IFR reprocessing facility currently under construction at Argonne National Laboratory near Idaho Falls.

Much work remains before metallic fuel can be considered fully licensable. Results to date, however, show that metallic fuel is readily and economically fabricated, is capable of achieving high exposures and long reactor residence times, and possesses unique and promising safety features.

ACKNOWLEDGEMENTS

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