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Steady-state Irradiation Testing of U-Pu-Zr Fuel to >18% Burnup

by

R. G. Pahl and R. S. Wisner
Argonne National Laboratory
P. O. Box 2528
Idaho Falls, ID 83403-2528

and

M. C. Billone and G. L. Hofman
Argonne National Laboratory
9700 S. Cass Avenue
Argonne, IL 60439

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DISCLAIMER

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R. G. Pahl and R. S. Wisner

Argonne National Laboratory
P.O.Box 2528
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M. C. Billone and G. L. Hofman

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ABSTRACT

Tests of austenitic stainless steel clad U-xPu-10Zr fuel (x=0,8,19 wt.%) to peak burnups as high as 18.4 at.% have been completed in the EBR-II. Fuel swelling and fractional fission gas release are slowly increasing functions of burnup beyond 2 at.% burnup. Increasing plutonium content in the fuel reduces swelling and decreases the amount of fission gas which diffuses from fuel to plenum. LIFE-METAL code modelling of cladding strains is consistent with creep by fission gas loading and irradiation-induced swelling mechanisms. Fuel/cladding chemical interaction involves the ingress of rare-earth fission products. Constituent redistribution in the fuel had not limited steady-state performance. Cladding breach behavior at closure welds,in the gas plenum,and in the fuel column region have been benign events.

INTRODUCTION

Argonne National Laboratory has been working for the last five years to develop and demonstrate the Integral Fast Reactor concept (IFR).[1] One of the key aspects of this demonstration is to convincingly demonstrate the safe and reliable performance of the metallic fuel (U-Pu-Zr) during steady-state operation and natural breach. This paper will document the first steps toward this end: the irradiation testing in EBR-II of three lead subassemblies X419, X420, and X421 to burnups exceeding 18 at.% (atomic % heavy metal).

The three lead IFR subassemblies consisted of 61 elements ~63.8cm (~25.1 in) long which were clad in 20% cold-worked D9 (so-called D9C1P). Each subassembly contained a complement of U-xPu-10Zr fuel (where x=0,8, and 19 wt.%). The fuel slugs were one piece solid cylinders 34.3 cm (13.50 in) long x 0.434

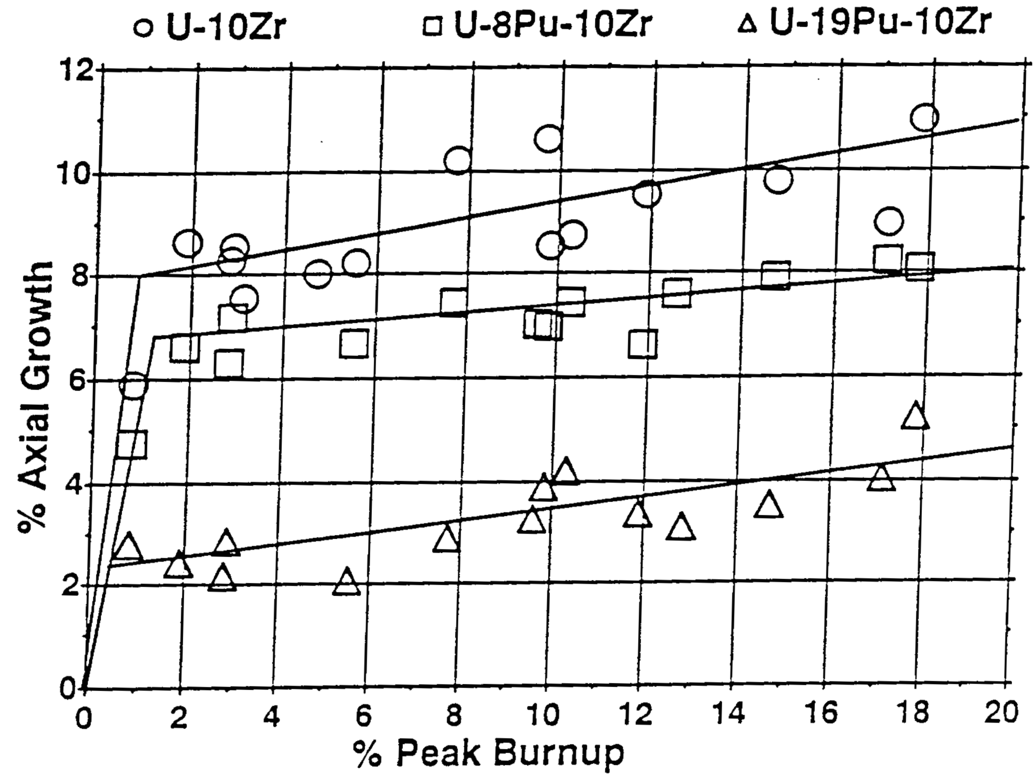

Fig. 1. Burnup dependence of the average axial fuel elongation.

cm.(0.171 in) in diameter, irradiated in the as-injection-cast condition.

The elements were sodium bonded at ~773 K (932° F) for no less than one hour. The cladding outer diameter was 0.584 cm (0.230 in) with a 0.381 cm (0.015 in) wall thickness. The as-built fuel to plenum volume ratio was approximately unity. A 0.107 cm (0.042 in) diameter spacer wire of 20% cold-worked D9 was welded to each end plug with a 15.24 cm (6.0 in) pitch. The closure welds were of the TIG type. Each experiment bore a unique tag gas mixture to aid in breach identification. The beginning-of-life peak inside cladding temperatures ranged from ~813 K (1004 °F) to ~853 K (1076 °F) in the three bundles. The three fuel types were suitably enriched in U-235 to operate in the range of ~377 W/cm to ~475 W/cm (~11.5kW/ft to ~14.5 kW/ft).

FUEL SWELLING AND FISSION GAS RELEASE

Unlike ceramic fuel, metallic fuel operates with a low fuel temperature gradient and retains sufficient fission gas to produce large volumes of fission gas bubbles at low burnup. The resulting swelling is freely promoted in the 73% smear density design of these lead test elements. Interlinkage of gas bubbles and rapid gas release at 1 at.% to 2 at.% burnup mitigates fuel-clad mechanical interaction because the porous fuel is so

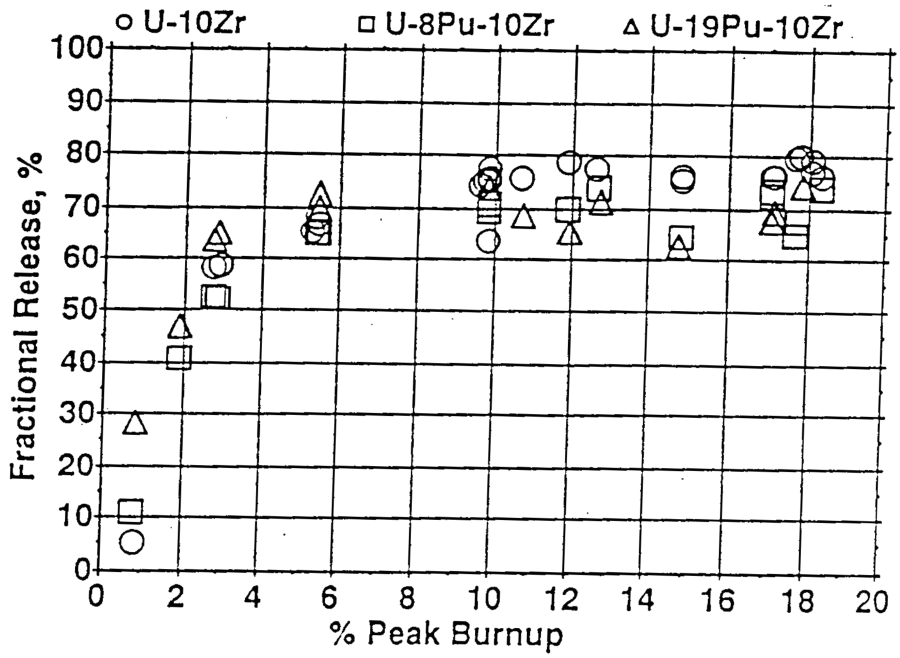

Fig. 2. Burnup dependence of the fission gas release fraction.

plastically compliant at temperature. This effect has been key to the high burnup capability of metallic fuel. Radial fuel growth dominates the swelling process; at ~2 at.% burnup cladding contact causes frictional forces to impede further axial growth. The initial apparent anisotropy has been related to radial gradients in creep strength and irradiation growth of non-cubic phases in the fuel.[2] At high burnup (between 15 at.% and 18 at.% burnup) the gap between fuel (especially U-19Pu-10Zr) and cladding tends to reestablish itself near core midplane due to the high swelling nature of the austenitic cladding at this fluence. Axial growth is an approximately linear function of burnup beyond 2 at.% burnup. Figure 1 shows the average length change as a function of peak burnup in the three fuel types. The 19% fuel shows the greatest degree of anisotropy, due in part to fuel slug cracking which accompanies constituent redistribution during the free-swelling regime below 2 at.% burnup.

The molar quantity of fission gas released to the plenum is nearly linear with burnup. As a consequence, the fractional release asymptotically approaches 80% of that generated at high burnup. This behavior can be seen in Figure 2 for the three alloys tested. Consistent with it's higher axial growth, and therefore greater volumetric swelling,the binary alloy shows slightly higher rates of gas release beyond about 10 at.% burnup.

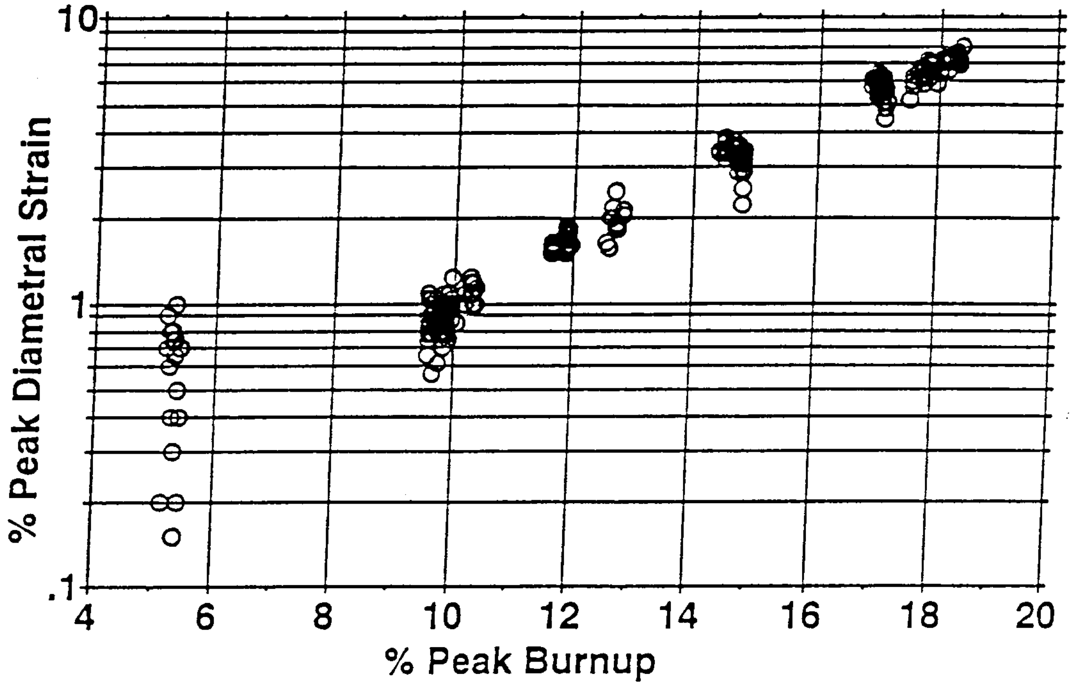

Fig. 3. Burnup dependence of the maximum cladding strain in the experimental D9 fuel elements.

CLADDING PERFORMANCE

The 20% cold-worked D9 cladding performed satisfactorily to the peak exposure reached (~19x10^22 n/cm^2 fast fluence). Peak diametral strain (creep plus swelling) ranged from an average of ~1% at 10 at.% burnup to ~7% at 18 at.% burnup as shown in Figure 3. Because fuel/cladding mechanical interaction is minimized by the low planar smeared density, the high fuel to plenum volume ratio promotes fission gas loading as the principal source of stress. The LIFE-METAL fuel performance computer code was fairly successful in predicting the cladding response in spite of the uncertainties of in-reactor operating conditions and the fact that high fluence swelling and creep correlations for this heat of steel are not particularly well-defined. These predictions are shown in Figure 4 for the case of U-10Zr fuel.

As asserted above, fuel/clad mechanical interaction was found to contribute little to the total creep strain at the burnup levels modelled. For the high burnup case, LIFE-METAL predicted that ~80% of the total strain is due to creep, and 20% due to swelling. Post-irradiation examinations of the high burnup fuel from X420 and X421 are continuing and will serve to benchmark the LIFE-METAL code predictions with immersion density measurements to derive cladding swelling values.

Fuel-clad chemical interaction was most prominent at the hotter elevations of the elements and at 17 at.%-18 at.% burnup took the form of a non-uniform transgranular layer <0.1 mm deep. Electron microprobe examinations of sibling fuel at 5 at.%-10 at.% burnup showed nearly complete depletion of nickel (from ~16 wt.% to ~1 wt.%) and significant enrichment in rare earth fission products (~23 wt.% of Nd,Ce,Sm,Pr, and La). Some loss of iron had also ocurred but no change in chromium content was found. This layer was extremely hard with low ductility at hot-cell temperatures and was about 3 times deeper in the 19% plutonium fuel than the binary fuel. The migration of the rare-earth fission products is enhanced in the coarse porosity of the ternary alloys and tends to agglomerate near the cladding inner surface within pre-existing cavities. Two major rare-earth phases have been observed, differing mainly in their palladium content. Their approximate compositions (wt.%) at 17 at.% burnup are: a)33 Nd,24 La,18 Ce,16 Pd,4 Pr,3 Pu,and 2 U; and b)34 Nd,28 Ce,28 La,6 Pu,and 4 Pr.

CHEMICAL REDISTRIBUTION

Multiphase boundaries present in the fuel during operation lead to annular zones which differ in swelling properties and composition, most prominent in the 19% Pu fuel.[3] By 2 at.% burnup, an interchange between zirconium and uranium occurs. Depending on fuel temperatures, this leaves either a zirconium-depleted shell (<2 wt.%) at mid-radius surrounding a zirconium-rich core or a zirconium depleted nugget at slug center encircled by zirconium-rich fuel. Pie-shaped cracks seen in transverse sections are common at the earliest stages of redistribution but are completely "healed" by fuel growth by 10 at.% burnup. Little redistribution occurs in the other alloys at low burnup. Preliminary electron microprobe data on U-19Pu-10Zr fuel at 18 at.% burnup showed the typical 3-zone structure. The central zone had approximately (wt.%) 14% Pu, 16% Zr (remainder U and fission products). This region was surrounded by a shell with 11% Pu,and only 4% Zr. The outermost region was particularly inhomogeneous with typical compositions of 18% Pu and 24% Zr. A qualitative analysis of a U-10Zr fuel slug at 18% burnup showed a factor of 2 higher in zirconium content at slug centerline versus fuel surface.

BREACH CHARACTERISTICS

The X419 test was reconstituted at 1 at.% and 3 at.% burnup for post-irradiation examination before it was terminated at 11.9 at.% burnup with no breaches observed. The X420 test was reconstituted at 5.5 at.% burnup prior to it's

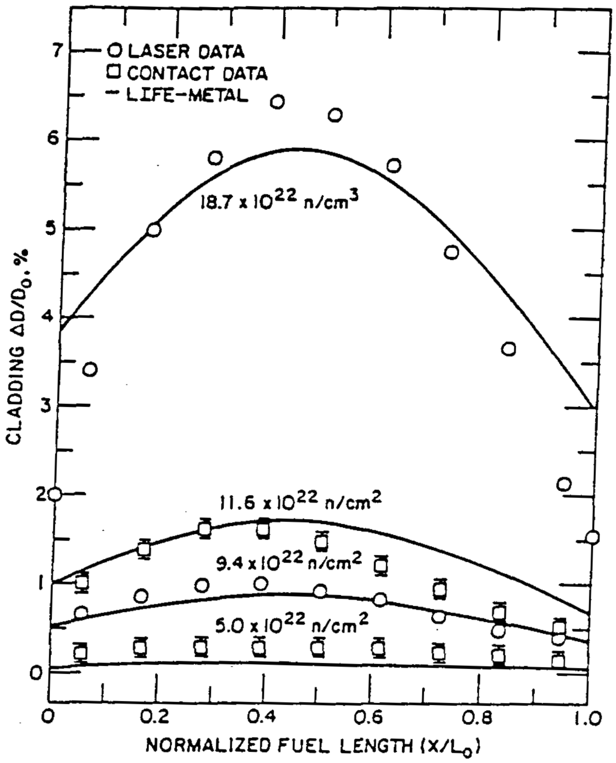

Fig. 4. Comparison betwee LIFE-METAL predictions and data for cladding diametral strain profiles at peak burnups of 5.4 at.%, 9.7 at.%, 11.8 at.% and 17.9 at.%.

first breach at 13.5 at.% burnup. Further reconstitution led to termination at 17.1 at.% burnup, prior to which ten additional breaches occurred. The X421 test was reconstituted at 10 at.% burnup and terminated at 18.4% peak burnup. It suffered from 2 breaches at >17.at.% burnup. Of these 13 breaches, all but 4 occurred in the fusion zone of the upper closure TIG weld remote from the fueled core region. 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 fitup has eliminated the stress-riser effect that caused these early failures, which were obviously unrelated to fuel type.

Three other breaches in the X420 test also occurred above the core region and can be considered non-fuel specific. Though examination of these failures is now just beginning, it would appear from sodium/cesium deposits that the failure site is in the plenum region. These failures occurred ~5-6" above the top of the fuel. Because this is a region of low cladding temperature and strain with no adjacent fuel we can now only speculate that defects during manufacture or reconstitution may be involved. As was the case for the weld breaches, the failure site exhibits little visible deformation and we presume that any cracks present are of a pinhole nature. No delayed neutron signals were associated with these plenum failures though fission gas loss was typically complete.

The only breach observed that may truly be fuel-specific occurred in element T084 from the X420 test at ~16.4 at.% burnup. A good estimate of the breach burnup can be made since tag gas signals and delayed neutron signals were coincident. The failure site is a hairline crack in the fuel region of this U-19Pu-10Zr element, ~9" from the bottom of the fuel column (X/Lo=.67). Expulsion of bond sodium and fission products led to a delayed neutron signal as a burst of ~20 minute duration, approximately 4 times background.

At the cladding outer surface, the crack is ~0.20 inches long by ~0.0002 inches wide and runs parallel to the element long axis (perpendicular to the cladding hoop stress). The main crack is intergranular with a few smaller cracks 2 to 3 grain diameters long clustered nearby. A shallow longitudinal "groove" could be seen on top of and adjacent to the main crack, possibly caused by abrasion with an adjacent element in this tight bundle of highly strained test elements (2/3 of the elements in the bundle had ~4% strain at this location). The crack was opposite T084's own wrapper wire at an azimuthal position where element/element interaction is maximum and tends to flatten the normal cladding circular cross-section. Laser profilometry confirmed the cladding ovality at this elevation.

The crack occurred in an area of intense fuel/cladding solid state interaction which was revealed by an oxalic acid etch. The maximum depth of interaction was ~0.15 mm (~0.006") and was centered on an azimuthal arc of increased interaction of ~30°. The fuel itself appeared to be unaffected by the breach as can be seen in the as-polished section in Figure 5. The annular zones of redistribution were concentric and resembled sibling fuel at like burnup. It is clear that fuel loss in this type of breach is not to be expected. Microhardness values in the interaction layer showed the usual degree of hardening (~750 DPH higher than the unaffected cladding). However, the D9 base metal along the crack path had lower hardness values than samples 90° and 180° away (typically 50 DPH less). It would appear that based on the observations of accelerated fuel/cladding chemical interaction and the cladding softening, nominal cladding temperatures were exceeded for an indeterminate amount of time in-reactor. At this high burnup, cladding dilation, and thermal bow possibly promoted local regions of distorted geometry and coolant flow. Stress rupture of the cladding would thus be promoted in areas of high cladding wastage and lowered creep strength.

EBR-II operated without consequence of this fuel column breach to the scheduled end of the run for an additional 34 days (~0.6 at.% burnup).

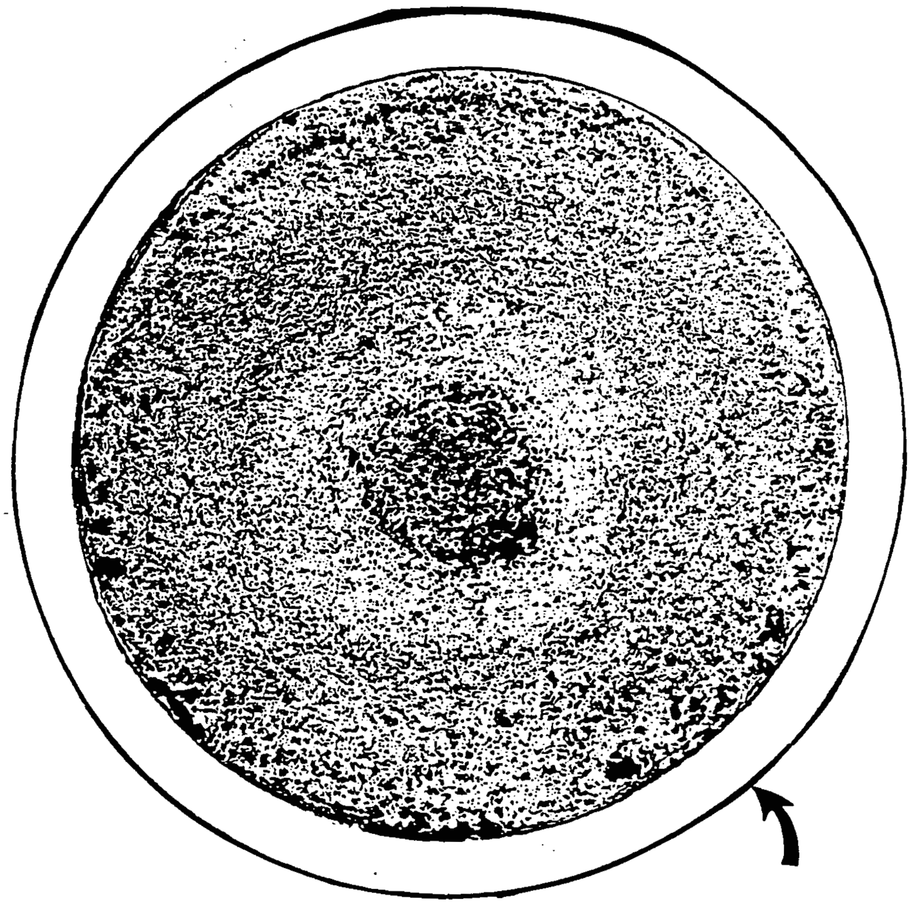

Fig. 5. Transverse section through the fuel column breach site. The arrow indicates location of the crack which is not visible at this magnification.

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