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Letter to the Editors

Irradiation behavior of U–Nb–Zr alloy dispersed in aluminum

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Abstract

Three U–Nb–Zr alloys (U–5Nb–3Zr, U–6Nb–4Zr, and U–9Nb–3Zr) were included in a screening irradiation test of low-enrichment aluminum matrix dispersion fuels. Fuel particles made from these alloys reacted readily with aluminum during fuel fabrication and post-fabrication annealing, resulting in large fuel plate thickness increases. Under irradiation, the behavior of U–5Nb–3Zr (wt%) alloy based fuel was poor at 41 at.% ^{235}U burnup, showing indications of incipient breakaway swelling. The post-irradiation microstructural characteristics of U–6Nb–4Zr based fuel were somewhat better than those of U–5Nb–3Zr, but is marginal at 70 at.% burnup. U–Mo based fuels generally show less reaction on fabrication and better fuel performance characteristics during irradiation.

1. Background

Aluminum matrix dispersion fuel is used in many research and test reactors due to its ability to operate at high power density. Over the last twenty-five years, there has been an ongoing effort to qualify LEU (low-enrichment uranium, <20 at.% ^{235}U) fuels for use in place of HEU (highly-enriched uranium) fuels [1]. Thus far, suitable fuels have been qualified for most research and test reactors [2]; only those operating at very high power densities cannot be converted to LEU with commercially available uranium silicide-based (U_3Si_2) fuel. To do this using existing fuel configurations and fabrication technology requires the use of fuel particles with a uranium density of at least 15 000 kg U/m^3 [3]. While few compound phases meet this criterion, many uranium alloys do, and a series of irradiation tests have been conducted on dispersion fuels using uranium–molybdenum and uranium–niobium–zirconium alloys as the fuel phase. A potential advantage of using niobium and zirconium as alloy constituents in these fuels is their low neutron absorption cross section relative to molybdenum. A brief report on the irradiation performance of U–Nb–Zr alloys under moderate test conditions is given here.

2. Fuel fabrication

Powder was produced for use in these experiments by filing a cast fuel pin with a tungsten carbide rotary file and collecting the chips produced [4]. Chemical analysis of the fuel alloy feedstock used to make the powder is given in Table 1. Up to 3 wt% of tungsten carbide (WC) and cobalt contamination was introduced during filing. Scanning electron microscopy/Energy dispersive spectroscopy (SEM/EDS) examination of the finished plates showed the WC contamination to be in the form of discrete particles separate from the fuel particles, although in a few cases, tungsten was found internal to fuel particles.

The hot rolling process [5] used to fabricate these specimens was conducted over approximately 6.3 ks (1.75 h) using a six pass schedule at 773 K. Final plate thickness was 1.27×10^{-3} m, with a fueled zone 4.2×10^{-4} m thick. Fuel particle loading for all test plates was nominally 25 vol%. After fabrication, the fuel plate specimens were subject to a 758 K 'blister' anneal for 3.6 ks (1 h) in order to identify bond defects in the fuel specimens. During the blister anneal, increases in

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Table 1
Chemical analysis of U–Nb–Zr fuel feedstock

Material	Total U (±0.5 wt%)[a]	^{235}U (±0.5 wt.%)	Nb (±10 wt%)	Zr (±10 wt%)	Mo (±10 wt%)	C (±25 wt%)	O (±25 wt%)
U–6Nb–4Zr	90.6	19.5	5.2	3.4	–	0.009	0.002
U–5Nb–3Zr	92.8	19.4	4.1	2.6	–	0.004	0.005
U–2Mo–1Nb–2Zr	96.4	19.4	0.7	0.7	1.8	0.006	0.004

[a] Relative measurement error at two standard deviations.

the thickness of fuel plates containing U–Nb–Zr alloys were noted [6], as indicated in Table 2. These changes in thickness were significantly larger than those observed for uranium–molybdenum alloys with the same total alloying content. Examples of polished cross-sections from the U–Nb–Zr alloy plate specimens after annealing are shown in Fig. 1. In all cases, the fuel particles reacted extensively with the aluminum matrix. Standardless SEM/EDS suggests that a $(U,Nb,Zr)Al_x$ type mixed intermetallic phase is forming. The formation of a $(U,Nb,Zr)Al_3$ reaction product was observed in other work [7]. Concomitant with this reaction, large voids formed within the fuel meat region. A volume increase of approximately 9% will occur on formation of a $(U,Nb,Zr)Al_x$ intermetallic phase from reaction of stoichiometric amounts of alloy and aluminum. At a fuel plate volume loading of 25%, this reaction would account for a maximum fuel meat volume increase of 2–3%. Void formation thus accounts for the majority of the 8–13.5% thickness change during annealing in the lower alloy specimens.

The amount of swelling due to reaction dropped sharply for alloy contents of greater than about 7 wt% in the U–Nb–Zr system, and greater than 4 wt% in the U–Mo system. Increasing the total alloy content above 10 wt% (as in the case of U–9Nb–3Zr) is not a viable option due to the corresponding decrease in uranium density.

Table 2
Thickness increases during fuel plate fabrication

Fuel alloy	Thickness change (%)
U–2Mo–1Nb–1Zr	13.5
U–5Nb–3Zr	12.4
U–6Nb–4Zr	4.8
U–9Nb–3Zr	4.1
U–4Mo	8.0
U–6Mo	0.1
U–8Mo	None
U–10Mo	None

3. Irradiation testing

Based on evaluation of post-fabrication microstructures, U–2Mo–1Nb–1Zr fuel specimens were determined to be unsuitable for irradiation, and the U–5Nb–3Zr alloy specimens were restricted to low burnup. U–6Nb–4Zr was irradiated along with a series of other alloys to approximately 70% burnup in the RERTR-1 and RERTR-2 fuel irradiation tests.

The fuel test plates were irradiated at the ATR (Advanced Test Reactor) at the Idaho National Engineering and Environmental Laboratory. A description of the irradiation experiment is given in more detail elsewhere [8]. Table 3 gives the calculated burnup and fission density of the test specimens discussed here. Calculations were compared to measured burnup values and found to be within ±2%. Peak fuel plate temperature was approximately 340 K. Included for comparison in this table is an example of a uranium–molybdenum alloy based fuel plate.

Table 3
Irradiation parameters for fuel test specimens

Composition nominal (wt%)	Plate No.	Average fuel density[a] (kg U m^{-3})	^{235}U burnup[b] (avg.%)	Meat fission density[c] (10^{27} m^{-3})	Fuel particle fission density[d] (10^{27} m^{-3})	Average fuel fission rate (10^{20} m^{-3} s^{-1})
U–6Nb–4Zr	F005	4700	70	1.3	4.8	2.4
U–5Nb–3Zr	I005	4300	41	0.8	2.9	3.6
U–10Mo	A005	4500	69	1.4	4.9	2.4

[a] Density calculated from X-ray absorption measurements.
[b] Burnup calculated from ATR core model.
[c] Meat fission density refers to the fission density averaged over the volume of fuel and aluminum in the fueled core of the specimen.
[d] Fuel particle fission density represents fissions only in the fuel particles.

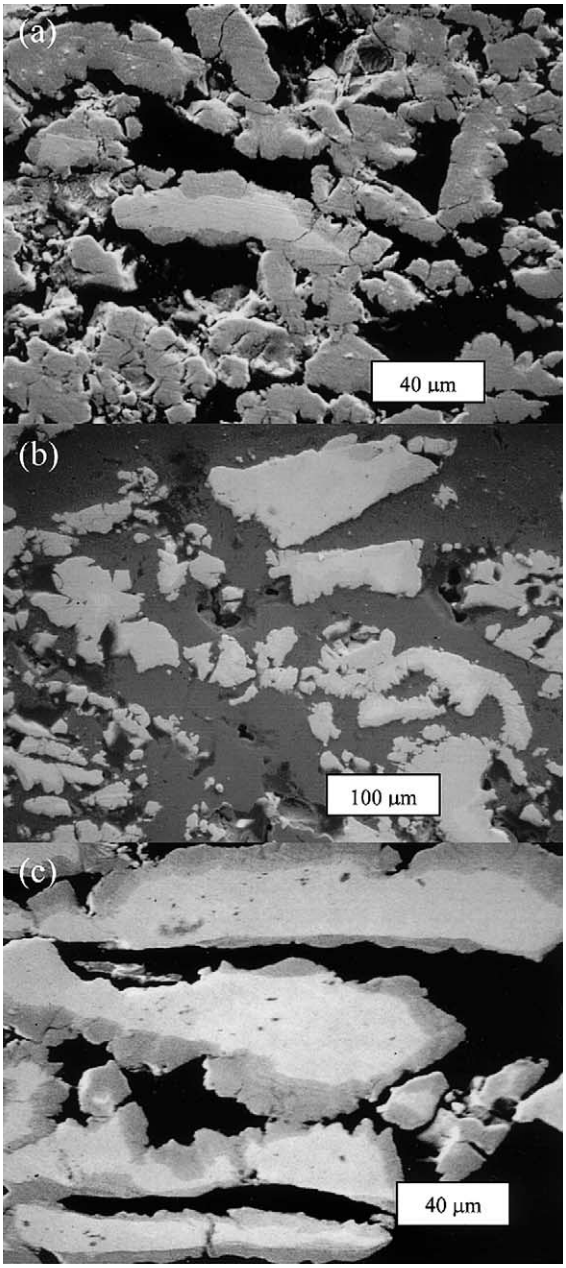

Fig. 1. Pre-irradiation microstructures of fuel test specimens. Dark background phase is the aluminum matrix, light phases are U–Nb–Zr and U–Mo–Nb–Zr fuel alloys, and intermediate contrast phases are (U,Nb,Zr,[Mo]) Al_x reaction products. (a) U–2Mo–1Nb–1Zr, (b) U–5Nb–3Zr, and (c) U–6Nb–4Zr.

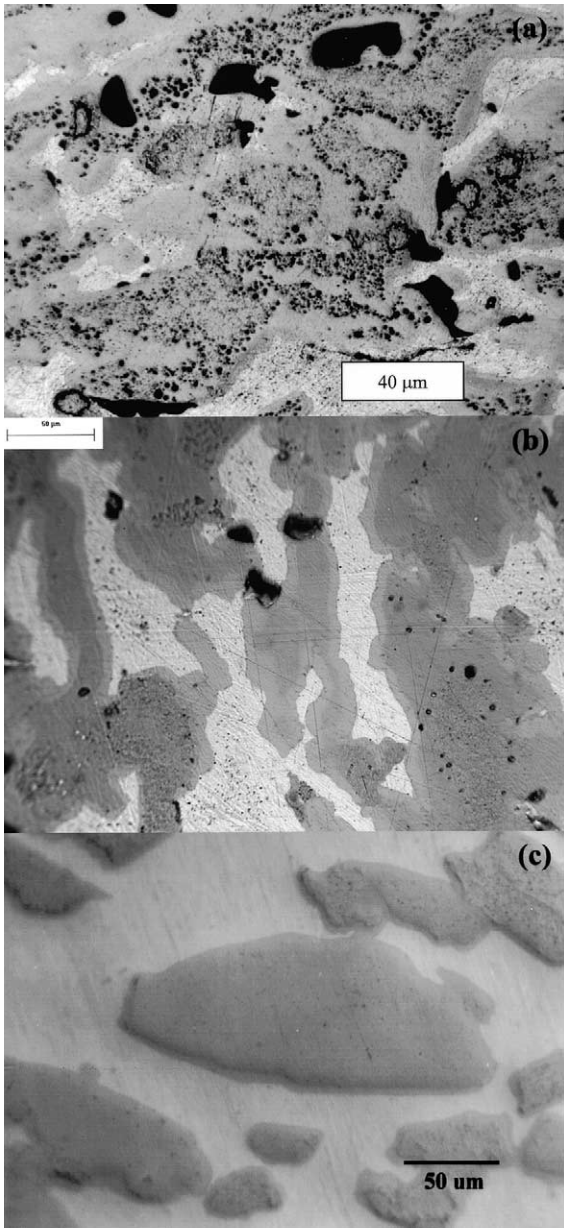

Fig. 2. Post-irradiation optical images. (a) U–5Nb–3Zr plate I005 at 41% ^{235}U burnup. (b) U–6Nb–4Zr plate F005 at 70% burnup. (c) U–10Mo plate A005 at 69% burnup.

4. Post-irradiation examination

An optical micrograph of U–5Nb–3Zr plate I005 at a fuel particle fission density of 2.9×10^{21} fissions cm^{-3} (42 at.% ^{235}U burnup) is shown in Fig. 2(a). Consistent with pre-irradiation characterization, there is extensive reaction of the fuel particles with the aluminum matrix. There is a population of large voids that are remnants of porosity formed during fabrication and annealing; the rounding of the pore edges that has occurred is commonly observed during irradiation of this type of fuel. The aluminide reaction product is relatively free of large gas bubbles. Visible around the periphery of each particle in light contrast is a zone of fuel/matrix interaction approximately 2×10^{-6} m in width that was formed during irradiation. The remainder of the fuel meat consists of the internal volumes of fuel particles that have not reacted with the matrix. The unreacted fuel contains fission gas bubbles of varying size, some of which are linking together to form larger gas pockets. This behavior has been noted as a precursor to breakaway swelling in other dispersion fuels [9,10].

Increasing the total content of niobium and zirconium by ~2 wt% leads to a noticeable differences in fuel behavior. A micrograph of U–6Nb–4Zr plate F005 after irradiation to a fuel particle fission density of 4.8×10^{21} cm^{-3} (70 at.% ^{235}U burnup), is shown in Fig. 2(b). Despite the increased burnup ($1.7 \times$ U–5Nb–3Zr burnup), the fuel exhibits desirable microstructural features relative to U–5Nb–3Zr. Fission gas bubbles in U–6Nb–4Zr at 70% burnup are fewer in number, smaller, and more uniformly distributed. This uniform, stable distribution of small bubbles is desirable in that it shows that fission gas does not have a tendency to coalesce into large pockets that are indicators of incipient breakaway swelling [11]. The fuel/matrix interaction zone that formed in-reactor is approximately 4×10^{-6} m thick at this burnup. Samples of U–9Nb–3Zr fuel were also fabricated and irradiated to 71% burnup. Destructive post-irradiation examination was not conducted on these specimens, however, after it became apparent that the lower uranium density class of U–Nb–Zr alloys offered no neutronic or fuel performance advantage over U–Mo alloys.

A micrograph of a U–10Mo fuel specimen (A005) after irradiation to a fuel particle fission density of 4.9×10^{26} fissions/m^3 (69% ^{235}U burnup) is shown in Fig. 2(c), for comparison to the U–Nb–Zr alloys. This specimen exhibits a uniform distribution of fine fission gas bubbles and less overall fuel/matrix reaction. There are no large gas pockets present. Table 2 shows that negligible fuel plate swelling occurred during fabrication, and post-fabrication microscopic examination showed that only small fuel particles (<5–10×10^{-6} m and some apparently thin regions near particle edges reacted [12]. An interaction layer of average thickness 2.7×10^{-6} m has formed on irradiation, representing a significantly smaller amount of matrix aluminum depletion than that present in U–6Nb–4Zr based fuel.

5. Summary and implications for use of U–Nb–Zr alloys in dispersion fuel

The post-irradiation microstructures of U–Nb–Zr alloys have been compared with those of U–Mo alloys irradiated in the same experiment under the same test conditions. The microstructure of U–5Nb–3Zr at 41% burnup shows features similar to those in fuels known to undergo breakaway swelling. An alloy with an additional 1 wt% of Nb and 1 wt% of Zr exhibited markedly improved post-irradiation microstructural characteristics, so that fuel based on the nominal composition U–6Nb–4Zr has marginally acceptable behavior under irradiation to 70 at.% ^{235}U burnup at 340 K. U–Mo alloys exhibit a stable population of small fission gas bubbles at ~70 at.% burnup and a lower fuel/aluminum reaction layer growth rate. Due to extensive fuel/matrix reaction and marginal fuel performance, the class of U–Nb–Zr alloys has been eliminated from further fuel testing. Subsequent to this test, a high power, high temperature test has been conducted using U–Mo alloys [13] with good results, and two additional tests to an ultimate burnup greater than 80 at.% are underway [14].

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