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Response of dynamically compacted tungsten to high fluence neutron irradiation at 423–600°C in FFTF

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Abstract

When pure tungsten produced by dynamic compaction at 95.3% theoretical density was irradiated in FFTF at temperatures of 423–600°C and neutron doses as high as 14.4×10^{22} n cm^{-2} ($E > 0.1$ MeV), it densified 2–3% and became very brittle. The brittle behavior resulted in failure at grain surfaces and appears not to be related to neutron-induced transmutation or segregation of transmutants. Based on density change measurements, it can be concluded that significant cavity formation did not occur at these high neutron exposures.

1. Introduction

Tungsten and tungsten-rhenium alloys are sometimes mentioned as candidates for plasma-facing components for fusion service [1–3]. There are essentially no data on pure tungsten at high neutron exposure and fusion-relevant temperatures, however, although some irradiation data on W–Re alloys have been published [4–7].

One of tungsten's advertised advantages is that it is inherently low-activation in nature, but unfortunately it is not low-transmutation in nature as well [8]. Tungsten easily transmutes to rhenium in most neutron spectra of interest; rhenium then transmutes to osmium, especially in mixed spectrum reactors [9]. Being a refractory metal, tungsten also is subject to embrittlement by contamination with interstitial elements or by irradiation hardening, leading to an elevation of its ductile-to-brittle transition temperature (DBTT) [3].

In order to obtain some information on the response of tungsten to both transmutation and irradiation hardening, an exploratory experiment was conducted to relatively high neutron exposure in the FFTF fast reactor. A dynamically compacted tungsten, with inherently high dislocation density, was selected for this experiment.

2. Experimental details

Dynamically compacted high purity tungsten was supplied by Lawrence Livermore National Laboratory at 95.3% theoretical density. In dynamic compaction processing, shock waves throw the particulates violently together such that heat is generated rapidly enough that the surfaces of the particulates experience very high temperatures while the particle interiors remain cool. This heat results in melting surfaces, creating bonds between particles without affecting the internal structure of the particles. Dynamic compaction inherently produces a highly deformed structure with the dislocation density far exceeding that obtained by conventional mechanical treatments.

Dynamically compacted tungsten was irradiated as standard 3.0 mm diameter, ~300 μm thick microscopy disks in the Materials Open Test Assembly located in the Fast Flux Test Facility. The specimens were contained in sealed helium-filled stainless steel packets. Two identical specimens were included in each packet. There were two packets at each irradiation temperature and one was removed after each MOTA sequence was completed. Irradiation proceeded in the mid-core region.

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Table 1
Irradiation conditions

Average temperature, °C	Neutron fluence n cm^{-2} ($E > 0.1$ MeV)	dpa (SS)
MOTA 2A		
423	8.53×10^{22}	36.1
520	8.58×10^{22}	36.3
600	7.64×10^{22}	32.3
MOTA 2B		
427	1.42×10^{23}	60.3
520	1.44×10^{23}	60.8
600	1.28×10^{23}	54.2

Active temperature control to ±5°C was maintained during irradiation at temperatures of 423–427°C, 520°C and 600°C for two consecutive irradiation sequences in MOTAs 2A and 2B. The first sequence reached doses of 32–36 dpa as calculated for stainless steel, depending on the irradiation temperature, and those specimens irradiated in both sequences reached 54–61 dpa (SS). The neutron exposures ranged as high as 1.44×10^{23} n cm^{-2} ($E > 0.1$ MeV) and are given in Table 1. At the FFTF midplane W is calculated to experience only 28% of the displacements per dpa that would be produced in stainless steel [9]. After irradiation, either in MOTA-2A, or in both MOTA-2A and 2-B, the specimens were examined using immersion density. The in-cell immersion technique employed has been optimized to yield very accurate values of density on radioactive microscopy disks, and has been shown to yield reproducible results within ±0.2% density change. Specimens from the high exposure sequence were then examined with scanning electron microscopy and energy dispersive *X*-ray analysis.

3. Results

As shown in Table 2, all specimens densified 2–3% independent of irradiation temperature, with most densification completed in the first irradiation segment MOTA-2A. Theoretical density was therefore not attained. In any given packet, however, the two individual specimens tended to develop almost identical changes in density. As shown in Table 2, there was only one exception to this behavior.

The specimens after the second irradiation sequence were found to be very brittle, and at 427°C the specimens from the MOTA-2B discharge broke into many small pieces during the density measurement procedure. Other specimens irradiated at higher temperatures were easily broken with tweezers. The fracture surfaces were primarily intergranular in nature, as shown in Figs. 1–4. There did not appear to be any significant deformation of the grain surfaces.

Calculations of the transmutation shown in Fig. 5 indicate that ∼1% Re and <0.1% Os would have formed in these in-core specimens at the highest exposure level [9], but EDS measurements of the fracture surface in the scanning electron microscope were unable to clearly

Table 2
Density change (%) after irradiation [a]

Preirradiation density g/cm^3	423°C MOTA 2A	427°C MOTA 2A, 2B	520°C MOTA 2A	520°C MOTO 2A, 2B	600°C MOTA 2A	600°C MOTA 2A, 2B
18.41	2.23	2.75	2.05 3.19	2.95	2.07	2.59

[a] Unless two values are given, the density change reported is the average of two separate and identical specimens which agreed within ±0.2% change in density.

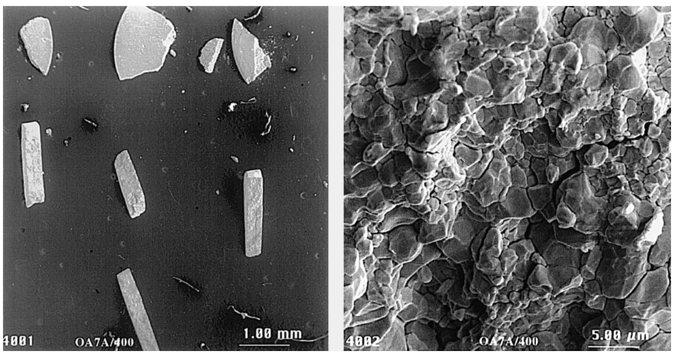

Fig. 1. Fracture surfaces due to failure occurring during density measurement of a specimen irradiated at 423–427°C to 1.42×10^{23} n cm^{-2} ($E > 0.1$ MeV).

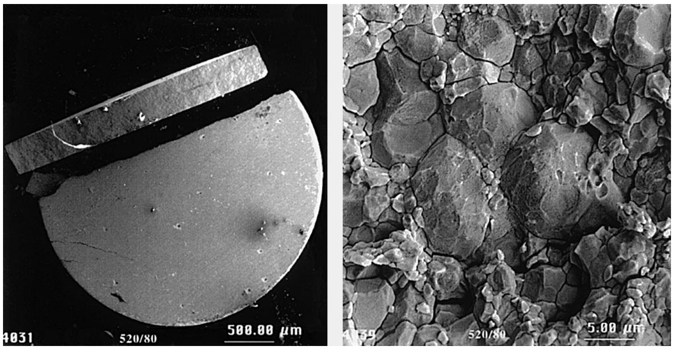

Fig. 2. Failure surfaces resulting from deliberately-induced fracture of a specimen irradiated at 520°C to 1.44×10^{23} n cm^2 ($E > 0.1$ MeV).

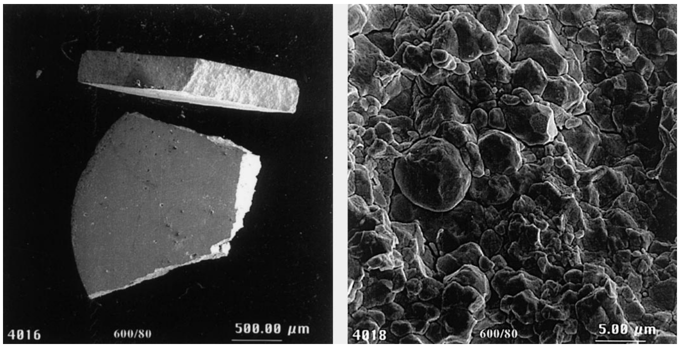

Fig. 3. Failure surfaces resulting from deliberately-induced fracture of a specimen irradiated at 600°C to 1.28×10^{23} n cm^2 ($E > 0.1$ MeV).

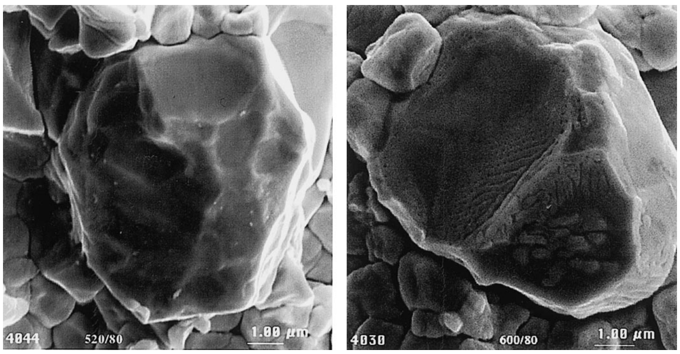

Fig. 4. Large grains on fracture surfaces at 520°C and 600°C at the highest neutron exposure, showing very little surface deformation.

identify either element, showing that their concentration levels near the failure surface were below the resolution limit of the EDS technique.

4. Discussion and conclusions

The temperature-independent densification appeared to be rather reproducible, and probably arose from an early filling of smaller pores. It is thought to most likely have resulted from radiation-enhanced diffusion and perhaps sputtering between particles. The appearance of the fracture surfaces suggests that some intergranular porosity still exists. The densification is not thought to have arisen from any transmutation-related changes in lattice parameter or phase stability.

The undetectibility of Re by EDS, especially on the fracture surface, also suggests that Re did not segregate

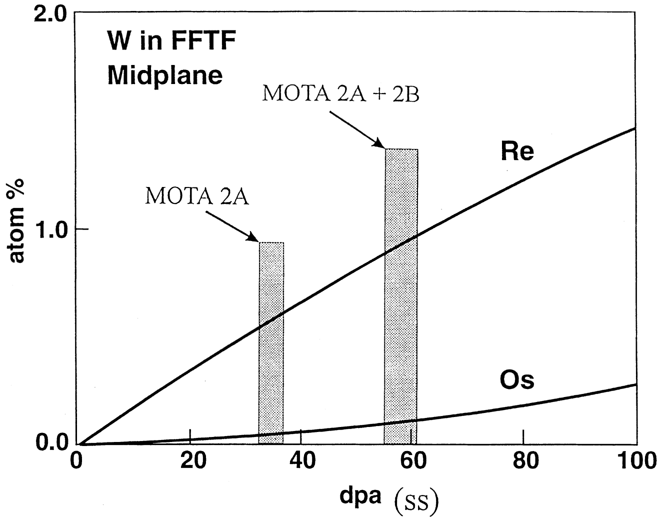

Fig. 5. Calculations of transmutation expected during FFTF–MOTA irradiation [9].

to the grain boundaries significantly. This is consistent with another observation where segregation was not observed at void surfaces in W–25 Re after irradiation at ~600°C in EBR-II [4].

Radiation-induced hardening of the grains and the resultant shift in DBTT, coupled with the existence of preexisting pores between the grains, probably accounts for the brittle behavior, which appeared to be the strongest at the higher neutron exposure. Since the transmutant Re only reached ~1%, one would not expect precipitation to have occurred, but it was unexpectedly observed in W–5 Re and W–11 Re alloys after irradiation in EBR-II [5]. Therefore this assumption may not be correct.

If this irradiation had been carried out below-core in FFTF or in a typical fusion device, the transmutation rate would have been much higher [9]. In some design studies such transmutation is thought to be beneficial, since Re additions to W before irradiation actually improve strength, ductility, recrystallization resistance and machinability, as reviewed in Ref. [10]. This expectation is judged by the authors of this paper to be rather too optimistic, however, since most of the available irradiation data on W and its alloys were developed in fast reactors (as reviewed in Ref. [11]), where the impact of transmutation is relatively smaller. The observation of radiation-induced precipitation in W–Re alloys even in fast reactors [5] probably means that Re formation is detrimental, however. Two studies have directly addressed the effects of transmutation due to thermal neutrons [12,13] and focused on the resultant losses in ductility.

In parallel research conducted in the same capsules as the tungsten specimens [14], dynamically compacted Ti-modified austenitic stainless steel was irradiated at identical temperatures and neutron doses. It was demonstrated that by tailoring the microstructure (i.e., dislocation density) and the microchemistry (i.e., C and Ti distribution in austenite) of the steel, bubble or void formation can be suppressed up to the highest neutron dose studied. Based on density measurements, one can similarly conclude that bubble or void formation was suppressed in the dynamically compacted tungsten. Microscopy of dynamically compacted Ti-modified austenitic steel showed [14] that some recovery of dislocation structure took place during neutron irradiation to 60 dpa at 600°C. On the other hand, the recrystallization temperature of tungsten is significantly higher than that of austenitic steel, thus enabling the probable retention of dislocation structure to higher service temperatures.

In general, it appears that dynamically compacted pure tungsten will not perform well in intense neutron fields at temperatures in the 400–600°C range, and its use for plasma-facing components should be carefully evaluated.

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