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Irradiation effects in tungsten-copper laminate composite☆

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HIGHLIGHTS

- Fusion reactors need a tough, ductile tungsten plasma-facing material.
- The unirradiated tungsten-copper laminate is more ductile than tungsten alone.
- After neutron irradiation, the composite has significantly less ductility.
- The tungsten behavior appears to dominate the overall composite behavior.

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ABSTRACT

Tungsten-copper laminate composite has shown promise as a structural plasma-facing component as compared to tungsten rod or plate. The present study evaluated the tungsten-copper composite after irradiation in the High Flux Isotope Reactor (HFIR) at temperatures of 410–780 °C and fast neutron fluences of 0.02–9.0 × 10^{25} n/m^2, E > 0.1 MeV, 0.0039–1.76 displacements per atom (dpa) in tungsten. Tensile tests were performed on the composites, and the fracture surfaces were analyzed with scanning electron microscopy. Before irradiation, the tungsten layers had brittle cleavage failure, but the overall composite had 15.5% elongation at 22 °C. After only 0.0039 dpa this was reduced to 7.7% elongation, and no ductility was observed after 0.2 dpa at all irradiation temperatures when tensile tested at 22 °C. For elevated temperature tensile tests after irradiation, the composite only had ductile failure at temperatures where the tungsten was delaminating or ductile.

1. Introduction

For future fusion reactors, there is a need for both armor tungsten, which shields the underneath structural materials from the high heat flux and particle flux from the plasma, as well as structural tungsten, which may act as armor tungsten as well as carrying a load or performing a function such as a pipe for cooling water [1]. The main challenges that need to be overcome to make a structural tungsten material are lowering the ductile to brittle transition temperature (DBTT) and improving the fracture toughness of tungsten [2]. There are several possible ways to improve these properties in tungsten such as alloying, grain boundary engineering, or creating a composite [2]. The method that has shown the most promise thus far is composites, here, namely laminate composites.

Tungsten foil is ductile at room temperature in tensile and bending tests due to the small grains, the high amount of mobile edge dislocations, and the “foil effect” of dislocations annihilating on the surface [3,4]. The DBTT is shifted to lower temperatures due to the effects the cold rolling has on the microstructure [5]. To create a successful composite requires utilizing an interlayer to join tungsten foils together into a macroscopic material while still maintaining the beneficial properties of the foil. For joining methods that use heat, the temperature should be kept below 1100 °C to retain the foil microstructure and prevent

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recrystallization of the foils [6]. Additionally, the interlayer should be something that does not form a brittle intermetallic with tungsten so that the interface remains sharp [6]. Copper (T_{melt} = 1085 °C) meets these requirements, so was chosen as the interlayer for this study even though the drawback of copper is that it would lower the operating temperature of the tungsten-based material.

Previous work has examined the Charpy and bending properties of the tungsten-copper laminate, or the similar copper silver eutectic alloy with tungsten laminate composite, with excellent results as compared to plate tungsten properties. The laminate had greatly improved Charpy results at all tested temperatures up to 500 °C compared to tungsten plate [2]. Many mechanisms of crack deflection in a laminate composite rely on the different layers having different properties [7], which is certainly the case for the tungsten-copper laminate. Even when annealed and recrystallized tungsten foil was used in the laminate, the laminate Charpy properties were significantly better than the tungsten plate up to 1000 °C [2]. The fabrication technology has advanced to produce pipes from the tungsten-silver copper composite which had superior properties to pipes cut from tungsten rods [2].

Although the previous work is promising, the remaining question is how the tungsten laminate performs under neutron irradiation. There is some evidence that nanostructured materials, such as tungsten foil, may perform better under irradiation due to the additional defect sinks [8], and the foil layers may allow dislocation annihilation on the surfaces [3] but this must be investigated after neutron irradiation. The specific change in tungsten's properties depend on the neutron energy spectrum, but irradiations conducted in reactors including JOYO, JMTR, the Oak Ridge Research Reactor, and the High Flux Isotope Reactor (HFIR) all agree that tungsten hardens and embrittles over a wide range of irradiation conditions and temperatures up to at least 900 °C [9,10]. Thus, for any fusion reactor, a successful tungsten-based material would have to mitigate the negative effects of neutron irradiation on tungsten's behavior in addition to overcoming tungsten's inherent brittleness. In this study, tungsten-copper laminate material was irradiated in HFIR at temperatures between 410 and 780 °C to fast neutron fluences of 0.02–9.0 × 10^{25} n/m^2, E > 0.1 MeV, which are 0.0039–1.76 displacements per atom (dpa) in tungsten, to determine if the ductile copper, unique microstructure of the tungsten foil, and additional interfaces at the foil surfaces could improve the behavior under neutron irradiation compared to tungsten alone.

2. Methods

Copper sheets 0.1 mm thick were brazed to the rolled tungsten sheets, 0.1 mm thick and 99.97% pure. For this study, the composite consisted of three tungsten layers alternating with two copper layers for a total thickness of 0.5 mm. The thickness of the copper interlayer influences the strength of the composite, thinner is better; the thermal conductivity of the composite, thicker is better; and the maximum operating temperature of the composite, thinner is better [6]. For the 0.1 mm case used here, the thermal conductivity is higher than the 0.025 and 0.010 mm cases previously fabricated [6], but this layer thickness is essentially macroscopic to the defects as different strengthening mechanisms only take over at layer thicknesses less than approximately 1 μm [11].

For fabricating the composite, a tungsten weight was placed on the foil stack while it was heated in a high vacuum furnace to approximately 1100 °C for brazing using the method described in Ref. [2] with more details on the tungsten-copper version of the laminate in Ref. [6]. The tungsten foils had grains elongated in the rolling direction with approximately an aspect ratio of 1:30 [4]. The surface of the tungsten foils had primarily (100)<011> texture, which for BCC tungsten means that the preferred cleavage planes are at 45° to the rolling direction [4]. Tensile samples in size SSJ-2, with outer dimensions 16 mm × 4 mm × 0.5 mm, were cut from the laminate such that the tensile direction matched the rolling direction, and the normal direction of the foils was also the normal direction of the tensile bars.

The tensile samples of the composite were irradiated in rabbit capsules in the HFIR at selected temperatures between 410 and 780 °C to fast neutron fluences of 0.02–9.0 × 10^{25} n/m^2, E > 0.1 MeV. The fill gases used in the rabbits were He, Ne, and Ar with the precise gas mixture chosen for each rabbit to achieve the desired irradiation temperature. Displacements per atom (dpa) values discussed in this paper are only for the tungsten and use 0.195 dpa = 1 × 10^{25} n/m^2 (E > 0.1 MeV). This correlation for HFIR fluence to tungsten dpa is discussed in the article by Sawan [12], and this value agrees with the output from the SPECTER code [13]. Again, all dpa values mentioned in the text after this point are only for tungsten, but for comparison, using the SPECTER code for the same HFIR case as for tungsten above gives the conversion for copper as 0.948 dpa = 1 × 10^{25} n/m^2 (E > 0.1 MeV). Each rabbit capsule included several passive SiC temperature monitors. After irradiation, these temperature monitors were analyzed using thermal expansion from continuous dilatometry, and the data were analyzed using the recently developed software [14]. An ANSYS thermal model of each irradiation capsule was used to relate the calculated SiC temperature to the temperature experienced by the tungsten composite samples.

After irradiation, samples were examined in the Low Activation Materials Development and Analysis (LAMDA) Laboratory at ORNL. Tensile tests were performed at 22 °C and selected elevated temperatures at an extension rate of 0.02 in/min. Elevated temperature tests were performed in a vacuum furnace with background pressures during tests in the range of 10^{-5} to 10^{-6} Torr. For the nanoindentation, a loading rate of 100 μN/s was used for the tungsten layers and a rate of 50 μN/s was used for the copper layers; all indents were done to 500 nm depth. The different loading rates were chosen as a balance between collecting enough data points, better at slower rates, and reducing the drift and chance of room vibrations interrupting the test, better at faster rates. The nanoindenter used a Berkovich tip (3-sided pyramid) and was calibrated by the vendor during installation. The Analyst Data Analysis Tool version 2.3.4 © MTS Systems Corporation was used to analyze the nanoindentation data; the program uses the Oliver-Pharr data analysis procedure to determine the hardness from the nanoindentation data [15,16]. Fracture surfaces were examined with a Keyence digital optical microscope and a scanning electron microscope.

3. Results and discussion

Tensile tests were performed on the copper-tungsten samples at room temperature and selected elevated temperatures, summarized in Table 1. The Sample IDs in Table 1 refer to the engraved identifiers on the samples, which are tracked in the ORNL database. To aid with comparing the many different samples, throughout the text, unirradiated samples will be referred to by their Sample ID and their test temperature, for example, SW40-22 for the unirradiated sample SW40 tested at 22 °C. For irradiated samples, the text labels will include the Sample ID, dpa, irradiation temperature, and test temperature in that order. For example, SW15-0.43-710-22 was irradiated to 0.43 dpa at 710 °C and tested at 22 °C.

In Table 1 for brittle samples, no yield stress (YS) is listed, and the fracture strength is listed in the ultimate tensile strength (UTS) column. All the tensile tests were performed without extensometers, so the cross-head motion was recorded to estimate the elongation of the samples. Without extensometers, machine