Irradiation Damage Behavior in Novel High-Entropy Carbide Ceramics

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INTRODUCTION

High-entropy materials contain four or more elements in equal or near-equal concentration and form a stable single-phase crystal structure due to high configurational entropy. They are more thermodynamically stable than traditional materials at high temperatures due to their low Gibbs free energy. High-entropy alloys (HEAs) attracted extensive research interest in the last decade. Studies on metallic HEAs revealed their superior mechanical properties, corrosion resistance and irradiation tolerance. In contrast, very few high-entropy ceramic (HEC) materials have been discovered and investigated.

Although carbide ceramics such as ZrC and HfC have high melting temperatures and mechanical strength, irradiation-induced damage, such as amorphization, limits their structural material applications in Gen-IV nuclear systems. We hypothesized that the irradiation resistance of carbide ceramics can be improved through high-entropy effects, due to slow energy dissipation that can positively affect defect recombination dynamics.

This paper reports our preliminary results on the irradiation damage behavior of a novel HEC material, $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$. The irradiation damage study will provide fundamental knowledge in terms of microstructural stability and the nature of irradiation defects in this novel ceramic material. Due to the large neutron capture cross section in hafnium, the next step of material development will focus on HECs without hafnium element.

RESULTS

The starting powders are a mixture of HfC, TaC, TiC, ZrC and NbC with an equimolar composition. Bulk $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$ was synthesized by spark plasma sintering (SPS) at 2000°C under a pressure of 30 MPa.[1] XRD reveal that the $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$ possessed a single-phase rock salt structure, in which the five metal elements (Hf, Ta, Ti, Zr, and Nb) were randomly distributed on the cation site and C atoms occupied the anion site.

The $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$ samples were irradiated with 120 keV helium ions at 25 ºC at the Texas A&M Accelerator Laboratory. Transmission electron microscopy (TEM) characterizations showed that helium bubbles with a nominal diameter of 1 nanometer appeared at depths ranging from 220 to 470 nm following helium irradiation (Fig. 1b). The number density of helium bubbles was measured from the cross-sectional TEM images at different depth (Fig. 1a).

The simulated depth profile of implanted helium in $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$ (Fig. 1b) was calculated using SRIM (Fig. 1b). [2] The SRIM calculated helium distribution correlates well with the TEM derived number density of helium bubbles. The maximum helium concentration (~1.8×10^5 ppm) and the maximum number density of helium bubbles (0.00117 nm^{-3}) both appeared at a depth of around 380 nm. Helium bubbles were not observed in the TEM images when the helium concentration was below 25000 ppm. It is noted that despite substantial variation of the helium concentration along the depth, the diameters of helium bubbles are almost the same, i.e., around 1 nanometer. This indicates that the motion and coalescence of helium bubbles was significantly suppressed, which may be related to the chemical disorder and lattice distortion caused by the multiple principal metal elements.

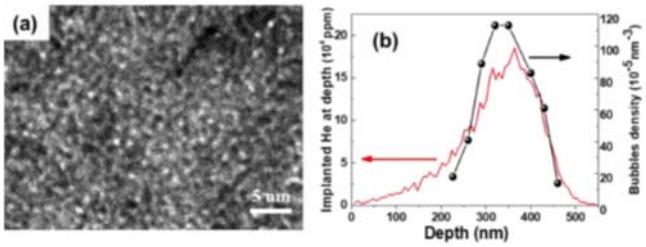

Fig. 1. (a) Bright-field TEM image of helium bubbles after 120 keV helium ion irradiation at 25 ºC. (b) The depth profiles of SRIM-calculated helium concentration and the TEM derived number density of helium bubbles.

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