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Thermal stability of helium bubble superlattice in Mo under TEM in-situ heating

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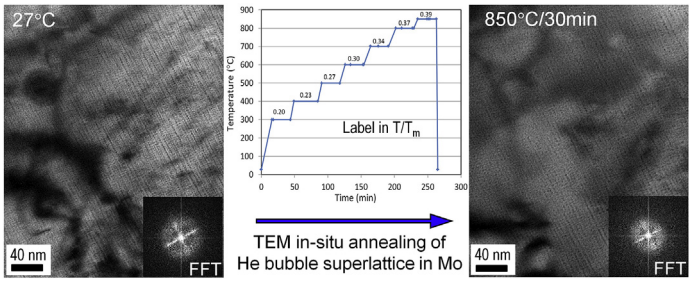
GRAPHICAL ABSTRACT



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ABSTRACT

Although the temperature window of helium ion irradiation for gas bubble superlattice (GBS) formation was found to be in the range of approximately 0.15–0.35 melting point in literature, the thermal stability of He GBS has not been fully investigated. This work reports the experiment using an in-situ heating holder in a transmission electron microscope (TEM). A 3.0 mm TEM disc sample of Mo (99.95% pure) was irradiated with 40 keV He ions at 300 °C to a fluence of 1.0E+17 ions/cm^2, corresponding to a peak He concentration of approximately 10 at.%, in order to introduce He GBS. In-situ heating was conducted with a ramp rate of ~25 °C/min, hold time of ~30 min, and temperature step of ~100 °C up to 850 °C (0.39T$_m$ homologous temperature). The result shows good thermal stability of He GBS in Mo with no noticeable change on GBS lattice constant and ordering. The implication of this unique and stable ordered microstructure on mechanistic understanding of GBS and its advanced application are discussed.

1. Introduction

Gas bubble superlattice (GBS) in metals irradiated with inert gas ions has been investigated extensively by Johnson and others in the past [1–14]. The formation of helium GBS in body-center-cubic (bcc) metal appears the most successful among all the inert gas ion irradiations. The possible mechanisms of the self-organization nanostructures as a result of irradiation are outlined by Ghoniem et al. [15] with a few recent additions [16,17]. GBS formations are reported in bcc, face-center-cubic (fcc), and hexagonal metals irradiated with noble gas helium, neon, and krypton ions. The degree of ordering varies depending on the material, ion species, and irradiation condition although in general the degree of bubble ordering is considered to be low. The work on helium-irradiated Mo indicates that a GBS occurs between the irradiation temperatures of ~0.15T_m and 0.35T_m (T_m the melting temperature in K) [13]. For Mo the

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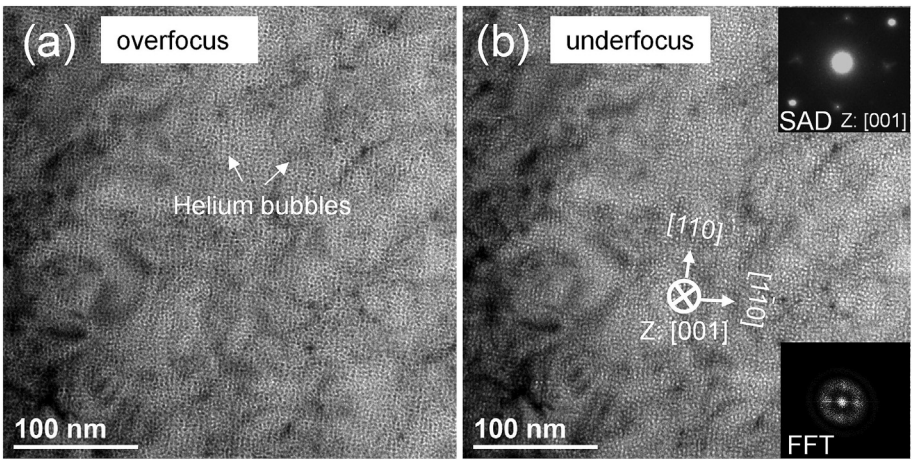

Fig. 1. TEM bright field images of helium gas babble superlattice imaged at zone [001] in (a) over-focus and (b) under-focus condition. Insets show the selected area diffraction (top) and fast Fourier Transform (bottom) that confirms the bubble ordering.

melting temperature and the corresponding irradiation temperature window to form He GBS are 2623 °C and 161–741 °C, respectively. The recent discoveries of highly-ordered GBS consisting of mostly fission gaseous atom Xe (Xe/Kr ~ 10) in the U-Mo fuels irradiated in reactors stimulated great interest on the GBS [18-21]. The degree of ordering in Xe GBS in irradiated U(Mo) fuel is significantly better than that reported in He GBS in pure metals in the literature. Xenon GBS is identified to have an fcc structure in the bcc host material of U-7Mo with an average bubble size and bubble lattice constant of ~3.1 nm and ~12 nm, respectively. This is contradictory to the GBS found from noble gas ion irradiations where all the GBS reported so far have the same structure as their host material. The exceptional performance of U-Mo fuel in geometrical stability, mechanical integrity and fission gas retention is attributed to the high stability of Xe GBS under extreme irradiation conditions up to intermediate fission density.

In addition to radiation stability, thermal stability of a GBS is equally important. The thermal annealing study on the GBS may lead to a better understanding of the development of this unique self-organized microstructural feature and its potential application to be used as a functional material over a broad temperature range. In-situ heating in TEM is a popular technique to investigate the dynamic response of the microstructure or chemical process as a function of temperature [22–28]. It allows tracking the thermal response of microstructural features such as defect clusters, bubbles, voids, loops, precipitates, dislocation, and grain boundary down to nanometer scale. The previous work on TEM in-situ heating of Xe GBS in the irradiated U-Mo fuel revealed exceptionally high thermal stability up to 850 °C ($0.72T_m$) [29]. Contradictory result for thermal annealing of He GBS in copper foil indicated bubble growth and coalescence at 325 °C ($0.44T_m$) and the formation of blisters at higher temperatures [30]. The objective of this work is to investigate the thermal stability of the He GBS in Mo under TEM in-situ heating. The result will be compared with literature to gain the insight of GBS thermal stability.

2. Experiment

The Mo sheet with a thickness of 250 μm and purity of 99.95 wt.% was acquired from GoodFellow. Helium ion irradiation for 3.0 mm TEM disc samples was performed at Los Alamos National Laboratory using an ion implanter with beam energy of 40 keV and an ion flux of 7.6×10^{12} ions·cm^{-2}·s^{-1} to a fluence of 1.0×10^{17} ions·cm^{-2} at an irradiation temperature of 300 °C. A JEOL-2010 TEM/STEM 200 kV microscope, equipped with a LaB_6 filament and a Gatan UltraScan 1000 digital camera, was used. TEM samples were prepared using twin-jet electrical polishing in a 12.5% sulfuric acid and 87.5% methanol solution at temperature of 5 °C to perforation. Fig. 1 shows the TEM bright field images of He GBS at zone [001] in over-focus and under-focus conditions along with the insets showing the selected area diffraction (SAD) from Mo at zone [001] and the Fast Fourier Transformation (FFT) where four faint spots and their orientation indicate bubble ordering coherent to the host material Mo.

The Gatan double-tilt heating holder (Model 652-Ta) used in this experiment has a type-R thermocouple spot-welded to the miniature furnace body to measure the furnace temperature. The actual temperature of the 3.0 mm Mo disc specimen in a heating holder is likely to be slightly lower than the measured furnace temperature. This is because heat transfer under vacuum is mainly by conduction and specimen temperature is affected by the uncertain quality of the mechanical contact between the furnace and the specimen. At high temperature, it is anticipated that the difference between the thermocouple reading and actual temperature on the specimen diminishes since heat transfer by thermal radiation starts to predominate. The in-situ heating was conducted with a TEM column vacuum of ~8.0×10^{-6} Pa (~6×10^{-8} torr), temperature ramp rate of ~25 °C per minute and a hold time of ~30 min starting at 300 °C with a step of 100 °C and finished at 850 °C which is the maximum safe operation temperature for this holder. The temperature vs. time along with the homologous temperatures for Mo is shown in Fig. 2.

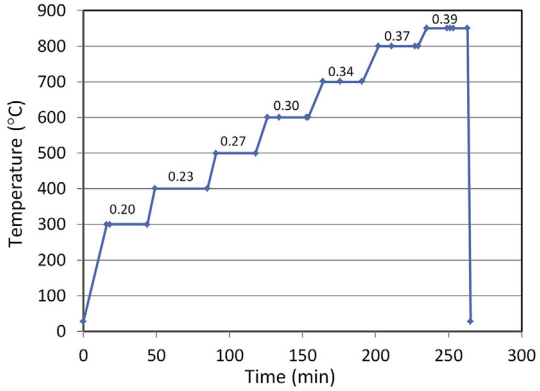

Fig. 2. TEM in-situ heating profile (temperature vs. time) and the corresponding homologous temperatures for Mo.