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Investigation of hydrogen bubbles behavior in tungsten by high-flux hydrogen implantation

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HIGHLIGHTS

- The formation and growing up behavior of hydrogen bubbles in tungsten were investigated by TEM observation.
- Hydrogen bubbles' size appeared larger with higher beam flux until saturated at a certain flux.
- Hydrogen-implanted W samples were annealed to understand the thermal annealing effect on the hydrogen bubbles.

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ABSTRACT

Hydrogen isotopes retention and bubbles formation are critical issues for tungsten as plasma-facing material in future fusion reactors. In this work, the formation and growing up behavior of hydrogen bubbles in tungsten were investigated experimentally. The planar TEM samples were implanted by 6.0 keV hydrogens to a fluence of $3.38 \times 10^{18}\ H \cdot cm^{-2}$ at room temperature, and well-defined hydrogen bubbles were observed by TEM. It was demonstrated that hydrogen bubbles formed when exposed to a fluence of $1.5 \times 10^{18}\ H \cdot cm^{-2}$, and the hydrogen bubbles grew up with the implantation fluence. In addition, the bubbles' size appeared larger with higher beam flux until saturated at a certain flux, even though the total fluence was kept the same. Finally, in order to understand the thermal annealing effect on the bubbles behavior, hydrogen-implanted samples were annealed at 400, 600, 800, and 1000 °C for 3 h. It was obvious that hydrogen bubbles' morphology changed at temperatures higher than 800 °C.

1. Introduction

The materials in the next generation of nuclear reactors will be subjected to greater extremes than those currently in service, such as higher temperatures and greater radiation induced damage levels [1–3]. Along with the implementation of International Thermonuclear Experimental Reactor (ITER) project, tungsten (W) is considered as a primary candidate material for the divertor upper baffle in future nuclear fusion devices, due to its low sputtering yield, low solubility for hydrogen isotopes and high thermal conductivity [3–5]. As plasma-facing materials, tungsten will be subjected to variety of irradiations, especially large flux (10^{22} – $10^{24}\ ions \cdot m^{-2} \cdot s^{-1}$) and low-energy (from tens of eV to several hundreds of eV) hydrogen (H) isotopes and helium (He) ions escaping the plasma, as well as energetic neutrons resulted from DT reaction [3–8]. Fusion neutrons will result in lattice damage throughout the bulk of materials and meanwhile, the implanted H/He atoms will unavoidably interact with this kind of lattice damage and finally affects the tungsten's performance as divertor material [4,5,9–11].

There have been plenty of studies on hydrogen isotopes behavior in tungsten including diffusion, permeation, trapping and desorption by intrinsic defects and irradiation induced damage [4,5,9–16]. These behaviors are supposed to lead to the generation of hydrogen bubbles whom are blamed for degrading the mechanical properties and the performance of the metal [4–8]. Particularly, it was confirmed from experiments that low-energy (from tens of eV to several hundreds of eV) hydrogen isotopes

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irradiation can form a surface layer a few micrometers deep consisting of bubbles, blisters, pores, fuzz, and further development to the surface swelling, blister bursting, and exfoliation [3–6,10–12]. While larger scale damage phenomena occur well below the theoretical damage threshold energy and beyond the ion penetration depth, fusion neutrons will produce lattice damage throughout the bulk of materials, thus posing additional concerns for hydrogen inventory in tungsten [1–5,8].

Experimentally, hydrogen implantation has been used as an efficient method for the evaluation of hydrogen retention in various metals, in which both the introduction of gas species and displacement damage happened simultaneously [9–11,17–21]. Thus, the nucleation and growth behaviors of hydrogen bubbles resulted from H-vacancy interactions could be identified [18–20]. For a minimum displacement energy of 40 eV in tungsten, hydrogens with a minimum energy of approximately 2.0 keV are required to create lattice displacements [11,17–19]. Therefore, hydrogens with an energy of 6.0 keV was employed to study the dependences of hydrogens' behavior on implantation conditions in tungsten, particularly beam flux and total fluence. While neutron-induced damage extends to a larger depth, it is noticed that keV hydrogen induced damage mainly concentrated in a narrow region below the surface (the damage zone).

In the present work, thinned W foils with a thickness of $\sim$ 100 nm were implanted with high-flux hydrogens to simulate properly the effect of displacement damage on hydrogens retention in W matrix. However, because of the high diffusivity, certain hydrogens released from the W foils as a result of the surface effect, which is different from the hydrogens diffusion and retention at depths far beyond the damage zone in the bulk tungsten [3–5,11,12]. Therefore, a focused study on the investigation of hydrogen bubbles' dependence on the implantation conditions was conducted by $ex-situ$ transmission electron microscopy (TEM) observation, which gives direct observations of hydrogen bubbles' morphology in the damage zone. While the irradiation at higher temperature was carried out to understand dynamic defect formation and recovery at the temperature environment closed to the actual fusion reactor, hydrogen implantation at room temperature followed by thermal annealing was expected to evaluate the elementary steps of defect recovery and vacancy aggregation [13,14,22–24]. Therefore, post-implantation thermal annealing was performed, which provides a relatively fundamental description of hydrogen bubbles' behavior at higher temperatures.

2. Experiment details

2.1. Materials and sample preparation

In this study, the commercial tungsten (purity 99.95%, thickness 0.1 mm, as-rolled) was provided by Goodfellow Cambridge Limited, England. Disk samples with a diameter of 3.0 mm were punched out from the as-received plate and mechanically polished (milled) to a thickness around 60 μm. Then, these samples were annealed at 1000 °C for 2.0 h in high vacuum ($\sim 10^{-3} Pa$) to reduce the concentration of defects and relieve the stress introduced in the polishing process [18,25]. After that, these disk samples were electrochemically polished by a twin-jet electro-polisher (MTP-1A, 0.5 mol/L $NaOH$ at room temperature) to be thin enough (<100 nm [18,19]) to obtain well-defined surfaces, which could be transparent under TEM observation. The metallography of these samples was observed firstly using a scanning electron microscopy (SEM, MIRA3, Tascan). It indicated that tungsten grains possess irregular structures with grain sizes ranging from several to tens of micrometers (Fig. 1a). In addition, local crystallographic texture of W matrix was acquired using electron backscatter diffraction (EBSD). Measurements were carried out at a SEM (EVO 18, Carl Zeiss) equipped with an EBSD detector (Nordlys Max2, Oxford Instruments), and a texture with <100> direction preferentially oriented parallel to the sample surface was obtained (Fig. 1b). Furthermore, the microstructures of these samples prior of hydrogen implantation were given by TEM (JEM-2100, JEOL) observations (Fig. 1c–d).

2.2. Hydrogen implantation

Thinned TEM samples were separated into two groups to study the hydrogen bubbles behavior with total fluence (as group G_1) and beam flux (as group G_2), respectively. Hydrogen implantations were carried out in a low-energy high-flux ion source platform (Pearbody scientific) with custom built end-stations located at School of Nuclear Science and Technology, Lanzhou University, China. The duoplasmatron–type ion source was used to extract hydrogen ions with energies in the range of $5.0 \sim 20.0$ keV. A summary of the implantation conditions on the planar TEM samples is presented in Table 1. During implantation, the round-shaped samples were orientated perpendicular to the beam direction, in a high vacuum chamber ($\sim 10^{-4} Pa$). The temperature of the sample holder was controlled at room temperature (RT) by water cooling system. In order to get a higher beam intensity, hydrogen clusters (H_3^+), i.e., a cluster consists of three hydrogen atoms but lost one of its electrons [6,11,18], was used in hydrogen implantation. For the fluence dependence investigation, four samples ($S_1 \sim S_4$) in group G_1 were used. The implantation energy of H_3^+ clusters was chosen 18.0 keV at a fixed flux of $2.08 \times 10^{14}\ H_3^+ \cdot cm^{-2} \cdot s^{-1}$, corresponding to an implantation energy of 6.0 keV for each hydrogen with a hydrogen flux of $6.25 \times 10^{14}\ H \cdot cm^{-2} \cdot s^{-1}$. These four samples were subsequently implanted to fluences of 7.5×10^{17}, 1.5×10^{18}, 2.25×10^{18} and $3.38 \times 10^{18}\ H \cdot cm^{-2}$, respectively. For the beam flux dependence investigation, four hydrogen fluxes (3.125, 6.25, 9.375, and $12.5 \times 10^{14}\ H \cdot cm^{-2} \cdot s^{-1}$) were chosen to implant TEM samples ($S_5 \sim S_{18}$) in group G_2. The chosen implantation energy of hydrogens was 6.0 keV with a total fluence of $2.25 \times 10^{18}\ H \cdot cm^{-2}$. As shown in Table 1, three or four samples in group G_2 were implanted with the same beam flux to ensure repeatability.

The distributions of as-implanted hydrogens (in units of at. %) and displacement damage (in units of dpa) were calculated by SRIM-2013 [26], in which a displacement energy of 90 eV in W matrix was applied [11,17]. While displacement damage peaked at $\sim$ 10 nm, the typical concentration of hydrogens is ranged to $\sim$ 100 nm with the peak located at $\sim$ 30 nm (Fig. 2). Therefore, all of the hydrogens were stopped within the thinned TEM sample. The maximum damage level is approximately 0.4 dpa at the peak position at a fluence of $7.5 \times 10^{17}\ H \cdot cm^{-2}$. However, the diffusion and annihilate of defects was not taken into account in this calculation.

2.3. Microstructural characterization

Following ion implantation, over-focus and under-focus techniques in TEM observation were used to detect hydrogen bubbles. The fact that dark dots (in over-focused condition) turned to bright (in under-focused condition) at the same area indicating they are bubbles/voids [18,19]. The size distributions of gas bubbles in tungsten were measured using digital micrograph software from under-focused bright field TEM micrographs. The gas bubbles can be resolved when their sizes are larger than 0.5 nm. It is noticed that the size and density of bubbles varies at various irradiation conditions (Figs. 4 and 6). In order to increase statistics, at least three different areas have been chosen for each sample to measure