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Hydrogen bubble formation and evolution in tungsten under different hydrogen irradiation conditions

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HIGHLIGHTS

- Direct and clear observation of hydrogen bubbles evolution by TEM is provided.
- The role of temperature playing in bubble formation and evolution is fully explored.
- Vacancy trapping mechanism is verified in this experiment.

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ABSTRACT

In order to see how hydrogen is behaving in tungsten and to understand the way bubbles form and grow up, specimens were irradiated by hydrogen ions from room temperature to 800 °C to fluence of 2.25×10^{21} m^{-2}. Experimental results show that higher temperature helped bubble acquire higher internal pressure, causing interstitial loop punching to happen. In this process bubbles' size grew and dislocation loops were formed but dislocation loops migrated away at and above 350 °C. And bubble number density reached peak value at 600 °C but then dropped dramatically at 800 °C. Because continuously increasing temperature would cause small bubbles dissolution or leaking out. Besides, high temperature also prevented tiny bubbles growing to be visible under TEM observation by their reaching equilibrium pressure before reaching threshold pressure for loop punching. In the other set of experiments, specimens were irradiated by low hydrogen fluence of 1×10^{20} m^{-2} at 600 °C, in which case few hydrogen bubbles appeared. With further increasing irradiation fluence, bubble number density quickly increased. Small bubbles tended to coalesce to become larger visible bubbles. And they continued to grow through loop punching until their internal pressure cannot support their size expansion any more.

1. Introduction

In a fusion reactor tungsten is proposed to be primary candidate material for plasma facing material and especially used as the divertor upper baffle and dome material [1]. Interaction of tungsten with H, D and T fuel particles whose energies are between eVs and keVs [2] as well as tritium inventory in tungsten is unavoidable and remains difficult problems for fusion engineering and design [3]. There have been plenty of studies on hydrogen isotopes behavior in tungsten including diffusion, permeation and transport of H, D in material [4,5] and H, D, T trapping by defects and other irradiation damage [6–12] are supposed to lead to generation of hydrogen bubbles [13–15] whom are blamed for degrading the mechanical properties of the metal [16] and causing surface blistering. Some theories of bubble formation mechanism have been proposed through various computational simulation methods [10,13–15,17,18] such as plastic deformation [17], dislocation loop punching and agglomeration of hydrogen-vacancy complexes [14,18], unfortunately direct observation of hydrogen bubble evolution in tungsten is rare. As quantification of the effect of ion-induced damage on tritium trapping, permeation, and retention in tungsten are recommended for high-priority research and development [3], this paper provides a relatively comprehensive

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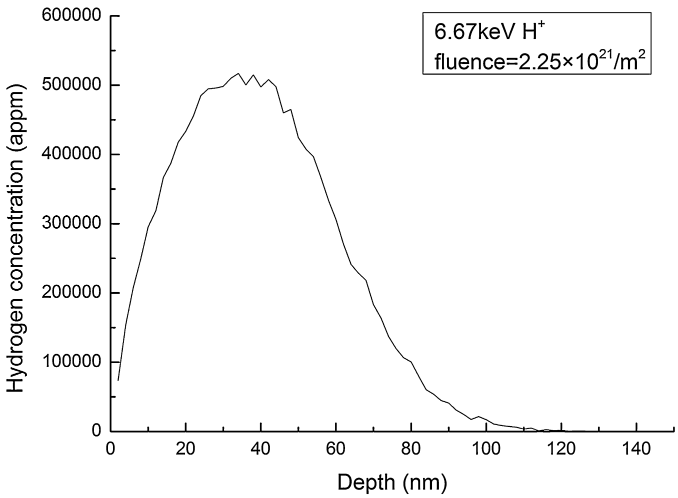

Fig. 1. SRIM calculation of 6.67 keV H^+ ion range distribution in tungsten.

description of hydrogen bubble formation in tungsten during hydrogen ion irradiation and bubble evolution with changing irradiation conditions by means of transmission electron microscope (TEM) observation.

2. Experimental procedure

Polycrystal tungsten with high purity of 99.97% was used in experiment. After annealed in vacuum at 1200 °C for 2 h, disk specimens of 3 mm diameter were punched out from flakes with thickness of 0.06 mm and were further mechanically polished (milled) to a thickness of around 30 μm. And these milled specimens were then electro-chemically polished by using a 1 wt.% NaOH polishing solution in a twin-jet electro-polisher to become thin enough and obtain well-defined surfaces to be able to be observed under a TEM in order to see their microstructure after irradiation.

TEM specimens were separated into two sets. W1, W2, W3, W4, W5 were put in set-one; W6, W7, W8 were put in set-two. Then specimens W1, W2, W3, W4, W5 were irradiated by 6.67 keV H^+ (20 keV $H_3{}^+$) beam at room temperature (RT), 350 °C, 500 °C, 600 °C, 800 °C to same fluence of 2.25×10^{21} m^{-2} (the flux was around 7.74×10^{16} m^{-2} s^{-1}); W6, W7, W8 were irradiated by 6.67 keV H^+ beam to different fluence of 1×10^{20} m^{-2}, 7.5×10^{20} m^{-2}, 1.5×10^{21} m^{-2} at 600 °C which can be compared with W4. Temperature was monitored by a thermocouple throughout the experiment where specimen was mounted on top of a heater by using fulmargin with the probe of the thermocouple touching the ion incident plane of specimen. Detailed irradiation conditions are listed in Table 1. The ion irradiation experiments were carried out on an ion implanter in the Accelerator Lab of Wuhan University.

From the calculation of SRIM2008 using a displacement energy of 90 eV [19] the projected depth of 6.67 keV H^+ in tungsten was within the range of 100 nm as shown in Fig. 1. After irradiation, these specimens were studied by a JEM-2010HT TEM which was equipped with the 200 kV accelerator voltage. The depth range of TEM observation among all specimens measured from the surface was fixed at ~100 nm which was estimated by counting the number of thickness fringes from the edge of the thin foil (the thickness fringes method).

3. Results and discussion

3.1. Hydrogen bubble evolution in tungsten with increasing temperature

3.1.1. Microstructure change in tungsten after hydrogen irradiation at RT

In Fig. 2, compared with the unirradiated specimen, many small dislocation loops with size ranging from 2 to 6 nm appeared in specimen W1 which was irradiated by 2.25×10^{21} m^{-2} hydrogen at room temperature. But no bubble could be found at such low temperature.

It is mentioned in Ref. [20] that the threshold energy for hydrogen ions to produce displacement damage in tungsten is around 2 keV. After 6.67 keV hydrogen ion irradiation, large amount of Frenkel pairs were introduced and dislocation loops nucleated and grew through interstitial atoms aggregation [7]. In this process the binding energy of two self-interstitial-atoms (SIA)s was as strong as 2.12 eV [8] and two SIAs could form a di-interstitial: $I + I = I_2$ which became the nucleus of an interstitial loop [21]. But the mobility for hydrogen atoms were very low at room temperature whose diffusion coefficient was $D_H = 4.1 \times 10^{-7} \exp(-0.39\,\text{eV/kT})\,\text{m}^2\,\text{s}^{-1}$ [22], and vacancies were only able to move when temperature were above 200 °C [23] due to their high migration energy of 1.7 eV [24] that is much higher than the migration energy of SIAs which is 0.054 eV [25]. Therefore hydrogen atoms could not move close to vacancies and hydrogen bubble nucleation through vacancy trapping mechanism was suppressed considering the fact that the trapping radius for a monovacancy to trap a deuterium atom is 0.59 nm [9].

3.1.2. Hydrogen bubble formation at 350–500 °C

In Fig. 2 when temperature increased to 350 °C, a few tiny (in fact they are so small that only when compared with the over-focused TEM picture of Fig. 3 can we recognize them) bubbles could be found in specimen W2. When irradiated at 500 °C numbers of bubbles whose average diameter was 2 nm with bubble number density 1.4×10^{23} m^{-2} appeared in specimen W3. But dislocation loops disappeared with increasing temperature. As for bubble number density, it was calculated by n/V, where n was number of bubbles and V is the total volume of the areas we selected to count bubbles and the areas' thickness we adapted was 100 nm.

After temperature increased to 350 °C vacancies started to move, and hydrogen atoms' mobility was enhanced, at the same time dislocation loops coalesced with each other and grew and escaped to the surface [20]. It is said that the number of irradiated-induced vacancy and SIA pair is 0.16 per 5 keV deuterium atom and 0.79 per 15 keV deuterium atom in tungsten [9]. So in this experiment, for about every six implanted 6.67 keV hydrogen atoms, one vacancy might be produced. Taking into account the escape of hydrogen near the surface, the rest hydrogen atoms were left behind for hydrogen bubble nucleation. Compared with SIA whose binding energy with a hydrogen atom is 0.33 eV, a vacancy has a much larger binding energy with a hydrogen atom which is 1.22 eV [26] that is also much stronger than intrinsic traps including impurities, grain boundaries and dislocations, therefore most of hydrogen was trapped by vacancies [9]. In this process, the first several hydrogen atoms would diffuse into the vacancy and occupy the internal surface of the vacancy until the internal surface was saturated—all the tetrahedral sites on each cube face around a vacancy were occupied [27]. As a result, the later incoming hydrogen atoms would have to enter and locate at the center of the vacancy, which leading to formation of H_2 molecules at the vacancy center. Following this hydrogen bubble nucleation started [10]. Further increasing