Current mapping of low-energy (120 eV) helium and hydrogen irradiated tungsten by conductive atomic force microscopy

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Abstract

Both conductive atomic force microscopy (CAFM) and transmission electron microscopy have been used to characterize the defects or He bubbles in low-energy (120 eV) H and He irradiated tungsten (W). By a comparative study, we find that the current mapping from CAFM is very sensitive in the detection of nanometer-sized defects in low-energy H and He irradiated W. Our calculation confirms that the resistance change in H and He irradiated W is strongly affected by the distance between atomic force microscopy tip and defects/He bubbles. CAFM can accurately detect defects/He bubbles in the W surface layer, however, it is infeasible to measure them in the deep layer (>20 nm), especially due to the existence of defects in the surface layer.

1. Introduction

Low-energy (100-500 eV) and large-flux helium (He) and hydrogen (H) irradiations from deuterium-tritium (D-T) thermonuclear fusion cause serious surface damages in tungsten (W) as one of the most important candidates for plasma-facing materials in ITER [1-3]. Traditionally, high-resolution transmission electron microscopy (TEM) has been utilized to evaluate the change in W surface topography, such as the formation of bubbles, blisters, holes, and fiber-form nanostructures [4,5]. However, the specimen preparation for TEM analysis is rather complicated, which typically includes polishing, coating deposition, and ion milling process. Specimens are seriously destroyed during the specimen preparation, and the microstructures to be observed might be changed. Electron irradiations in TEM system can also cause a change in the measured materials and accelerate their irradiation damage [6,7].

Previously, conductive atomic force microscopy (CAFM) has been successfully utilized to detect the nanometers-sized defects or bubbles in W crystals irradiated with H and He ions [8-10]. Briefly, CAFM is using a PtIr-coated tip to perform the current measurements in the contact mode. A constant sample voltage is applied between the conductive tip and the irradiated sample, and electron emission images through the irradiated samples are obtained simultaneously with topography. So, during the tip scanning, the surface morphology and the current image are obtained at the same time. Compared to TEM, CAFM measurements can have several advantages, as shown

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below: (1) CAFM measurement does not make any damage to the specimens measured; (2) CAFM measurement may provide a direct comparison between the surface topography and the conductivity or defect distribution of measured specimens; (3) CAFM current mapping is quite sensitive to a change in electron conductivity of measured specimens, and it may identify the defects in nanoscale or atomic resolution; (4) The conductivity of defects may be measured, which can be very helpful for understanding the compositions and growth process of defects. However, CAFM measurement can be only sensitive in detecting defects in the surface layer of specimen. During CAFM measurements, tip scans are carried out along a surface (x or y direction), and the effect of defect locations along the vertical (z) direction on current mapping still remains unclear.

In this paper we report CAFM and TEM measurements of low-energy (120 eV) H and He irradiated polycrystalline W. The size and density of defects in W are obtained, and the effect of defect locations on current mapping is analyzed. We show theoretically that the size and density of defects from the current mapping in CAFM are strongly dependent on the defect location along Z direction. Our simulation indicates that the current mapping in the CAFM measurements can be very effective in detecting nanometer-sized defects or He bubbles in the surface layer of irradiated W.

2. Experimental procedures

The H and He irradiations of polycrystalline W were performed at a temperature of 873 K by using our MIES described previously [10]. H irradiations were performed with a plasma beam of H^+ , H_2^+ and H_3^+ ions while He irradiations were carried out with He^+ beam. All H and He irradiations were performed at the energy of 120 eV and the fluence of 1.0×10^{25} ions/m². Prior to H and He irradiations, W specimens were annealed at 1273 K for 2 h in vacuum to reduce the large concentration of nanometer-sized defects. CAFM (Veeco DI 3100) was used to detect the nanometer-sized defects in irradiated W [8-10]. CAFM measurements were performed at a typical scan rate of 0.3 Hz by using a PrIr-coated tip in contact with the specimen. The surface topography of irradiated W was obtained with the current mapping at a constant voltage of 20 mV. FIB milling was utilized to process the samples for TEM analysis. Before focused ions beam (FIB) milling, a W coating with a thickness of 10 nm was deposited to protect the measured specimen. Then, thin samples with 50 nm in thickness were prepared, and JEM-2010 and H-9000UHR microscopes were used for TEM observation. However, the microstructures of specimens are usually changed or destroyed by the complex operation of FIB milling.

3. Results and discussion

Figure 1(a) and 1(b) shows the surface topography and current mapping of polycrystalline W irradiated with He plasma, respectively. The wave-like surface microstructures are orientated in a certain direction [11]. Plenty of nanometer-sized defects can be observed from the current mapping. These defects are relatively insulating while their edges are more conductive, indicating that nanometer-sized He

bubbles are formed in the irradiated W. The characteristic shape of defects and their distribution is consistent with the measured surface topography. The growth of He bubbles, resulting in surface swelling is related to He diffusion and accumulation in the W surface layer. The formation of wave-like microstructures can be due to the cascading slipping of W atoms along dense-packed faces during the growth of He bubbles [8]. Prior to irradiation, W samples were also measured by CAFM. Current image showed that the distribution of conductive defects was irregular, and their density was much lower. The depth at which irradiated defects are retained may be typically divided into two layers: (i) the surface layer (<10-20 nm), and (ii) the deep layer (>20 nm). The TEM image in Fig. 1(c) shows that large He bubbles with sizes of 5-10 nm are formed in the W surface layer while smaller bubbles with sizes of <2-3 nm are observed in the deep layer. This indicates the strong He accumulation in the surface layer of W. The formation of large-sized He bubbles results in the surface swelling of irradiated W lattice. Figure 1(d) shows the size and density of He bubbles measured by CAFM and TEM. There are serious differences about the data obtained from these two methods. When the size of He bubbles is >10 nm, the size and density measured by CAFM are consistent with the ones measured by TEM. This indicates that CAFM may accurately detect large He bubbles in the surface layer. However, small He bubbles in the deep layer are invisible for CAFM measurements.

Surface topography and current mapping of low-energy H irradiated W are shown in Fig. 2(a) and 2(b), respectively. The current mapping in Fig. 2(b) contains plenty of defects with their typical sizes of >20-30 nm, and these defects are relatively conductive. Our previous study showed that at an increasing H fluence small defects may diffuse and coalesce into large defects in W [10]. The size and density of defects are strongly dependent on H fluence, and ordered arrangements of defects are formed at relatively high H energy and irradiation temperature. Our recent study shows that due to low-energy H irradiation at 373 K, the W surface layer contains a high density of nanometer-sized defects, resulting in the thermal instability of W atoms in the surface layer and a periodic surface degeneration of the W lattice [12]. The surface topography of H irradiated W in Fig. 2(a) is consistent with its current mapping in Fig. 2(b). The formation of nanometer-sized defects in the W surface layer can result in surface swelling and surface degeneration of W lattice. The TEM image in Fig. 2(c) shows that the surface layer destroyed due to surface swelling is about 8-10 nm thick. Plenty of small defects are formed inside the W lattice, and the distance between these defects and the W surface is in the range of 10-30 nm. Compared to He atoms, H atoms diffuse more deeply into W lattice at an elevated temperature, as observed from TEM images in Figs. 1(c) and 2(c). The size and density of the defects measured by CAFM and TEM are shown in Fig. 3(d). The size of defects measured by CAFM is much larger than the one measured by TEM while their density from CAFM measurement is lower than the one measured by TEM. This indicates that CAFM measurement is just sensitive in detecting the defects inside the surface layer. TEM measurement is inaccurate in detecting the defects in the surface layer. The small H defects in the deep W layer are not detectable by CAFM.

To analyze the effect of defect location in the W lattice on current mapping in

CAFM measurements, we calculate the resistance (R_T) between AFM tip and sample holder when W with defects is scanned by the AFM tip. The three-dimensional structure of the resistance is schematically shown in Fig. 3, where the electrons emitted from AFM tip flow through W containing one defect. The AFM tip with its radius of r_T is scanned along x or y axis over the W surface. It is proposed that the conical structure with its apex angle of 2θ is formed between the center of the nanotip and the defect in W. The distance between the center of the nanotip and the defect in W is L_D , and the radius of the defect is r_D . When considering that electrons emit from the AFM tip through W in a hemispherical shape, R_T can be calculated as

$$R_T = \frac{1}{\frac{1}{R_c} + \frac{1}{R_r}} \quad (1)$$

where R_c and R_r are the resistance of conical structure and the one of rest part in the hemispherical structure, respectively. R_r may be expressed as

$$R_r = \frac{1}{2\pi r_T \cos \theta} \quad (2)$$

where $\theta = \arctan \frac{r_D}{L_D}$. If no defect exists, the resistance (R_0) of the hemispherical structure can be expressed [6]

$$R_0 = \int_{r_T}^{\infty} \frac{\rho_0 dr}{2\pi r^2} = \frac{\rho_0}{2\pi r_T} \quad (3)$$

where ρ_0 is the resistivity of W. R_c can be calculated as

$$\begin{aligned} R_c &= \int_{r_T}^{L_D - r_D} \frac{\rho_0 dr}{2\pi(1 - \cos \theta)r^2} + \int_{L_D - r_D}^{L_D + r_D} \frac{\rho_D dr}{2\pi(1 - \cos \theta)r^2} + \int_{L_D + r_D}^{\infty} \frac{\rho_0 dr}{2\pi(1 - \cos \theta)r^2} \\ &= \frac{1}{2\pi(1 - \cos \theta)} \left(\frac{\rho_0}{r_T} - \frac{\rho_0}{L_D - r_D} + \frac{\rho_D}{L_D - r_D} - \frac{\rho_D}{L_D + r_D} + \frac{\rho_0}{L_D + r_D} \right) \end{aligned} \quad (4)$$

where ρ_0 and ρ_D are the resistivity of W and the defect, respectively. Combining Eqs. (1), (2), and (4), one obtains

$$R_T = \frac{\rho_0 (\rho_0 (L_D^2 - 2r_D r_T - r_D^2) + 2\rho_D r_D r_T)}{2\pi r_T (\rho_0 (L_D^2 - r_D^2 - 2r_D r_T \cos \theta) + 2\rho_D r_D r_T \cos \theta)} \quad (5)$$

By combining Eqs. (3) and (5), the change ($\Delta R(l, z) / R_0$) in resistance can be obtained

$$\frac{\Delta R(l, z)}{R_0} = \frac{R_T - R_0}{R_0} = \frac{2R_T (\rho_D - \rho_0) (1 - \cos \theta) r_D}{(\rho_0 (l^2 + z^2 - 2r_D r_T \cos \theta - r_D^2) + 2\rho_D r_D r_T \cos \theta)} \quad (6)$$

where $l = \sqrt{x^2 + y^2}$. z is the vertical distance between W surface and the defect. If $L_D \gg r_D$ and $\rho_0 \gg \rho_D$, Eq. 3 can be approximated by

$$\frac{\Delta R(l, z)}{R_0} \approx -\frac{r_T r_D^3}{(l^2 + z^2)^2} \quad (7)$$

This indicates that $\Delta R(l, z)/R_0$ values are not dependent on ρ_0 and ρ_D , but they are mainly determined by r_D , l and z . Fig.4(a) shows the variation of $\Delta R(l, z)/R_0$ as a function of l for different values of z , assuming $r_D = 2nm$. r_T was assumed to be approximately 1 nm [13,14]. The circular and non-conductive defects can be obtained from CAFM measurements. The $\Delta R(l, z)/R_0$ value is strongly dependent on z value. The maximum $\Delta R(l, z)/R_0$ value at $z = 2nm$ is almost 6 orders of magnitude higher than the one at $z = 50nm$. The maximum $\Delta R(l, z)/R_0$ value greatly decreases by 3 orders of magnitude when z increases from 2 to 10 nm. Thus, CAFM is very sensitive in detecting conductive defects with a depth of <5-10 nm. It is impossible to detect defects in the deeper layer (>20nm) due to the existence of defects in the surface layer. This indicates that the conductive defects in H-irradiated W are from the W surface layer. If the size of the defects detected from the current mapping is proposed to be full width at half maximum (FWHM) of $\Delta R(l, z)/R_0$ values, its radius significantly increases with the z value (Fig. 4(b)). Our calculations show that the maximum $\Delta R(l, z)/R_0$ value increases by 2 orders of magnitude when r_D varies from 2 to 10 nm, indicating that CAFM is only sensitive in detecting larger-sized defects in W.

If $L_D > r_D$ and $\rho_D \gg \rho_0$, Eq. 3 can be written as

$$\frac{\Delta R(l, z)}{R_0} \approx \frac{r_D^2}{2(l^2 + z^2)} \quad (8)$$

The $\Delta R(l, z)/R_0$ values at $r_D = 2nm$ and $z = 2-50nm$ are plotted as a function of l , as shown in Fig. 5(a). The maximum $\Delta R(l, z)/R_0$ value at $z = 2nm$ is about 3 orders of magnitude lower than the one at $z = 50nm$. Figure 5(b) shows that the

maximum $\Delta R(l, z)/R_0$ value is strongly dependent on z value, and CAFM is very sensitive in detecting large-sized defects in surface layer. Our CAFM measurements confirm that the large-sized and non-conductive He bubbles in the surface layer are detectable while the small He bubbles in the deep layer are invisible from the current mapping of He irradiated W. Increasing z value from 2 to 50 nm leads to an obvious increase in the radius of detected defects.

4. Conclusions

In summary, by a direct comparison between CAFM and TEM methods, we have demonstrated that CAFM measurement is rather simple, and it does not make any damage to H and He irradiated W. The current mapping from CAFM measurement is very sensitive in the detection of nanometer-sized defects in low-energy H and He irradiated W, which can play a crucial role in understanding the surface degeneration of H and He irradiated W. However, the defects or He bubbles in the deep layer (>20 nm) are not detectable by CAFM. Our calculation confirms that during AFM tip scan, the resistance change in the irradiated W is strongly dependent on the distance between tip and defects/He bubbles. The resistance change increases by 3-6 orders of magnitude when the distance varies from 2 to 50 nm.

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Figure 1

FIG. 1. (a) Surface topography and (b) current mapping of the polycrystalline W irradiated with He plasma. (c) TEM image of the polycrystalline W irradiated with He plasmas. (d) Size and density distributions of He bubbles in W, measured by CAFM and TEM.

Figure 2

FIG. 2. (a) Surface topography and (b) current mapping of the polycrystalline W irradiated with H plasma. (c) TEM image of the polycrystalline W irradiated with H plasma. (d) Size and density distributions of H defects in W measured by CAFM and TEM.

Figure 3

FIG. 3. Schematic diagram of the resistance between AFM tip and sample holder, where the electrons emitted from AFM tip flow through W containing one defect. The AFM tip with its radius of r_t is scanned along x or y axis over W surface. The conical structure with its apex angle of 2θ is formed between the center of the nanotip and the defect in W. The distance between the center of the nanotip and the defect in W is R_D , the horizontal distance between the center of the nanotip and the defect is l , the vertical distance between the center of the nanotip and the defect is z , and the radius of the defect is r_D .

Figure 4

FIG. 4. (a) $\Delta R(l, z)/R_0$ values obtained at $r_D = 2\text{nm}$ as a function of l . (b) The variations of $\Delta R(l, z)/R_0$ values and defect radius measured with the z value. In this approximation, $L_D \gg r_D$ and $\rho_0 \gg \rho_D$. r_T is assumed to be 1 nm.

Figure 5

FIG. 5. (a) $\Delta R(l, z)/R_0$ values obtained at $r_D = 2\text{nm}$ as a function of l . (b) The variations of $\Delta R(l, z)/R_0$ values and defect radius measured with the z value. In this approximation, $L_D > r_D$ and $\rho_D \gg \rho_0$.