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Blistering on tungsten surface exposed to high flux deuterium plasma

H.Y. Xu [a, b, *], W. Liu [b, **], G.N. Luo [c], Y. Yuan [d], Y.Z. Jia [b], B.Q. Fu [b], G. De Temmerman [e, f]

[a] Center of Interface Dynamics for Sustainability, Institute of Materials, CAEP, Chengdu 610200, China
[b] Key Laboratory of Advanced Materials, School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China
[c] Institute of Plasma Physics, Chinese Academy of Sciences, Hefei, Anhui 230031, China
[d] School of Physics and Nuclear Energy Engineering, Beihang University, Beijing 100191, China
[e] FOM Institute DIFFER-Dutch Institute for Fundamental Energy Research, Trilateral Euregio Cluster, Edisonbaan 14, 3439 MN Nieuwegein, The Netherlands
[f] ITER Organization, Route de Vinon-sur-Verdon CS 90046-13067, St Paul Lez Durance Cedex, France

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ABSTRACT

The blistering behaviour of tungsten surfaces exposed to very high fluxes ($1–2 \times 10^{24}/m^2/s$) of low energy (38 eV) deuterium plasmas was investigated as a function of ion fluence ($0.2–7 \times 10^{26}$ D/m^2) and surface temperature (423–873 K). Blisters were observed under all conditions, especially up to temperatures of 873 K. The blister parameters are evaluated with blister size, blister density and surface coverage. The blister size always peaked at less than 0.5 μm and no blister larger than 10 μm is observed even at high fluence. The blister densities are found in high magnitude of 10^6 blisters/m^2, with the surface coverages lower than 2%. The formation of cracks in the sub-surface region was observed by cross-section imaging. Changes in blister size and shape with fluence and temperature suggest processes of predominantly nucleation and subsequent growth of blisters. The smaller blister size is considered to be caused by a combination of flux-related effects such as enhanced defect formation in the near surface region, reduced deuterium diffusivity and relatively short exposure times.

1. Introduction

Tungsten has recently been chosen as the divertor material from the start of ITER operations, and is also a leading candidate as plasma-facing material in future fusion reactors due to its high melting point, high thermal conductivity, low sputtering erosion and low H isotope retention [1,2]. Plasma facing components (PFCs) in the divertor region of a fusion device have to be able to withstand large power densities (up to 10 MW/m^2) and intense D/T/He particle fluxes (10^{22}–$10^{24}/m^2/s$) [3]. Bombardment of surfaces by such fluxes of energetic particles can lead to significant surface modification [4,5], which could lead to a deterioration of the material thermo-mechanical properties [5,6] and to an enhanced uptake of tritium [7,8], which is important for the long-term D-T operation.

Blister formation has been widely studied for tungsten exposed to deuterium ion/plasma bombardment both in linear plasma devices and in tokamaks [5,8–24]. Discussion on the precise mechanism leading to blistering is ongoing [10–13], and is believed to be related either with plastic deformation caused by deuterium super-saturation and the accompanying stress, or with elastic or plastic deformation caused by gas pressure inside cavities [14]. Overall, all the basic issues are correlated to trapping and diffusion of deuterium. After implantation, deuterium diffuses far beyond the implantation range and is trapped by defects, such as vacancy/vacancy clusters, dislocations and grain boundaries or other lattice imperfections. The incident ion fluence (i.e. total number of particles impinging per unit surface area) and surface temperature are important factors affecting deuterium retention and blistering. Typically, blisters with diameters between 1 and up to 300 μm are reported on tungsten surfaces with elliptic outlines or rounded domes, depending on the exact experimental conditions [15–18]. Both the blister density and size are reported to grow with the incident fluence by overlapping and/or coalescence of neighbouring blisters [13]. For increasing temperatures, the blister size increases while the blister density and surface coverage decreases [13]. Suppression of blister formation has been reported at temperatures above ~700 K by several researchers [10–13,15] except at 900 K by Ye et al. [9]. As a possible reason it was proposed that a

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* Corresponding author. Center of Interface Dynamics for Sustainability, Institute of Materials, CAEP, Chengdu 610200, China.
** Corresponding author.
E-mail addresses: donaxu@163.com (H.Y. Xu), liuw@mail.tsinghua.edu.cn (W. Liu).

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high diffusivity of deuterium at high temperatures prevents local accumulation of deuterium leading to a too low concentration of deuterium to initiate blistering [13].

While blister formation has been extensively studied for plasma fluxes in the range 10^{19}–10^{22}/m^2/s, this is far below the expected peak ion flux in the ITER divertor (~10^{24}/m^2/s during the D-T operations) [25]. Recently, the appearance of dense blisters with size up to a few hundred nanometres in diameter was reported by several researchers [19–21]. The changing of blistering behaviour with increased temperature is different from the case with flux less than 10^{22}/m^2/s [21]. And a new type of surface modifications-nanostructures, associated with the enhanced generation of defects in the implanted zone [20,22,26,27], has been reported for tungsten exposed to high flux D plasma. It is thus of interest to investigate how these high flux conditions affect the formation of blisters and how the blistering occurs with fluence and surface temperature.

2. Experimental

The material used in the present study is high purity (>99.95 wt %) rolled tungsten, which was supplied by Advanced Technology & Materials CO. Ltd. (China). It is produced by powder sintering followed by warmly cross-rolling (~1473 K) to 75% reduction in thickness (final thickness ~3 mm) which results in a homogeneous microstructure. The main impurities were Mo, Fe, O below 15 ppm, and C around 50 ppm. Disc specimens with a diameter of 30 mm and a thickness of 3 mm were polished mechanically and electrochemically to mirror-like surfaces and then stress-relieved at 1273 K for 1 h at a background pressure of 5×10^{-4} Pa.

D plasma exposures were carried out in the Pilot-PSI linear plasma generator, which is uniquely capable of producing plasmas with a deuterium ion flux in the range 10^{23}–10^{25}/m^2/s and with energy fluxes up to 50 MW/m^2 [28]. The composition of D plasmas was safely assumed to be dominated by D^+ ions and the neutral species in front of the target were neglectable by 't Hoen [29]. The plasma beam with a Gaussian profile are obtained, with the electron density (~0.6–2×10^{20}/m^3) and temperature (<2 eV) determined by Thomson scattering ~17 mm away from the plasma-exposed surface. The peak ion flux of ~ 1–2×10^{24}/m^2/s was determined from the measured electron density and electron temperature in the centre. The ion energy was fixed to ~38 eV by negatively biasing the samples. A fast infrared camera (FLIR SC7500-MB) was used to monitor the surface temperature of the sample, and was cross-calibrated with a spectral pyrometer during the experiment. To reach the required fluence for each sample, several consecutive shots with duration of 10 s and/or 75 s were carried out, with efficient heating-up and cooling down within ~1–2 s. Different surface temperatures were obtained by inserting different layers of grafoil between the samples and the water-cooled sample holder.

The surface morphology of the tungsten samples was observed by a scanning electron microscope (Tescan XM 5136-SEM), both before and after D plasma exposure. The sub-surface morphology of the samples was analysed with a field emission SEM after cross-sectioning by a focused ion beam (TESCAN LYRA3 FEG-SEM/FIB, 30 kV Ga^+).

3. Results

3.1. Blisters on W surface

Fig. 1 shows typical SEM pictures from a sample exposed in Pilot-PSI to a plasma fluence of 7×10^{26} D/m^2 at a surface temperature of 423 K and illustrates the formation of blisters with irregular shapes. Compared to Fig. 1 (a), blisters are formed markedly after D plasma exposure, as shown in Fig. 1 (b). Basically, blisters can be classified into two categories depending on their height-to-diameter ratio: most blisters exhibit a flat shape extending within the surface while some blisters appear with relatively higher domes. Examples of those two categories are highlighted in Fig. 1 (b) by arrows. The flat blisters (indicated by the red arrows) appear like overlaps and/or coalescence of small blisters, while terraces can be found on the top surface. Analyses of images taken with a 45° tilt angle, the maximum ratio of blister height to diameter is found to range from about 0.02 to 0.2 which is much lower than the value of 0.7 reported by Shu et al. [12]. Compared with the flat blisters, the blisters with high dome (pointed to by white arrow) usually appear with smooth surfaces and shapes close to round or elliptic, with a relatively constant height-to-diameter of about 0.2.

The distribution of blister size is evaluated from five images taken from the exposed area of the surface (Fig. 1 (c)). The blisters are sketched from the top-viewed images while the size is determined by the equivalent circle diameter from the sketched areas. The number of blisters used to determine the size distribution is shown in the right axes of Fig. 1 (c), with the size width of 0.1 μm is used throughout the paper. The blister size peaks at 0.3–0.4 μm, as noted by the most popular diameter d_{mp}, with the average size $d_{average}$, counted by arithmetic mean, at ~0.46 μm and the maximum diameter is found to be less than 3.2 μm.

3.2. Sub-surface morphology corresponding to blisters

To obtain more details on the materials' sub-surface modifications induced by blistering, the cross-section of the sample has been examined by SEM after FIB milling; the result is shown in Fig. 2. Cracks/extended voids are observed below the top surface. The intra-granular cracks observed below the surface of flat-dome blisters in Fig. 2 (a), appear parallel to the surface and are found at a depth less than 200 nm. For the larger blisters, multiple cavities within the projected area are observed, as shown in Fig. 2 (b): a cavity is observed along the grain boundary, extending much deeper into the bulk, and another intra-granular crack parallel to the surface at shallow depth. It should be mentioned that those cracks/cavities are not observed on the material before plasma exposure. Similar cracks/extended voids were observed by S. Lindig et al. [10] and Y.Z. Jia [21]. However, much deeper distribution (more than several μm) was found under relatively low flux (10^{22} D/m^2/s) in Ref. [10].

3.3. Effect of fluence on blistering behaviour

The influence of the plasma fluence on the blistering behaviour was examined for fluences in the range 0.2×10^{26} D/m^2 to 7×10^{26} D/m^2, while the other conditions were held constant. Fig. 3 shows the corresponding top-viewed SEM images. Blisters with irregular shapes are already showing up at the lowest fluence of 0.2×10^{26} D/m^2 (corresponding to an exposure time of 10 s). With the fluence increase, larger blisters appear, as shown in Fig. 3 (b)-(d). An orientation dependence of blistering is found on the exposed surface, showing that blistering occurs preferentially on some grains while some others are relatively immune to blistering. Regarding to [22], it is assumed that these grains have surface orientations near (111) and (100), correspondingly.

The blister size distribution was analysed from top-viewed images and is shown as a function of ion fluence in Fig. 4. Several images (4–10) as shown in Fig. 3 were analysed for each size distribution. A. bimodal distribution is observed on all samples, with a dominant peak below 0.5 μm, and another one around 1–2 μm,