Influence of particle flux density and temperature on surface modifications of tungsten and deuterium retention

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ABSTRACT

Systematic study of deuterium irradiation effects on tungsten was done under ITER – relevant high particle flux density, scanning a broad surface temperature range. Polycrystalline ITER – like grade tungsten samples were exposed in linear plasma devices to two different ranges of deuterium ion flux densities (high: $3.5–7 \cdot 10^{23}$ D^+/m^2 s and low: $9 \cdot 10^{21}$ D^+/m^2 s). Particle fluence and ion energy, respectively 10^{26} D^+/m^2 and $\sim$38 eV were kept constant in all cases.

The experiments were performed at three different surface temperatures 530 K, 630 K and 870 K. Experimental results concerning the deuterium retention and surface modifications of low flux exposure confirmed previous investigations. At temperatures 530 K and 630 K, deuterium retention was higher at lower flux density due to the longer exposure time (steady state plasma operation) and a consequently deeper diffusion range. At 870 K, deuterium retention was found to be higher at high flux density according to the thermal desorption spectroscopy (TDS) measurements. While blisters were completely absent at low flux density, small blisters of about 40–50 nm were formed at high flux density exposure. At the given conditions, a relation between deuterium retention and blister formation has been found which has to be considered in addition to deuterium trapping in defects populated by diffusion.

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1. Introduction

Tungsten is currently considered as the main plasma-facing candidate material for future fusion devices due to its high melting point, good thermal properties and low erosion yield and fuel retention. However, bombardment of tungsten surfaces by high fluences of low energy particles can lead to microstructural changes of a thin surface layer, such as erosion, dust formation, and undesirable fuel accumulation [1]. Experiments have shown that tungsten is also prone to blistering under deuterium irradiation. Previous investigations performed under particle fluxes of 10^{21}–10^{22} D^+/m^2 s demonstrated a dependence of the blister formation on surface temperature [2], incoming particle mix, tungsten surface properties and particle fluence [3–7]. The most favorable exposure temperature for blister formation is around 500 K. Roughening of tungsten surface and exposure temperatures above $\sim$950 K suppress the blister formation. However, when

referring to blistering in tungsten and its specific formation conditions one also has to take into account the production history of the material, its properties, and preparation process [7].

Plasma facing materials in ITER will have to perform under the impact of particle flux densities that vary by several orders of magnitude (10^{19}–10^{24} $m^{-2}s^{-1}$), surface temperature (370–1370 K) and ion energy (0.1–100 eV) [8]. In the present study, the role of particle flux density on plasma-material interaction has been investigated by exposing polycrystalline tungsten samples to deuterium plasmas, at a given fluence (10^{26} D^+/m^2), ion energy ($\sim$38 eV) and two different deuterium ion flux densities ($\sim$$10^{22}$ to $\sim$$10^{23}$ D^+/m^2 s). The data on tungsten exposure to high surface temperature and particle flux density were scarce therefore this study presents a broad and systematic investigation of tungsten under the expected ITER divertor conditions [9].

2. Experimental set-up

The tungsten grade used in the experiments was chosen according to the specifications approved for ITER material selection and

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was provided by Negele Hartmetall [10]. Tungsten of a purity of 99.94% was rolled in rods of 20 mm in diameter with grains elongated parallel to the rolling direction to insure a better heat transport and prevent delamination [1]. Circular samples of 14 mm diameter and 4 mm thickness were cut from the tungsten rods in a perpendicular direction to the grain elongation and polished mechanically to a mirror finish. Before exposure, samples were annealed at 1270 K for one hour and cleaned in an ultrasonic bath in acetone for 15 min at 30 °C. Tungsten grain size on the surface was ~40 μm as determined by electron backscatter diffraction analysis (EBSD).

The first set of experiments was done at the linear plasma device Magnum-PSI [11] under ion flux densities expected in the ITER divertor. A deuterium fluence of 10^{26} D^+/m^2 was accumulated after 9–12 plasma shots of 11.9–26.5 s where 10–20 min was allowed between the shots for the magnets to cool down. Flux densities were $3.5 \cdot 10^{23}$ D^+/m^2 s, $4.6 \cdot 10^{23}$ D^+/m^2 s and $7 \cdot 10^{23}$ D^+/m^2 s at the surface temperatures of 530 K, 630 K and 870 K respectively. The shot-to-shot reproducibility of the surface temperature was within a range of ±50 K. Ion energy of about 38 eV was reached by applying a negative potential on the samples. They were actively cooled during the plasma operation and the temperature was varied only due to the heating by the plasma.

The second set of experiments was performed at lower particle flux density at the PSI-2 linear plasma device which operates at steady state [12]. Samples were exposed at the ion flux density of $9 \cdot 10^{21}$ D^+/m^2 s reaching the same total fluence of 10^{26} D^+/m^2, ion energy ~38 eV and surface temperatures as in the first set of experiments. Temperature variation was achieved by changing the area of contact between the sample and the actively cooled sample holder (using different sizes of grafoil inter-layers). A quadrupole mass spectrometer was used to monitor the level of impurities in the plasma. The surface temperature was measured by infrared camera and calibrated with a thermocouple.

Scanning electron microscopy (SEM) was used to investigate the surface modifications and cross section images were taken after cutting the sample by means of a focused ion beam (FIB). The total deuterium retention was determined by thermal desorption spectroscopy (TDS) method by heating the samples linearly (approximately) up to 1273 K for one hour (0.35 K/s) in an ultravacuum chamber. During the heating up process it was possible to desorb deuterium which was retained in the volume and measure in-situ with a mass-spectrometer.

3. Results and discussion

Investigations made with scanning electron microscope (SEM) on the samples exposed to the high particle flux densities, revealed two types of blisters. At 530 K, small nanostructures of 20–30 nm diameter (Fig. 1a) and large ones of ~0.1 to 1 μm (~10^{13} m^{-2}) were detected. A clear dependence of blister distribution on grain orientation was observed on the modified surface which could be attributed to the plastic deformations as a consequence of high local stresses in addition to the slip systems mechanism [13]. With surface temperature increasing to 630 K, the density of large blisters decreased (~$5 \cdot 10^{12}$ m^{-2}) and the size of small blisters increased to ~40 nm (Fig. 1b). A cross-sectional cut using focused ion beam (FIB) showed the presence of voids a few hundreds of nanometers below the surface (Fig. 1b1). The high particle flux density exposure at 870 K (Fig. 1c) revealed only small blisters of a few tens of nanometers.

In case of exposure to low particle flux density at 530 K, blisters of a few micrometer were present at a lower concentration (~$4 \cdot 10^8$ m^{-2}) (Fig. 1d) compared to the high flux exposure. These structures were rarely distributed on the surface and their flat-top

shape has been reported previously [14]. On the sample exposed to 630 K at low flux (Fig. 1e), the presence of two types of blisters was detected ~2 μm and ~40 to 100 nm, whereas at 870 K the surface was not modified.

In Fig. 2 are given higher resolution SEM images of samples exposed to high particle flux density to show more clearly the presence of the nanostructures. The grain marked by a "X" sign in the Figs. (11a) and (2A) was modified by the presence of very small nanostructures of the order of a few tens of nanometer, resembling a spongy structure. At 630 K (Fig. 2B), the small nanostructures became more visible as their size increased whereas density decreased. At 870 K (Fig. 2C) were detected two types, blisters in the tens nm-range and blisters in the hundred nm-range as indicated with red circles [13]. Their density decreased and the size increased to about ~50 nm with increasing temperature.

To summarize the visual observations with the electron microscope and compare with previous investigations from the literature, in Fig. 3 is plotted the blister formation domain and size distribution relation to surface temperature and ion flux density. The total ion fluence in all cases is of the same order (~10^{26} D^+/m^2), surface temperature is in the range ~300 to 870 K and ion energy varies from 35 eV to 70 eV. Besides the symbols, in the graph is given the diameter of blisters in micrometer. The graph is arbitrarily sectioned in three regions in order to point out the blister diameter distribution. In the blue region are included exposures done at low flux and temperature where a considerable variety of blister diameters can be noticed. However, at higher particle flux density, within the same region, the blister diameter decreases as their density increases.

As seen from the white region on the graph, no surface changes are reported for temperatures higher than 700 K. For the present experiments, it is important to be emphasized the presence of small blisters (40–50 nm) at elevated particle flux densities and temperatures which were first observed by Xu [13]. These findings demonstrate a clear dependence of surface morphology changes of tungsten under deuterium particle impact on surface temperature and particle flux density.

It has been proposed that blisters are formed from specific kind of defects in the material such as vacancies, interstitials, and dislocations which act as traps for deuterium [18]. The binding energies of deuterium in these traps can be calculated by analyzing the shape and peak temperature of the TDS spectra [19]. To illustrate, in Fig. 4 are shown desorption data of deuterium during the heating up while the modeling of the spectra to find the binding energies for each case is ongoing.

The TDS spectrum of samples exposed at 530 K at high particle flux density has a peak at 840 K and a shoulder at 730 K (Fig 4). This indicates the presence of more than one type of traps with different binding energies. On the other hand, the deuterium desorption from the sample exposed at 530 K at lower flux density has a single peak temperature at around 875 K. The desorption profile of deuterium from the sample exposed to high particle flux density at 630 K mimics the variations of the temperature profile which is not entirely linear in the range 600–800 K as a result of a failure of the TDS setup during the measurements. The largest amount of deuterium from the sample exposed at high particle flux at 870 K was desorbed at around 865 K in a single and sharp peak whereas deuterium retention at 870 K was insignificant for the exposure at lower flux density.

The integrated amount of deuterium from the TDS profiles is given in Fig. 5. The total amount of desorbed deuterium for the low flux density cases are in a good agreement with the data provided by Alimov et al. [20]. Deuterium retention at 530 K and 630 K is higher for low flux exposures due to the temperature and time dependence of diffusion [23]. Shorter exposure time at Magnum-PSI compared to PSI-2, leads to lower diffusion of deuterium at