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Dislocation-mediated trapping of deuterium in tungsten under high-flux high-temperature exposures

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ABSTRACT

The effect of severe plastic deformation on the deuterium retention in tungsten exposed to high-flux low-energy plasma (flux ~10^{24} m^{-2} s^{-1}, energy ~50 eV and fluence up to 5×10^{25} D/m^2) was studied experimentally in a wide temperature range (460–1000 K) relevant for application in ITER. The desorption spectra in both reference and plastically-deformed samples were deconvoluted into three contributions associated with the detrapping from dislocations, deuterium-vacancy clusters and pores. As the exposure temperature increases, the positions of the release peaks in the plastically-deformed material remain in the same temperature range but the peak amplitudes are altered as compared to the reference material. The desorption peak attributed to the release from pores (i.e. cavities and bubbles) was suppressed in the plastically deformed samples for the low-temperature exposures, but became dominant for exposures above 700 K. The observed strong modulation of the deuterium storage in "shallow" and "deep" traps, as well as the reduction of the integral retention above 700 K, suggest that the dislocation network changes its role from "trapping sites" to "diffusion channels" above a certain temperature. The major experimental observations of the present work are in line with recent computational assessment based on atomistic and mean field theory calculations available in literature.

1. Introduction

Tungsten (W) is the main candidate plasma facing material (PFM) of a fusion reactor (see e.g. Refs. [1,2]). Although W clearly has a number of advantages, the retention and permeation of hydrogen isotopes (HI) remain a concern for both the efficient operation and safety of the reactor, given the extreme operation conditions that the PFM must sustain during service (e.g. see recent review by Tanabe [3]).

While at the low-temperature limit (450 K) the bulk permeation is suppressed and the formation of HI bubbles and cavities occurs mainly in the sub-surface region [3], at the high-temperature limit (1000 K and above), a significant contribution of the bulk retention at a depth of millimeter and beyond is to be accounted for. Another important point is that sub-surface trapping, expressed in the formation of bubbles and accompanied by plastic deformation and blistering, is a non-equilibrium process primarily defined by the combination of plasma flux, surface temperature (and its gradient towards the bulk) and material microstructure [3]. The latter truly plays a crucial role because it defines the density and strength of trapping sites inducing the formation of HI clusters that cannot nucleate otherwise in the perfect bulk, due to the absence of hydrogen self-trapping in bcc W [4,5]. Moreover, the initial microstructure of the PFC material will undergo constant modification due to the formation of plasma-induced defects (such as bubbles [6] and dislocation tangles [7]), neutron irradiation lattice defects (Frenkel pairs and their clusters [8]) and thermal fatigue-induced defects (dislocation pile-ups and slip bands [9]). That is why significant effort is currently devoted to understanding the role of the initial microstructure in the HI permeation, dynamic trapping and release.

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Among the naturally existing and radiation-induced defects, the following are of special relevance with respect to HI retention: vacancies (and small vacancy clusters), voids, grain boundaries and dislocations. Trapping at dissolved interstitial impurities (C, O) is also possible, but generally considered to be too weak to initiate nucleation of HI bubbles at temperatures above 450 K [3]. Focus is, therefore, put on studies allowing to reveal (and preferably understand) the role played by each type of the above mentioned defect microstructures.

Implantation with high-energy H ions is an efficient way to address trapping at point defects, as one can select the beam energy in the narrow range needed to create Frenkel pairs without producing collision cascades [10]. Investigation of ion pre-implanted samples is more complex, but allows one to probe samples that contain both dislocation loops and voids simultaneously [11]. Comparison of results obtained using single crystals and recrystallized polycrystalline grades allows to reveal the trapping efficiency of grain boundaries (GBs). Recently, a comparison of HI retention and microstructure after high-flux deuterium (D) plasma exposure was performed in recrystallized and heavily plastically deformed samples [12,13]. All microstructural features, except from the dislocation density (increased by two orders of magnitude), remain the same after plastic deformation. The exposures were performed at 450 K (low-temperature limit) in the dose range of 8×10^{24}–3×10^{26} D/m^2/s, using the high-flux linear plasma accelerator PILOT-PSI.

The obtained results have shown that heavy plastic deformation increases the total D retention by ~50%, as compared to the reference recrystallized sample. The contribution of D trapping by pores to the total retention is suppressed by plastic deformation up to a certain dose and increases beyond it. This has been explained by the dual role of the dislocation network: on one hand offering preferential nucleation sites, and on the other hand delaying the formation of HI bubbles to a higher dose as compared to the reference sample, due to the increased density of trapping sites. Hence, the plastically-induced dislocation network delays the formation of large HI bubbles, but such delaying capacity is limited.

To extend our understanding of the role played by plastic deformation in the HI diffusion, trapping and release processes, it is important to screen the effect of temperature and in particular to reveal whether the dislocation-enhanced retention also occurs above 800 K, where the retention in low-dislocation density grades (i.e. coarse-grained and recrystallized grades) strongly decreases [14]. In this work, we perform a series of high-flux exposures in the 470–1000 K range, using the same flux exposure parameters, detection/analysis techniques and material as in earlier studies [12,13].

2. Experimental details

The pristine material used in this study was a polycrystalline W of 99.99% purity, provided in a rod form by Plansee AG [15]. The main impurities, as reported by the manufacturer, are listed in Table 1. The removal of initial residual stresses was performed by a thermal treatment at 1273 K, followed by recrystallization at 1873 K for 1 h. Generally, the microstructure of the as-received material consisted of large random grains (separated by high-angle GBs) that contained subgrains (separated by low-angle GBs), while dislocations were present both inside the grains as well as at low-angle GBs. Upon thermal annealing (at 1873 K), microstructural recovery was expressed in a limited subgrain growth, removal of a significant amount of subgrains and reduction in the dislocation density. The grain size of the as-annealed material typically varied in the 50–150 μm range and the dislocation density was about 5×10^{12} m^{-2}.

The samples intended for tensile plastic deformation were cut from the as-annealed rods. To release the stress introduced by electric discharge machining (EDM), the tensile samples were re-annealed at 1273 K for 1 h in an argon (Ar) atmosphere. Several tensile tests were performed at 873 K in air at a deformation rate of 0.2 mm/min to reach 28% deformation. The latter approximately corresponds to the ultimate tensile strength of the studied W grade. Transmission electron microscopy (TEM) and electron backscatter diffraction (EBSD) samples were cut from the middle of the samples and re-annealed at 873 K for three hours prior to the examination to remove the EDM-induced defects without affecting the dislocation microstructure.

The microstructure of the annealed and plastically-deformed tungsten samples was studied. Scanning electron microscopy (SEM) analysis of the as-annealed samples revealed the growth of random grains up to 150 μm, and the strong reduction of the fraction of low-angle GBs down to about 20%. Examples of EBSD grain orientation maps are given in Fig. 1a and b for as-annealed and plastically-deformed samples, respectively. Three main crystallographic orientations of the W bcc lattice (i.e. ⟨111>, <001⟩ and ⟨101⟩) can be identified on the EBSD maps. Fig. 1a and b shows that the large random grains are practically free of subgrains and that small grains (5–10 μm) are rarely observed.

In the case of the deformed material (Fig. 1b), the effect of tensile loading is obvious and one can see that the grains are elongated in accordance with the orientation of the sample with respect to the tensile loading direction. However, the applied deformation did not result in grain refinement that was clearly detectable by means of EBSD. To provide a deeper insight in the possible microstructural modifications induced by plastic deformation, a TEM investigation was performed.

TEM samples ($10 \times 3 \times 1.25$ mm^3) were extracted from the middle of both as-annealed and as-deformed specimens (plates with dimensions of $10 \times 10 \times 1.25$ mm^3) by EDM. These pieces were then mechanically polished from both sides using SiC paper (grits: 220, 500, 1200 and 4000) to achieve 70–100 μm thickness and further cut with a wire cutter into pieces to fit 3-mm diameter TEM grids. They were polished again from both sides with 4000 SiC paper to remove the remnants of glue, rinsed in acetone and ethanol and then glued on ∅ 3-mm copper grids with an aperture of 1 mm. Finally, the specimens were polished electrochemically with a solution of 1.5 wt% NaOH in water using an applied voltage of 15 V. The specimens were investigated by means of a JEOL 2010 TEM operating at 200 kV and a JEOL 3010 TEM operating at 300 kV. Examples of bright field TEM images of dislocations observed in as-received and plastically-deformed samples are presented in Fig. 1c and d. The presence and density of dislocations was inhomogeneous in the as-annealed sample, even within a single grain. After plastic deformation, dislocations became evident everywhere in the sample visible area, while the scatter in their spatial distribution was reduced.

The average dislocation density was measured based on the method described in Ref. [16]. Several calculations at different specimen areas were performed to get an average dislocation density. Each calculation requires a TEM micrograph, corresponding selected area diffraction (SAD) pattern and convergent beam electron diffraction (CBED) pattern. In the Digital Micrograph software, which is provided with the image sensor of the TEM, a circle is drawn randomly in an image and the number of its intersections with dislocation lines is counted. The dislocation density is then calculated as $\rho = 2N/Lt$, where N is the number of intersections of the circle with dislocation lines, L is the circle diameter, and t is the local thickness of the specimen in the area of the image. The length of the circle is automatically calculated by the Digital Micrograph software. The local thickness of the specimen is