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Surface modifications and deuterium retention in polycrystalline and single crystal tungsten as a function of particle flux and temperature

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ABSTRACT

The effects of particle flux and exposure temperature on surface modifications and deuterium (D) retention were systematically investigated on four different tungsten (W) microstructures. As-received, recrystallized, and single crystal W samples were exposed to D plasmas at surface temperatures of 530–1170 K. Two different ranges of D ion fluxes (10^{22} and 10^{24} $D^+m^{-2}s^{-1}$) were used with the ion energy of 40 eV and particle fluence of 10^{26} D^+m^{-2}. Increasing the particle flux by two orders of magnitude caused blister formation and D retention even at temperatures above 700 K. The main effect of increasing the particle flux on total D retention was the shifting of temperature at which the retention was maximal towards higher temperatures. Diffusion-trapping simulations were used to fit the thermal desorption spectroscopy (TDS) release peaks of D, yielding one or two types of trapping sites with de-trapping energies around 2 eV.

1. Introduction

Tungsten is the selected plasma-facing material to be used on the high-heat flux region in the ITER divertor due to its favorable properties, such as high melting temperature and thermal conductivity, low hydrogen retention and low erosion yield [1,2]. Exposure to energetic particles such as hydrogen isotopes (D and tritium (T)), helium (He) and impurities coming from the plasma, causes gas retention and surface modifications (i.e. formation of blisters, bubbles, fuzz, cracking, etc.), which may lead to an increase in the erosion yield, dust formation and deterioration of material thermal and mechanical properties [3–5].

Plasma-facing materials in ITER will be exposed to particle fluxes that vary spatially by several orders of magnitude (10^{20}–10^{24} $D^+m^{-2}s^{-1}$), ion energies 0.1–100 eV and surface temperatures in the range 370–1370 K [2]. Recent studies have shown that blistering and D retention in as-received polycrystalline W occur even at temperatures above 700 K when exposed to plasma fluxes of 10^{24} $D^+m^{-2}s^{-1}$ and above. At higher particle fluxes, the local D concentration is higher for the same temperature and hence the supersaturation happens at even higher temperatures. Therefore, a higher temperature is required for inward diffusion to balance the incoming flux and avoid local supersaturation [6–9]. Trapping of D at grain boundaries and intrinsic defects in the material, such as dislocations and vacancies, which act as centers for bubble nucleation, leads to blister formation [7,10,11,12]. The impact of material microstructure on the observed flux effects on blister formation and D retention, however, has not yet been systematically studied.

This paper is built upon previous work [6,7] and it addresses the role of grain boundaries and intrinsic defects on D diffusion and trapping in addition to reporting the particle flux effects on retention and surface morphology changes. Recrystallized W samples with negligible dislocation density and single crystals were used to separately assess the impact of grain boundaries and dislocations on D trapping. Results are compared with previously reported data on as-received polycrystalline W samples and TDS

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profiles were modeled with the rate equations approach implemented in the Coupled Reaction Diffusion Systems (CRDS) code [13].

2. Experimental

Two types of polycrystalline W samples with different grain sizes, small grain (SGW 99.94%) with 20 μm diameter and large grain (LGW 99.94%) with 40 μm diameter, were used as received from the supplier. Details of the sample preparation for SGW and LGW are described in [6] and [7]. Recrystallized W samples (RECW) were prepared by thermally treating LGW samples at 2273 K for 30 min which resulted in the formation of grains with an average size of 50 μm. Single crystal W samples (SCW) were supplied by Mateck GmbH (MaTecK Material-Technologie & Kristalle GmbH n.d.). The purity of the material was 99.999% and the (110) crystal orientation had an accuracy of $\leq 0.1^{\circ}$.

Samples were cut in discs with a diameter of 14 mm and 4 mm thickness. In order to ensure consistent and comparable results in terms of surface analysis techniques and D retention in the material, all W samples were mechanically polished to a mirror finish ($R_a \leq 40$ nm). Afterwards, they were ultrasonically cleaned in alcohol and acetone to remove the polishing impurities and subsequently outgassed at 10^{-6} mbar at 1273 K for 1 hour. Scanning electron microscopy (SEM) images of RECW and SCW samples before plasma exposures are shown in Fig. 1.

W samples were exposed to D plasmas at surface temperatures of 530–1170 K to two different ranges of D ion fluxes (10^{22} and 10^{24} $D^+m^{-2}s^{-1}$) and the particle fluence was kept constant in all cases (10^{26} D^+m^{-2}). High particle flux exposures were carried out in Magnum and Pilot-PSI linear plasma devices [14,15] while low particle flux experiments were conducted at PSI-2 [16]. Changing the physical contact with the water-cooled sample holder allowed sample temperature variation. A fast infrared camera (FLIR SC6500MB) was used for the temperature measurements in three plasma devices. The details of the experimental setup, plasma properties and sample mounting specifics are described elsewhere [6,7].

The post-mortem analysis was done using scanning electron microscopy (SEM), thermal desorption spectroscopy (TDS) and secondary ion mass spectroscopy (SIMS) to investigate the surface modifications, total D retention and near-surface D depth distribution [6,7]. Quantitative D depth profiles were obtained with nuclear reaction analysis (NRA) at the tandem accelerator at IPP-Garching in Germany. A ^{3}He ion beam was used to probe the D content by detecting and analyzing the products of the nuclear reaction D (^{3}He,p) α [17]. During the measurements, the proton and alpha detectors were set at 135° and 102° angles respectively, and the beam spot was about 1 mm^2. The ^{3}He ion beam energy was varied in the range of 0.5–6 MeV. The calculation and deconvolution of experimental proton and alpha particle energy spectra recorded at various incident ^{3}He ion energies were done using the SIMNRA [18] and NRADC [19] codes and energy dependent cross-sections [20,21].

3. Results and discussion

After exposure to pure D plasma, a pronounced dependence of surface modifications on initial material microstructure, incident particle flux and surface temperature was observed. Results on surface modifications of SGW and LGW samples are described in previous publications [6,7]. To summarize briefly, at low particle fluxes (10^{22} $D^+m^{-2}s^{-1}$) blister formation was suppressed for surface temperatures above 700 K, while at high particle fluxes (10^{24} $D^+m^{-2}s^{-1}$) blisters were still observed even on samples exposed at 1170 K. This is illustrated by SEM images of SGW and LGW samples exposed at 1170 K at high flux shown in Fig. 2 that demonstrates a variety of blister size and areal density.

After exposure to low particle flux, no blisters were formed on RECW samples for surface temperatures higher than 520 K. At 520 K blisters were formed and had a diameter of 10–20 μm and a flat top surface. From the cross-sectional cut with the focused ion beam (FIB) on a blister, cracking along the grain boundary, starting at a depth of 4 μm and extending down to 16 μm was observed (Fig. 3). Alimov et al. found similar flat topped blisters on W samples recrystallized at 2073 K and exposed to similar plasma conditions at 595 K [22]. Balden et al. found such blisters to appear at a rather low areal density after exposure to an ion flux of 10^{20} $D^+m^{-2}s^{-1}$ at 500 K and referred to these surface modifications as extrusions [23]. No blisters were found at high particle flux on RECW samples, independently of the surface temperature. We hypothesize that the lack of blistering at high temperatures may be related to the low amount of nucleation sites (e.g. dislocations) on recrystallized W after annealing and higher probability of D to diffuse through grain boundaries after being thermally activated.

Since the fluence was kept constant throughout all the experiments, the exposure time at low flux was longer compared to the high flux therefore; higher amounts of D were trapped in the defects at low exposure temperature [24]. At low flux and high temperatures, long exposure times promote deeper D diffusion into the bulk. At high flux, larger concentration gradients of D can build up in the near-surface region, potentially leading to stronger material damage and/or faster growth of D filled bubbles or voids. At the same time, diffusion of D deeper into the material leads to D trapping at intrinsic lattice defects, such as vacancies, dislocations and grain boundaries. Dislocation loops and grain boundaries can

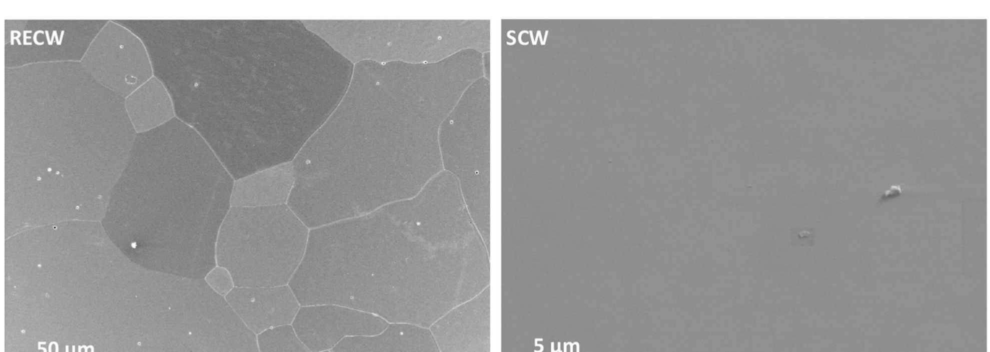

Fig. 1. Scanning electron microscope (SE mode) images of recrystallized (RECW) and single crystal (SCW) samples taken before the plasma exposure.