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Abstact

Surface topography and deuterium retention in re-crystallized tungsten have been examined after exposure to a low-energy (38 eV/D), high-flux (10^{22} D/m^{2}s) deuterium plasma at ion fluences of 10^{26} and 10^{27} D/m^2 and various temperatures. The methods used were scanning electron microscopy equipped with focused ion beam, thermal desorption spectroscopy, and the D(^{3}He,p)^{4}He nuclear reaction at ^{3}He energies varied from 0.69 to 4.0 MeV. During exposure to the D plasma, blisters with various shapes and sizes depending on the exposure temperature are formed on the W surface. At the temperatures above 700 K no blisters appear. The deuterium retention increases with the exposure temperature, reaching its maximum value of about 10^{22} D/m^2 at 480-530 K, and then decreases to about 10^{19} D/m^2 at 800 K.

Keywords: Blistering; Deuterium; Deuterium retention; High ion flux; Tungsten

1. Introduction

As plasma-facing material for fusion reactors, tungsten (W) will be subject to intense fluxes of energetic deuterium and tritium ions and neutrals. This implantation process leads to concerns about hydrogen isotope inventories after long-term deuterium-tritium plasma exposure. Most of the results on hydrogen isotope retention and recycling for W have been reviewed by Causey and Venhaus [1, 2], and Skinner et al [3]. Available data ([3] and references therein) have shown that the hydrogen isotope retention in W materials exposed to high-flux hydrogen plasmas differs from that for ion implantation. In addition, there is evidence of blistering occurred on tungsten surface exposed to hydrogen plasmas with ion energies well below the displacement threshold [4-8]. Not much is known about the hydrogen retention in different W materials irradiated with low-energy hydrogen ions and exposed to low-energy, high-flux hydrogen plasma. Especially, the temperature dependence of blistering and hydrogen retention in tungsten exposed to the low-energy hydrogen plasmas at high ions fluences (above 10^{26} ions/m^2) was not studied systematically. In this study we made an attempt to make up for this deficiency.

2. Experimental

Polycrystalline tungsten (A.L.M.T. Corp., Japan) with a purity of 99.99 wt.% fully re-crystallized at 2070 K for 1 hour after cutting and polishing was used. The linear plasma generator used for delivering a plasma beam comparable to the edge plasma at ITER divertor is described elsewhere [9]. To generate a D plasma, D_2 gas was filled in the plasma generation section to a pressure of 1 Pa. In doing so, a plasma beam highly enriched with species of D_2^+ to over 80% was obtained [9]. The bias voltage of -80 V was applied to the W sample resulting in an incident energy of 38 eV/D, taking into account the plasma potential of about -4 V measured by a Langmuir probe. The incident deuterium ion flux was fixed at 10^{22} D/m^2s, and the samples were exposed at ion fluences of 10^{26} and 10^{27} D/m^2. The sample was passively heated by the plasma itself and the exposure temperature was set by the thermal contact between the sample and the cooled holder.

The D concentration in the plasma-exposed W samples was measured by means of the D(^{3}He,α)H reaction, where both, the α particles and protons were analyzed. To determine the D concentration at larger depths, an analyzing beam of ^{3}He ions with energies varied from 0.69 to 4.0 MeV was used. The proton yields measured at different ^{3}He ion energies allow measuring the D depth profile at depths of up to 7 μm [10].

Total deuterium retention in the W samples was monitored ex-situ using thermal desorption spectrometry (TDS). An infrared heater was used to heat the samples at a ramp rate of 0.5 K/s and the sample temperature was raised to 1300 K. HD, and D_2 molecules released during TDS run were monitored by quadrupole mass spectrometer (QMS). To calculate the relative contribution of the recorded HD and D_2 masses to the total release of deuterium, the partial currents of the QMS were normalized as described in Ref. [11]. A standard D_2 leak with an inaccuracy smaller than 10% was employed to calibrate the QMS after each TDS analysis.

The surface topography was examined by scanning electron microscopy (SEM), whereas the three-dimensional sub-surface morphology of the plasma-exposed W samples was analyzed by a field emission SEM combined with a focused ion beam (HELIOS NanoLab 600, FIB). The Ga ion beam allowed surface cross-sectioning after selecting a special micrometer-sized feature on a material, investigating its surface topography and analysing its morphology beneath the surface. For the cross-sectioning, the surface normal was aligned to the ion beam. To reduce artifacts caused by the cross-sectioning (e.g., curtaining effect), prior to cutting the investigated surface was coated in-situ with a Pt-C film. For this purpose, a gaseous $C_9H_{16}Pt$ was injected and decomposed by scanning ion beam into a solid fraction on the selected surface.

3. Results

3.1. Surface topography and sub-surface morphology

After exposure to a fluence, Φ, of 10^{26} D/m^2 at temperatures, T_{exp}, in the range from 320 to 370 K, only sparse low-dome blisters with sizes of a few microns or less are observed (Fig. 1 (a)). At T_{exp} = 400-500 K, the blisters become much denser and the domes of blisters become higher. Peculiar changes occur at T_{exp} = 520-570 K, where two kinds of blisters appear: large blisters with sizes of a few tens of microns and small cone-shaped blisters with diameters of less than a few μm (Fig. 1 (c)). According to electron backscattering

diffraction analysis, small blisters appear preferably on the grains with nearly (111) surface orientation [7]. At exposure temperatures in the range from 580 to 670 K, the small blisters disappear, whereas the large blisters become sparser with the increasing temperature (Fig. 1 (e)).

The increase in fluence to 10^{27} D/m^2 leads to the appearance of dense and low-dome blisters with sizes up to 15 μm even at T_{exp} = 320-370 K (Fig. 1 (b)). Two kinds of blisters appear already at exposure temperature of 460 K (Fig. 1(d)). At higher exposure temperatures, the temperature dependence of blistering at $\Phi = 10^{27}$ D/m^2 (Fig. 1 (f)) is similar to that at $\Phi = 10^{26}$ D/m^2.

For both fluences, no blisters appear at the exposure temperatures above 700 K.

Three-dimensional sub-surface morphology of blister-like structures at $\Phi = 10^{27}$ D/m^2 ranges from fine intragranular crack-like defects with width of ~0.1 μm and length of ~5 μm at T_{exp} = 360 K (Fig. 2 (a)) to large cavities (hundreds of cubic microns) at T_{exp} = 595 K (Fig. 2 (c)). At exposure temperature of about 600 K the defects accumulate at grain boundaries and, therefore, the cavities reach easily depth up to 20 μm. On the contrary, at lower temperatures they lead to strongly stressed regions (hundreds of cubic microns) and cracks at grain boundaries and inside the crystallites. The surface blister-like structures and the defects underneath are correlated along crystallographic orientations of the W grains in accordance to the low-indexed sliding systems [12].

3.2. Depth profiles

In the re-crystallized W exposed to the D plasma at T_{exp} = 330 K to $\Phi = 10^{26}$ D/m^2, the deuterium depth profile is characterized by a sharp near-surface concentration maximum of 1-2 at.%, and, at depths above 1 μm, by a concentration of about 10^{-2} at.% slowly decreasing into the bulk (Fig. 3 (a)). As the exposure temperature increases up to 530 K, the D concentration in the near-surface decreases, whereas the concentration at depths of 1-3 μm reaches the maximum value of about 1 at.%. Further increase of the exposure temperature leads to a uniform decrease of the D concentration (Fig. 3 (a)).

As the ion fluence increases to 10^{27} D/m^2 at T_{exp} = 320 K, the D concentration at depths of 1-2 μm ia significantly increased (Fig. 3 (b)). At T_{exp} = 480 K, the D profile demonstrates, in addition to the near-surface peak, the concentration of about 1 at.% with no decrease into the bulk up to a depth of 7 μm. For both ion

fluences, the maximum D concentration is about 1 at.%, and this D concentration is achieved at the exposure temperatures of 480-535 K. This high D concentration could be due to accumulation of D_2 molecules in cavities created during the D plasma exposure.

3.3. Total retention:

In the re-crystallized W exposed to the D plasma at $\Phi = 10^{26}$ D/m^2, the deuterium retention is (2-3)×10^{20} D/m^2 at T_{exp} = 320 K and, as exposure temperature increases, rises to its maximum of about 7×10^{21} D/m^2 at T_{exp} = 530 K and then decreases down to about 10^{19} D/m^2 at T_{exp} = 775 K (Fig. 4). Increasing the fluence to 10^{27} D/m^2 at exposure temperatures in the range from 320 to 500 K results in significant increase of the D retention. At $\Phi = 10^{27}$ D/m^2 the maximum retention of about 10^{22} D/m^2 is observed at T_{exp} = 480 K. At exposure temperatures of 550-650 K, the D retention at $\Phi = 10^{27}$ D/m^2 decreases but remains higher by a factor of about 3 than that at $\Phi = 10^{26}$ D/m^2. At T_{exp} above 750 K, the D retention is about 10^{19} D/m^2 for both ion fluences (Fig. 4).

From the comparison of the TDS and NRA data (Fig. 4) it is clear that at $\Phi = 10^{26}$ D/m^2 and irradiation temperatures up to 700 K about 50% of retained deuterium is localized in the sub-surface layer up to 7 μm. However, at $\Phi = 10^{27}$ D/m^2 the fraction of deuterium retained at depths above 7 μm significantly increases.

4. Discussion

For tungsten irradiated with D ions at energies well below the displacement threshold, the mechanism of plastic deformation due to deuterium super-saturation [13] must be considered for modification of the sub-surface structure and formation of trapping sites for deuterium [14, 15]. During exposure to low-energy, high-flux D plasma, the D concentration in the implantation zone greatly exceeds the solubility limit and stresses the matrix lattice until plastic deformation occurs to alleviate these tensions [13]. This deformation is assumed to be responsible for the generation of vacancies, vacancy complexes and microscopic cavities at depths of several micrometers and the concurrent accumulation of diffusing deuterium.

On the other hand, according to the theory of formation of superabundant vacancies in the presence of interstitial hydrogen atoms [16], the formation energy of a vacancy is decreased owing to vacancy-hydrogen cluster formation and the configurational entropy of the system at high hydrogen concentration. Recently Shu [17] considered that the mechanism of vacancy formation due to the lowering of the vacancy formation energy by trapping of deuterium can also be valid for tungsten exposed to low-energy, high-flux D plasma. The deuterium-vacancy clusters may diffuse deeply into the bulk and agglomerate resulting in microscopic cavities.

At long-term irradiation the diffusing D atoms recombine on the cavity surfaces, increasing thus the D_2 gas pressure inside these cavities. The cracks, distorted areas, and large cavities beneath the surface (Fig. 2) are caused by stress exceeding the toughness of the material. The stress is may be increased by the gas overpressure inside the cavities. At elevated temperatures exceeding the brittle-to-ductile-transition temperature, the stress can be relaxed by dislocations moving along lattice planes through the whole crystallite leading to the cavities at the grain boundaries [18]. This corresponds to the observed material migration above the surface, i.e., the blister-like surface topography. However, at near-room temperatures the stress relaxation results in plastic deformation and crack formation inside the grains. Thus, the temperature dependence of the structure modification correlates well with the temperature dependence of fracture toughness for W [18, 19].

A thorough discussion about mechanisms of the blister nucleation and growth would require additional experimental information, such as transmission electron microscopy and SEM studies. It should also be accompanied by model calculation taking into account the stress field caused by hydrogen super-saturation within the near-surface layer, formation and diffusion of superabundant deuterium-vacancy clusters in bcc metals, and the movement of dislocations along crystallographic orientations.

In closing it is pertinent to note that the fluence and temperature dependence of the D retention is a consequence of the sub-surface structure modification and accumulation of deuterium in the cracks and cavities.

5. Summary

Significant temperature dependence of surface morphology and deuterium retention is found for re-crystallized W exposed to low-energy (38 eV/D), high-flux (10^{22} D/m^{2}s) D plasma at ion fluences of 10^{26} and 10^{27} D/m^2. At temperatures of 320-400 K, only sparse blisters with diameters of 0.5-2 μm are formed on the W surface. In this case the D depth profiles are characterized by a near-surface concentration maximum of 1-2 at.% and, in the sub-surface layer (at depths from 1 to 7 μm), by a concentration of 0.02-0.2 at.% slowly decreasing into the bulk. As the exposure temperature increases, the blisters become much denser. Two kinds of blisters appear: large blisters with sizes of 10-30 μm and small cone-shaped blisters with diameters of less than a few μm. As this takes place, the D concentration in the sub-surface layer reaches 1 at.%. No blisters appear at temperatures above 700 K, and the D concentration at depths of several micrometers is about 10^{-3} at.%.

Stress-induced plastic deformation caused by deuterium super-saturation within the near-surface layer and formation of superabundant deuterium-vacancy clusters are suggested as mechanisms for nucleation and growth of microscopic cavities at depths up to several micrometers. At long-term plasma exposure, the diffusing D atoms recombine on the cavity surfaces, increasing thus the gas pressure inside these cavities, which may support the formation of blister-like structures.

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Figure captures

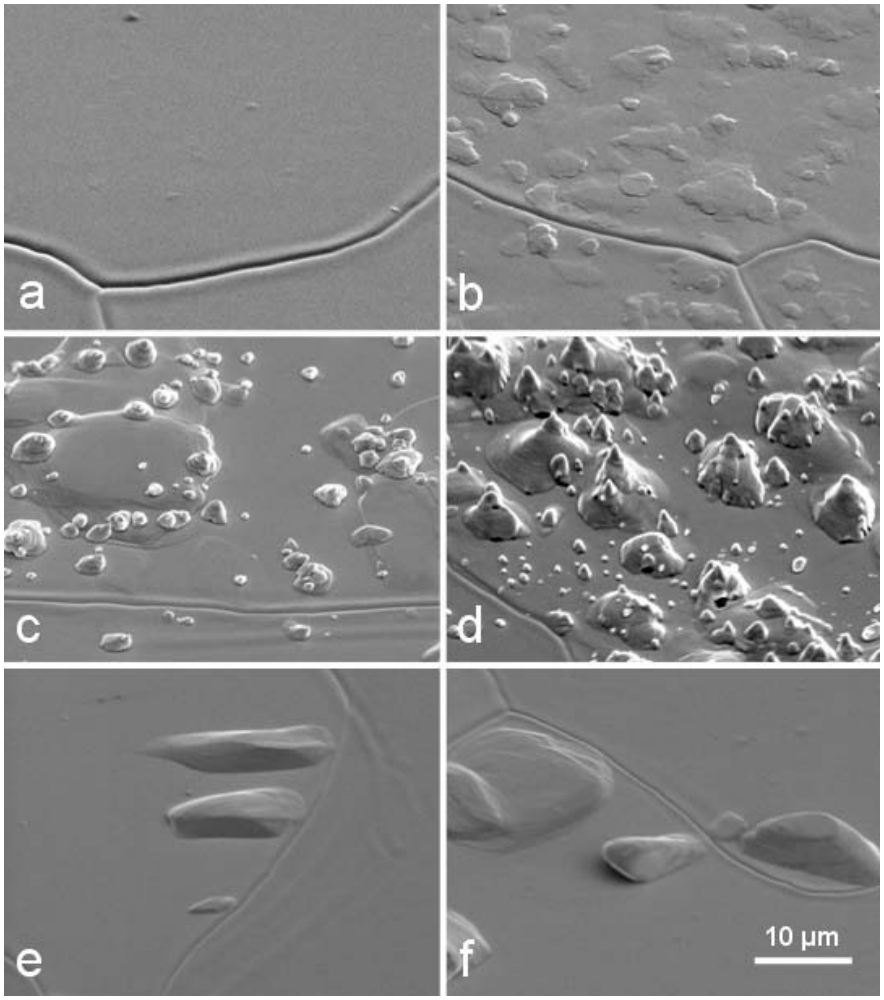
Figure 1. SEM images of re-crystallized W exposed to low-energy (38 eV/D), high-flux (10^{22} D/m^2 s) D plasma with ion fluences of 10^{26} D/m^2 (a, c, e – left part of the figure) and 10^{27} D/m^2 (b, d, f – right part of the figure) at temperatures of 360 K (a, b), 530 K (c), 460 K (d), 600 K (e), and 595 K (f). The magnification of all images is the same. The surfaces were tilted at an angle of 45° to the electron beam.

Figure 2. Cross sectional images of re-crystallized W exposed to low-energy (38 eV/D), high-flux (10^{22} D/m^2 s) D plasma with an ion fluence of 10^{27} D/m^2 at 360 K (a), 480 K (b), and 595 K (c). Take note that prior the cross-sectioning the surfaces were coated with a Pt-C film. The surfaces were tilted at an angle of 52° to the electron beam.

Figure 3. Depth profiles of deuterium retained in re-crystallized W exposed to low-energy (38 eV/D), high-flux (10^{22} D/m^2 s) D plasma with ion fluences of 10^{26} (a) and 10^{27} D/m^2 (b) at various temperatures.

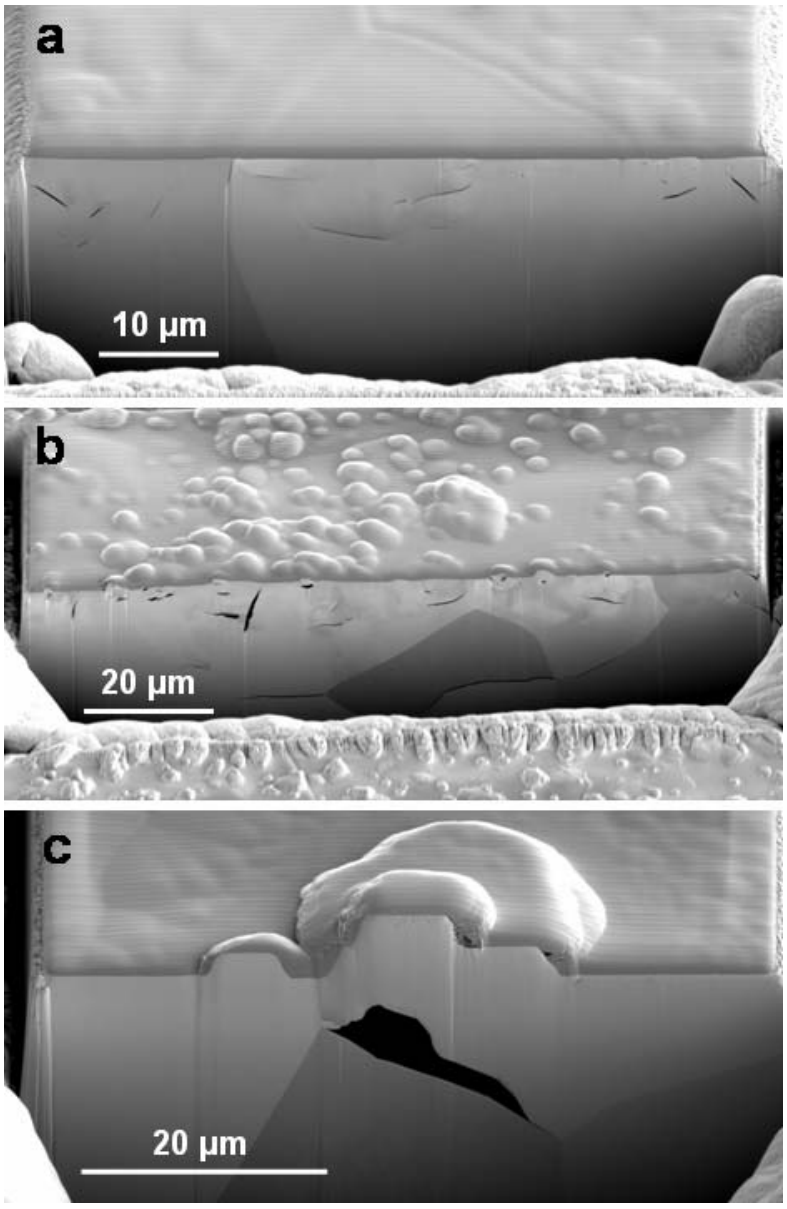
Figure 4. Deuterium retention in re-crystallized W exposed to low-energy (38 eV/D), high-flux (10^{22} D/m^2 s) D plasma to ion fluences of 10^{26} and 10^{27} D/m^2, as a function of the exposure temperature. The total deuterium retention was determined by thermal desorption spectrometry (TDS) (solid points), whereas the D retention up to a depth of 7 μm was measured by nuclear reaction analysis (NRA) (open points).

Fig. 1



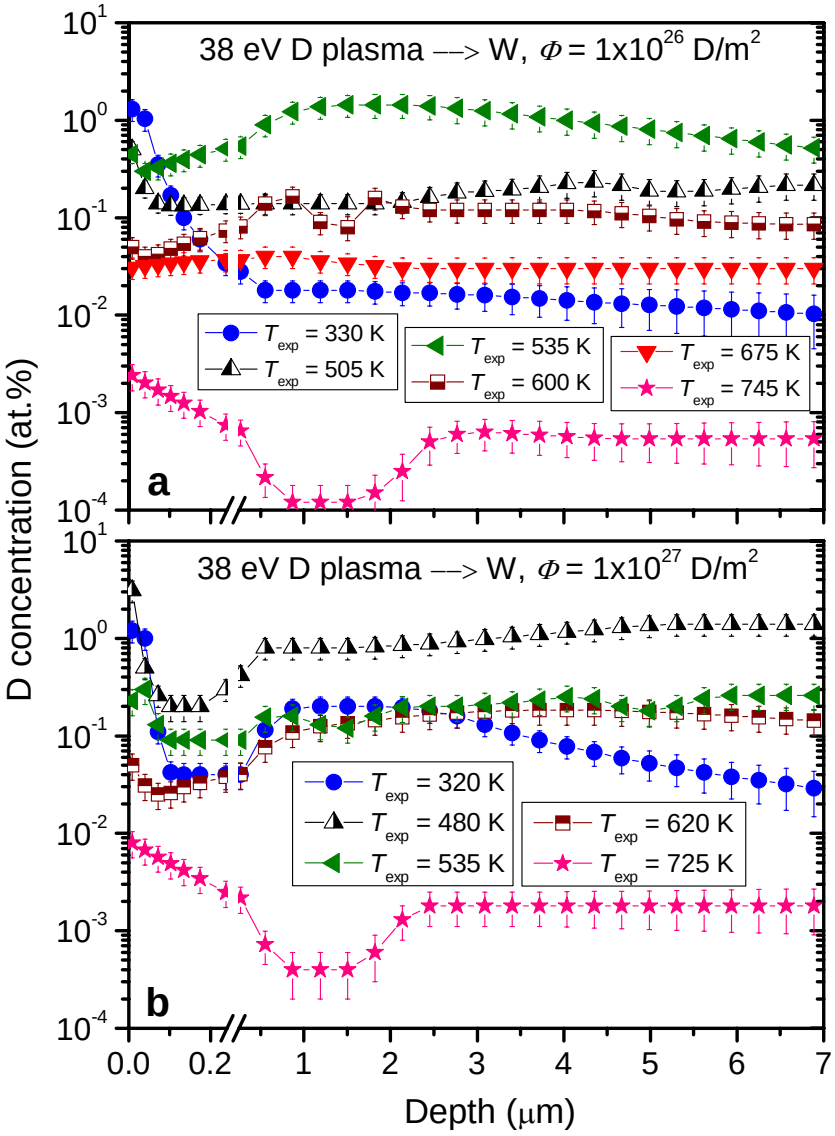
[00119]_(Alimov)_one column

Fig. 2



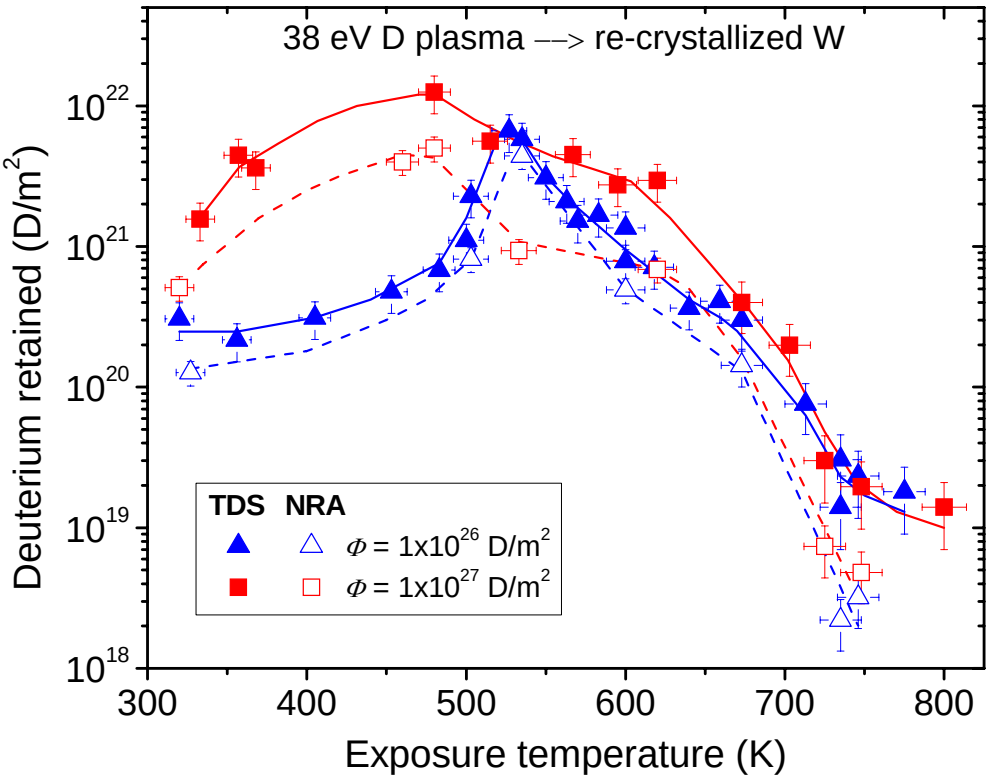
[00119]_(Alimov)_one column

Fig. 3



[00119]_(Alimov)_one column

Fig. 4



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