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Impact of plastic deformation on retention under pure D or He high flux plasma exposure

A. Bakaeva\textsuperscript{a,b}, D. Terentyev\textsuperscript{a,*}, T.W. Morgan\textsuperscript{c}, A. Dubinko\textsuperscript{a}, W. van Renterghem\textsuperscript{a}, L. Tanure\textsuperscript{c,d}, K. Verbeken\textsuperscript{d}

\textsuperscript{a} SCK-CEN, Nuclear Materials Science Institute, Boeretang 200, 2400 Mol, Belgium
\textsuperscript{b} Department of Applied Physics, Ghent University, St. Pietersnieuwstraat 41, 9000 Ghent, Belgium
\textsuperscript{c} FOM Institute DIFFER, De Zaale 20, 5612 AJ Eindhoven, The Netherlands
\textsuperscript{d} Department of Materials, Textiles and Chemical Engineering, Ghent University (UGent), Technologiepark 903, 9052 Ghent, Belgium

\section*{A R T I C L E  I N F O}

Keywords:
- W\textsuperscript{2}g tungsten
- He-H exposure
- High flux plasma
- Deformation

\section*{A B S T R A C T}

The retention of deuterium (D) and helium (He) is studied in pure tungsten after high flux mono-plasma exposure. The recrystallized and plastically deformed tungsten samples are studied to clarify the impact of the material microstructure, in particular dislocation density, on the trapping and release of D and He. Thermal Desorption Spectroscopy (TDS) measurements are performed to reveal the release stages and quantify the retention. Preliminary transmission electron microscopy study was applied to clarify the microstructural modifications induced by the plasma exposure to support the discussion and conclusions. It has been demonstrated that plastic deformation causes considerable suppression of He release within the explored limit of the TDS temperature – 1300 K. This is opposite to what is found for the pure D exposure, where the plastic deformation evidently enhances the D retention, given equivalent exposure conditions in terms of surface temperature and ion fluence.

\section*{1. Introduction}

Due to a number of unique properties, tungsten is considered as the plasma-facing material for the ITER divertor and DEMO first wall \cite{1}. Interaction of tungsten with fusion-relevant plasma ions, such as deuterium, tritium and helium, causes modifications in the surface microstructure (e.g. blistering, swelling, etc.) as well as in its mechanical properties (e.g. increased hardness, appearance of micro-cracks). This originates from the penetration of the gaseous atoms inside the material, the formation of bubbles and local plastic deformation, induced by thermal stresses. The impact on the microstructural changes is defined by the irradiation conditions (see e.g. \cite{2}) and initial microstructural state. Overall, a plasma facing material will undergo modification of its initial properties due to the accumulation of gas atoms in the sub-surface region (see \cite{3} for a recent overview) and lattice defects. In addition to the ion-driven plasma exposure, the plasma surface material/component will be a subject to cyclic thermal loading and neutron irradiation \cite{4}. These processes will cause the plastic deformation and generation of lattice defects such as voids and dislocation loops \cite{5}. To project how the plasma-material interaction will undergo in the material subjected to considerable plastic deformation, it is important to perform comparative study engaging reference and plastically deformed material to clarify possible impact of the plastic deformation with respect to the total retention and kinetics of the release of plasma components.

Our earlier works were dedicated to investigation of the impact of plastic deformation on the retention and release under pure D exposure \cite{6-8} in the ITER specification tungsten subjected to recrystallization at 1600\textdegree C (referred to “reference material” in this work). It has been generally concluded that under low temperature exposure conditions (i.e. below 600 K) a dislocation network facilitates the nucleation of defects and thereby enhances the trapping of D. Overall, this leads to an increase in D retention seen as amplification of the release stages on the thermal desorption spectrum (TDS), however the positions of the stages do not shift remarkably, implying that similar types of traps operate in the plastically-deformed material.

As of now, there is no experimental information reporting the impact of the plastic deformation on the He trapping and release. Most of the studies were performed to understand the impact of He seeding in the mixed exposures or influence of He energy, flux and fluence. Alimov et al. performed TDS analysis after high flux helium-seeded plasma exposures of recrystallized pure W (ALMT grade) \cite{2}. The energies of D...
and He ions were fixed at 38 eV and 76 eV, respectively. The TDS analysis revealed three major release stages at 550 K, 650 K and 800 K. Seeding of He ions into the plasma at exposure temperatures below 350 K did not change strongly the D retention, while at temperatures above 400K the D retention decreases significantly compared to that for the pure D plasma exposure. This was taken as an argument indicating that He seeding results in the confinement of D retention in the sub-surface region preventing long range diffusion. With respect to He retention itself, it was found not to depend on the He ion concentration in the D plasma. He retention was seen to increase with the exposure temperature from about 2 × 10¹⁹ He m⁻² at 340 K up to about 3 × 10²⁰ He m⁻² at 810 K. This was interpreted as thermally activated diffusion, which apparently is enhanced above 550-650 K, being the temperature of the release stage of He as measured by TDS.

Baldwin et al. and Finlay et al. [9, 10] also studied He retention under mixed D-He plasma and dual beam ion exposures, respectively. Several important results were demonstrated by Finlay et al.[10]. The addition of 3% He in the ion flux reduces D retention at 300 and 500 K for all probed He/D ion range ratios. He retention is affected by ion energy and temperature, but not by He-to-D ratio. The release peak of D, located around 500K, slightly shifts towards lower temperature due to the addition of He in the plasma, which is probably due to the modification of the binding energy profile in the mixed He-D-vacancy clusters. It was also noted, in line with earlier experiments, that the addition of He greatly decreases D diffusion and increases D trapping up to 1 μm depth, similar to the results reported in [2, 11]. Comparison of D and He depth profiles obtained by NRA showed that addition of He apparently reduces D diffusion and enhances its trapping in a region that extends ~1 μm deeper than the He layer detected. This suggests that the stress field, created by the He bubbles [12], extends much further than the layer of high He concentration.

Yajima et al. [13] has performed in-situ transmission electron microscopy (TEM) analysis to explore features of He trapping and release and related them to TDS measurements. Pure W (recrystallized) was exposed to the high flux He plasma to simulate ITER divertor conditions. Three major peaks around 550 K, 800 K and 1400 K were identified by TDS. The TEM measurements done using the same heating ramp reported that the major part of He bubbles was observed directly after exposure, while under the annealing, almost all bubbles disappeared before the temperature of 1200 K was reached. Hence, the release peak around 1400 K could be attributed to the deep diffusion of He and diffusion controlled (accounting for the re-trapping at the sub-surface) release at high temperature.

In this work, we continue our investigation of the effect of plastic deformation on the retention and release of plasma components. In particular, we perform plasma exposure using pure He at temperature of 480 K, and exactly the same W samples, as in our previous works [6-8] done for pure D exposures, to ensure full consistency. The TDS and TEM measurements are provided to make one-to-one comparison between pure D and pure He exposure and clarify the impact of the plastic deformation.

2. Experimental details

Polycrystalline W with purity of 99.99%, provided by Plansee AG was used in this study and in our previous works details on impurities and microstructure could be found [7, 14]. Reference material (REF) represents itself W rod recrystallized at 1873 K for 1 hour. The microstructure of REF sample consisted of randomly oriented grains, separated mainly by high-angle grain boundaries, with a grain size was in the range of 50-150 μm, as shown in Fig. 1(a). The dislocation density is measured by TEM to be (2-4) x 10¹² m⁻², and typical images of the sub-grains and dislocation lines resolved by TEM are shown in Fig. 2 (a,b).

Plastic deformation was applied by performing tensile loading at 873K in air with a deformation rate of 0.2 mm/min to reach 28% deformation, which approximately corresponds to the ultimate tensile strength of the studied W grade at 873 K (after recrystallization). The microstructure of both REF and plastically-deformed (PD) samples was studied using Scanning Electron Microscopy (SEM) by electron back

![Fig. 1. Inverse polar figure map of the EBSD scan of the (a) REF and (b) DEF samples. A sketch in fig. c shows the tensile sample and direction of the loading applied. On the schematic figure on the right hand side, direction A1 shows the deformation axis, A3 – normal to the EBSD scan.](image-url)
scattering diffraction (EBSD) analysis. Examples of EBSD inverse polar maps before and after the deformation are given in Figs. 1(a) and (b), respectively. Grains become elongated in accordance with the orientation of the sample with respect to the tensile loading direction, the mean size reduces to 25-80 µm range. The impact of the plastic deformation on the TEM-visible microstructure is shown in Fig. 2 (c,d).

TEM samples were extracted from the middle of the deformed specimen and TEM samples were prepared in the same conventional way as described in details in our previous works [15-17]. The specimens were investigated by means of a JEOL 3010 TEM operating at 300 kV. It has been revealed that after plastic deformation, dislocations became evident everywhere in the sample’s visible area. The resulting dislocation density in the PD sample was found to be about 2 × 10^{14} m^{-2}, i.e. nearly two orders of magnitude higher than that in the REF sample. The dislocation density was measured by counting the number of intersections with dislocation lines made by random strips drawn on micrographs. To ensure adequate comparison of different sub-surface layers, most of the micrographs are oriented with the primary <111> slip directions pointing to the top of the page. The same method was applied to characterise the dislocation density in the plasma-exposed samples.

Exposures to deuterium plasma were performed at the linear plasma generator Pilot-PSI [18], employing a high-density plasma mimicking the ‘sub-displacement threshold’ plasma-wall interaction conditions expected in the ITER divertor. The energy of the deuterium ions was about 50 eV (controlled by negatively biasing the target), while energies in excess of 900 and 450 eV are required to initiate atomic displacement in W for D and He respectively. Although the plasma beam is non-uniform, the size of the sample corresponded to the full width at half maximum, i.e. 10 mm, which ensured limited temperature and flux gradients across the surface during the exposure, as was measured and confirmed in-situ by an infra-red camera (FLIR A645 sc). The parameters of the exposure are provided in Table 1. The flux was calculated

<table>
<thead>
<tr>
<th>Material (REF/PD)</th>
<th>He / D Flux (ions/ m^{2}/s)</th>
<th>Fluence (ions/m^{2})</th>
<th>Time (sec)</th>
<th>Temperature (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>REF D</td>
<td>1E + 24</td>
<td>4.9E + 26</td>
<td>490</td>
<td>470</td>
</tr>
<tr>
<td>PD D</td>
<td>1E + 24</td>
<td>4.9E + 26</td>
<td>490</td>
<td>470</td>
</tr>
<tr>
<td>REF He</td>
<td>2.1E + 24</td>
<td>6.3E + 26</td>
<td>300</td>
<td>480</td>
</tr>
<tr>
<td>PD He</td>
<td>2.0E + 24</td>
<td>6E + 26</td>
<td>300</td>
<td>480</td>
</tr>
</tbody>
</table>

Table 1
Summary of exposure conditions and parameters.

Fig. 2. TEM dark field images of (a,b) REF and (c,d) PD samples in the non-exposed condition. The images are taken to demonstrate the sub-grain structure (a,c) and appearance of dislocation lines inside the grains (b,d).
from the plasma electron density and electron temperature, as measured by Thomson scattering [19] at a distance of around 25 mm upstream from the plasma-facing surface. Special care was taken to perform all exposures as close as possible to 480 K temperature to remain consistent with our previous studies [6, 7, 20]. Pilot PSI is one of few unique machines which provides an ultra-high flux (up to $10^{24}$ particles/m²/s) and hence high power beam which makes it more difficult to precisely control the surface temperature and some variation can occur due to extremely small differences in sample clamping, sample thickness, plasma conditions, etc. That is why, in the case of He exposures the surface temperature was slightly different from that of D exposures. However, the difference of 10K should not impact the conclusions.

After the exposure, TDS was applied to measure the release of He and D in the tungsten samples. The maximum temperature under TDS cycle was 1273 K, the heating rate used was 0.5 K s$^{-1}$ and the holding time at the maximum temperature was 5 min. The release flux of molecular HD and D$_2$ and He was measured by the quadrupole mass spectrometer (QMS). Quantification of the mass four signal (corresponding to the release of D$_2$ or He) was performed using a calibrated D$_2$ and He leak. The temperature-dependent deuterium release spectra were analysed on the basis of the standard Gaussian distribution function:

$$I \propto \exp\left(-\frac{(T - T_m)^2}{2 \Delta T^2}\right)$$  \hspace{1cm} (1)

where $I$ denotes the peak intensity, $T_m$ is the temperature position of the peak and $\Delta T$ is the full width at half maximum. Each raw TDS curve was fitted using a linear superposition of three Gaussian functions.

3. Results and Discussion

The TDS spectra measured for the REF and PD samples are shown in Fig. 3. One can clearly see that the integral retention after D exposure is enhanced by the plastic deformation, while the effect is opposite in the case of He exposure. In particular, it appears that plastic deformation defects enhance D retention in the TDS spectra after 600 K. The well-pronounced peak around 900K likely corresponds to the desorption of D from voids which is well known to occur in the temperature range of 800–1000 K (see e.g. [21]). In fact, the high temperature peak is also present in the reference material but it is not so highly pronounced in the current exposure conditions. The detailed study of the TDS spectra on the fluence and exposure temperature under pure D plasma exposure is reported in [7, 8].

In the case of He exposure, the reduction of the He retention is observed over the whole TDS temperature range and visually peak positions for the release do not change with the presence of plastic deformation. This observation suggests that the origin of defects in which He atoms are trapped is the same. The dislocation network, induced by the plastic deformation, clearly does not change the mechanism of trapping but affects its intensity. This can be linked to either the nucleation or growth rate of He bubbles being affected by a high density of dislocations. If one assumes that the dislocation network acts as channels for fast diffusion of He in and outside the bulk, the nucleation of He clusters in subsurface area would indeed be suppressed.

The Gaussian fit of the recorded spectra is given in Fig. 4. The symmetric Gaussian fit reasonably reproduces the TDS spectrum, except for a narrow spike around 1150K registered for He exposure in REF sample. It should be noted that such sharp and narrow peaks were also registered in [13] for pure recrystallized tungsten, however, their origin was not discussed. It is reasonable to assume that such a sharp release could be induced by the migration of He bubbles, which in turn leads to an abrupt increase in the release rate, contrary to the more gradual atomic evaporation of He from the He bubbles.

All the spectra could be well fitted with three major release stages, whose characteristics are reported in Table 2. In the case of D exposure, the peaks are located around 600 K, 690-730 K and 840-880 K. In the case of He exposure, the peaks are located around 570 K, 750 K and 1020 K. The release stages after He plasma exposure, performed at 1300 K, are quite close to the results reported previously [13], but in this study the low temperature release stage is found around 330–350 K as reported by Yajima et al. [13]. This deviation might be attributed to the different exposure temperatures employed, which resulted in the formation of fuzz in the case of Yajima’s experiment not observed in our study. The positions of the high temperature peaks, located in the range 800–1200K [10, 13], broadly agree with those observed in the present work.

Following the results presented in Table 2, we can state that indeed plastic deformation does not change the positions of the peaks measured in the He-exposed samples. As a result of the plastic deformation, the intensity of the first release peak decreases by a factor of two, for the second one – by a factor of three and for the third peak – by a factor of two. In the case of D-exposure, the applied plastic deformation mainly affected the intensity of the second and the third release stages.

Finally, we can mention that the release of D is completed within about 1000K, which is in general agreement with many previous studies done after pure D exposure (see introduction for references). Whereas, the release of He is very likely uncompleted, given the maximum TDS temperature applied here (being limited by technical capacity). Indeed, one should expect further release of He above 1300 K, and final conclusions can be done only by performing TDS up to 2000 K.

Based on the measured signals in the available TDS temperature range, the total retention in the REF and PD samples is compared in Fig. 5. The latter shows that plastic deformation suppresses the He retention by a factor of three. Keeping in mind that a fraction of He is very likely still retained, a reasonable explanation for the observed effect of the plastic deformation can be the process of backward diffusion of He.
via dislocation lines, whose density is much higher in the PD sample. The enhanced diffusion via dislocation lines was at least found for hydrogen in W by means of atomistic calculations [22]. Unfortunately the first principle calculations were not performed for He-dislocation system in tungsten, up to our best knowledge. The presence of the dislocation networks may also suppress the self-trapping of He, which otherwise is considered to be the major mechanism leading to the formation of He-vacancy clusters and their further growth in the bubbles by punching self-interstitials in tungsten bulk (see review [23] and references sited therein). Another explanation could be an increased number density of He bubbles (due to enhanced growth of He bubbles on dislocations) near surface that also enhance backward diffusion. In the current experiment, the exposure conditions were too complicated to easily single out the main mechanism responsible for the observed effect.

In our earlier works, we have performed dedicated TEM analysis to investigate the incident exposed surface and sub-surface region to clarify how deep the plasma-induced microstructural modification penetrates the sample in the case of pure D exposure. In [15-17], it was found that the characteristic depth of the plasma-induced microstructural modification is about 10–15 µm only, and beyond that depth, the material recovers its initial microstructure. At this, the main impact of the plasma exposure on the microstructure is the strong increase in the dislocation density on the top surface (by two order of magnitudes), moderate increase within 1-5 µm range and restoration of the initial microstructure beyond 10 µm.

To clarify the impact of He exposure, we performed TEM analysis of the top surface and sub-surface region at a depth of 1-5 µm. The top surface samples were prepared by single-side polishing (i.e. removing material from the un-exposed side), the others by standard double-side polishing. It found that acceptable quality of the TEM analysis could be not delivered for the single-side polished sample due to the strong
roughness of the exposed surface. This is in line with the well-known fact that He bubbles grow very close to the surface and therefore induce blistering and roughening of the surface. Thus, one-to-one comparison for the surface region could not be delivered. The examples of the TEM images in the sub-surface region of D and He exposed samples are provided in Fig. 6. The TEM analysis was performed for both REF and PD samples. For the D exposed sample, we found the increase of the dislocation density up to $(2-4) \times 10^{13} \text{ m}^{-2}$ i.e. by about one order of magnitude as compared to the reference value. In the He-exposed samples, the increase of the dislocation density was much higher, and in the shown presented region the dislocation density reached a value of $(1-2) \times 10^{15} \text{ m}^{-2}$.

The complete TEM study of He-exposed samples is undergoing now. Due to the strong impact of He exposure on the surface roughness, it is impossible to apply the same methodology as was applied for pure D exposures in our earlier works. Thus, the preparation of the lamellas with the focus ion beam (FIB) is needed. To clarify the extension of the plasma-induced damage at a depth beyond 5 µm, we used FIB to fabricate the TEM lamella and make SEM scan provided in Fig. 7. Although the surface quality still requires improvement, it can be seen that grain refinement is evident in the sub-surface region with a depth of 1 µm or even less. This points to the fact that the top surface region is indeed heavily deformed (pattern of small sub-grains) while the region within next 10 µm is nearly free of the grain refinement. Thus, He exposure affects the material microstructure much stronger than D exposure, at least within the range of 5 µm from the surface.

4. Conclusions

We have performed a set of pure D and He high flux exposures at 470–480 K. The exposures were performed on the samples made of ITER specification tungsten provided by Plansee AG in the recrystallized state and plastically deformed condition (28% deformation at 873 K). TDS and preliminary TEM measurements were performed to
compare the effect of plastic deformation on the retention and release in the case of D and He exposure conditions. The release was measured by TDS up to 1300 K.

On the basis of the results presented and discussed above, we can summarize the following observations:

(i) In plastically deformed tungsten, three major release stages for He are observed just as in the reference recrystallized material. The positions of the peaks are the same in both types of samples but their amplitudes differ depending on the dislocation density of the material. The positions of the release peaks are in good agreement with the results previously published for pure He exposures. Overall, the plastic deformation reduces the total retention of He by a factor of three following the TDS measurements done up to 1300 K.

(ii) Preliminary TEM characterization performed for REF samples showed that in the sub-surface region He exposure induces much severe increase of the dislocation density as compared to the D exposure. Whereas the depth at which the microstructure recovers to the original bulk pattern is similar in both He and D exposed samples, and this depth is about 15-20 µm.

Further study by applying high temperature TDS is needed to substantiate the conclusions regarding the outgassing of He. Earlier studies show clearly that He release may last up to 1600-1800 K. More in-depth TEM investigation is also needed before and after TDS to distinguish clearly the impact of exposure itself and possible annealing induced by the TDS cycle.

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Supplementary materials

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