Deuterium retention in radiation damaged tungsten exposed to high-flux plasma

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9 Conclusions and outlook

This thesis presents the results of deuterium retention experiments on pre-irradiation damaged tungsten exposed to high-flux ($\sim 10^{24} \text{ m}^{-2} \text{s}^{-1}$) deuterium plasmas. Chapters 4 and 5 describe the effect that pre-irradiation with high energy $W^{4+}$ ions has on the trapping of deuterium. The penetration of deuterium into the pre-irradiation damaged material under high-flux plasma exposure is described in chapter 6. Chapter 7 discusses surface modifications created during high-flux plasma exposure, and its effect on deuterium retention. In addition, the effect of pre-irradiation damage on surface modifications was studied. Finally, the deuterium retention in $\mu$m thick tungsten films with varying density and nanostructure was investigated in chapter 8.

In this chapter we gather all the results from previous chapters to discuss the progress made addressing the main research question:

What is the effect of pre-irradiation damage on the deuterium retention in tungsten under high-flux plasma exposure?

As the underlying motivation for this research is the applicability of tungsten in the divertor of ITER, the implications of our findings towards understanding tritium retention under neutron irradiation in the ITER divertor and suggestions for continuation in this area are described in the outlook (section 9.2).

9.1 Conclusions

9.1.1 Main results

In chapter 4 it was shown that the deuterium retention in pre-irradiated targets after high-flux plasma exposure at surface temperatures below 525 K saturates at a concentration of 1.4 at.%. This saturation occurs at a pre-irradiation damage level of 0.2 dpa. The saturation originates in the ion damaging mechanism and was not affected by the high plasma flux. At 0.2 dpa, the concentration of vacancies and interstitials is at such a high level that every newly created vacancy or interstitial automatically recombines with one already present in the material. The average saturated volume per incident MeV ion is $3 \times 10^4 \text{ nm}^3$. Saturation still occurs at a damage level of 0.2 dpa after plasma exposure at higher surface temperatures (above 800 K), although the absolute level of deuterium retention strongly decreased (chapter 5). This reduction is caused by the mobility of vacancies which results in their annealing and clustering (appendix 5.A). A low occupation level of the defects caused by increased deuterium mobility was found not to be relevant.

The penetration of deuterium into pre-irradiated tungsten was measured to be very low at self-biasing conditions and only a fraction of $10^{-5} - 10^{-7}$ of deuterium from the incoming plasma beam is retained within the material (chapter 6). To explain this low penetration, we propose a mechanism in which the deuterium atoms form a chemisorbed layer at the surface. Incoming deuterium ions ($\sim 5 \text{ eV}$) therefore can not directly enter the
material, but interact instead with this deuterium surface layer. The energy is effectively transferred from incoming ions to atoms at the surface, preventing the ions to directly penetrate. Actually, an additional thermal process is needed to allow chemisorbed deuterium to enter the tungsten.

Experiments with target biasing during plasma exposure showed various type of surface modifications as discussed in chapter [7]. We distinguished three types of surface modifications: blisters originating from inter-granular cavities, protrusions arising from intra-granular cavities and nanometer structure formation. The micrometer-sized blisters grow in size and quantity with exposure time. We found less formation of these blisters on pre-irradiation damaged targets. Protrusions, typically hundreds of nanometer in size, were only observed after exposure to high-flux plasmas. The driving mechanism of blisters and protrusions is therefore most probably super-saturation within the tungsten lattice that leads to cavity growth and plastic deformation of tungsten.

Finally, micrometer thick tungsten layers with different structures (density and crystallite size) were exposed to deuterium plasmas. The thin tungsten films show a considerably higher retained fraction than the bulk polycrystalline tungsten, even when compared to pre-damaged tungsten samples. In fact, the lower the density of the thin tungsten film, the higher the deuterium retention and the lower the temperatures at which desorption takes place.

### 9.1.2 Undamaged tungsten

In order to isolate the effect of radiation damage, we first need to understand deuterium retention in tungsten under plasma exposure only. Figure 9.1 shows the retained deuterium fraction as function of incoming plasma fluence. Here, all data discussed in this thesis without pre-damage have been grouped. The fluence represents the average incoming deuterium ions per surface area and is calculated by integration of the flux profile (equation [2.1]) over the surface multiplied by the plasma exposure time and divided by the surface area. The retained fraction is defined as the deuterium stored in the sample, measured by TDS, divided by the total incoming deuterium ion fluence. Note that these samples differ in exposure conditions, such as bias, surface temperature and surface condition (as-received or mirror polished). Also, the time between plasma exposure and TDS measurement varied between two weeks and a few months. The bulk of the experiments was performed at low surface temperature, which means that the surface temperature in the centre is at maximum 560 K. The two experiments that were carried out at high surface temperature are indicated by a grey filling of the data points.

Targets exposed at low temperature and at self-biased conditions are shown by orange data points. Targets that had a rough surface (as-received) are indicated with a square and mirror polished samples with circles. The orange line is drawn to guide the eye. The slope of this line is chosen to be proportional to $\sqrt{t}$, indicating diffusion behaviour. Note that the retained fraction of all self-biased targets is very low and only $\lesssim 10^{-7}$ of the deuterium ions in the plasma beam. As self-biased samples showed no surface modifications, the deuterium appears to be retained in intrinsic defects, which are homogeneously distributed throughout the material ($10^{-4} - 10^{-6}$ atomic fraction).
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Figure 9.1: Deuterium retained fraction of undamaged samples plotted versus the plasma fluence. Self-biased exposures are shown in orange: Ch.3 represent the samples from section 3.4.2, Ch.4 the undamaged sample from chapter 4, Ch.6 II is exposed during the measurement series of chapter 6, and Ch.5 the undamaged sample exposed to high surface temperature (chapter 5). The biased (−40 V) samples are shown in blue: Ch.3 are the samples from section 3.4.2, Ch.7 represents the undamaged samples from chapter 7. Additionally, a sample that was exposed to high temperature is shown (carried out during the measurement series of Ch.7).

The targets that were exposed at a bias of −40 V are plotted in blue. Again, a line to guide the eye is drawn to indicate diffusion scaling. Although the retained deuterium has increased with an order of magnitude with respect to the self-biased targets, it is still only a small fraction with respect to the incoming plasma beam. As discussed in chapter 7, deuterium is predominantly retained in defects created by plasma exposure induced damage, such as blisters and protrusions that originate from inter-granular and intra-granular cavities, respectively. The modifications and the trapped deuterium are present well beyond the implantation region and extend micrometers into the tungsten material.

The self-biased sample exposed at high surface temperature contains a higher retained fraction than the sample exposed at low surface temperature, mainly because more deuterium could diffuse into the tungsten. On the contrary, the biased high temperature sample retained less than the low temperature counterpart. No blister formation was found on this sample, which is in agreement with fewer traps being available for the deuterium.
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9.1.3 Radiation damaged tungsten

In chapter 4 it was shown that the saturation of pre-irradiation damage is reached at 0.2 dpa. Figure 9.2 shows all targets that were pre-irradiated to a damage level of 0.2 dpa or higher. The as-received samples with rough surfaces are indicated by diamonds, while triangles represent all mirror polished samples. It must be noted, that the rough samples were pre-damaged over a smaller radius ($r_{\text{dam}} = 6 \text{ mm}$) than the plasma exposure ($r_{\text{plasma}} = 8 \text{ mm}$). For the mirror polished samples $r_{\text{dam}}$ was 9 mm.

The self-biased samples are shown in red (mirror polished) and purple (as-received). It seems that the as-received targets have somewhat higher deuterium retention. The red data points follow a $\sqrt{t}$ behaviour as described in chapter 6. Note that the TDS results are an average over the whole sample surface and therefore yield lower values than the locally retained fraction in the centre (NRA results in figure 6.3).

Pre-damaged targets were also exposed to a high-flux plasma under biased conditions. The biased targets (–40 V) are plotted in green in figure 9.2. Compared to the polished targets at unbiased conditions, a higher deuterium fraction is implanted. In this way the total retained fraction increases with about an order of magnitude. The overall retention level however, remains again low and is around $10^{-5} - 10^{-6}$.

In conclusion, the retained fraction is very low under high-flux plasma exposure.

![radiation damaged tungsten](image)

Figure 9.2: Deuterium retention of samples pre-irradiated to a damage level of >0.2 dpa. Ch.4 represents the samples from chapter 4, Ch.4 II are samples from a follow-up measurement series, Ch.5A is the sample from appendix 5.A, Ch.6 corresponds to the measurement series of chapter 6 and Ch.6 II to a follow-up experiment under similar conditions. The green data points represent the pre-irradiated targets exposed at –40 V.
Direct extrapolation of figure 9.2 gives a rough estimation for the tritium retention in the ITER divertor at the end of its lifetime (i.e. after $2 \times 10^7$ s), which corresponds to a fluence of $2 \times 10^{31}$ m$^{-2}$. Predictions for the divertor end-of-life damage level is 0.6 dpa, this is above the saturation of the pre-irradiation damage of 0.2 dpa. The worst case scenario is represented by the green line that corresponds to the retention in both pre-irradiation damaged and plasma damaged targets. The retained fraction at the fluence of $2 \times 10^{31}$ m$^{-2}$ is only $1.1 \times 10^{-8}$, or in absolute terms $2.2 \times 10^{23}$ m$^{-2}$. In the case that we assume that deuterium and tritium are equally distributed in the $\sim 50$ m$^2$ divertor, this corresponds to $\sim 30$ g tritium. A divertor operating at $<560$ K retains only 4% of the allowed tritium in-vessel inventory.

9.2 Outlook

Although the results show low levels of retention, care should be taken to extrapolate these results over four orders of magnitude to the relevant regime for ITER. It is possible that other effects will occur at long exposure times. Therefore, it is important that future experiments focus on accessing the high-fluence regime. The superconducting magnet upgrade that is planned for Magnum-PSI will give the opportunity to explore this fluence region. The second note is that this work solely focusses on the deuterium retention properties of tungsten pre-damaged with high energy tungsten ions. Neither the change of mechanical properties nor secondary effects of neutron irradiation damage, such as embrittlement, swelling and transmutations are considered. These are of course relevant aspects for a working divertor. Another difference between the extrapolation and the ITER divertor is the material temperature. Most of this thesis was carried out at low temperature, i.e. $<550$ K, while only a few experiments were performed at temperatures above the vacancy mobility of tungsten. Nevertheless, we have observed a reduced deuterium inventory as a result of the annealing of vacancies at high temperature. However, at the same time the diffusion speed increases with temperature. Therefore, it is of interest to study the effect of temperature on deuterium penetration in more detail. One could think of monitoring diffusion during plasma exposure at elevated surface temperatures, MeV ion pre-irradiations at elevated temperatures and/or interchanging the order of MeV irradiation, heating and plasma exposure.

Another interesting research line would build further on the experiments reported in chapter 8 and concerns the fundamental process of diffusion. In this thesis, we assumed for the diffusion coefficient in the TMAP7 simulations a perfect tungsten lattice. However, in the experiments we used polycrystalline materials that contain grain boundaries. Grain boundaries might act as a diffusion barrier, thereby slowing down diffusion, or act as ‘high ways’ and increase the diffusion. Nanostructured layers with varying density would provide an interesting tool to investigate the diffusion process in tungsten.

The last concern is that blister formation was found to occur beyond the implantation region. Although we believe that blister formation happens by plastic deformation, the exact mechanism is not yet known. Future studies will need to focus on the process of blister formation. One of the key mechanisms that remains unknown is the interstitial or
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mobile concentration during plasma exposure. The determination of the in-situ mobile concentration would greatly help to understand how much pressure is built up in the tungsten lattice and cavities as well as to understand how cavity growth takes place.

Our results suggest that the tritium retention in the divertor of ITER will not be problematic. Nevertheless, still many challenges exists for the use of tungsten in the divertor of ITER.