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(18th May 2015 – 22nd May 2015)
Aix-en-Provence, France

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Mechanical and microstructural changes in tungsten due to irradiation damage

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Abstract. Stress-relieved pure tungsten received three damage levels (0.1, 0.25 and 0.5dpa) by self-tungsten ion beam irradiation at room temperature. Positron annihilation spectroscopy showed the formation of mono-vacancies and vacancy clusters after ion beam exposure. In the first irradiation step (0-0.1dpa) some splitting up of large vacancy clusters occurred which became more numerous. For increasing dose to 0.25dpa, growth of the vacancy clusters was seen. At 0.5dpa a change in the defect formation seems to occur leading to a saturation in the lifetime signal obtained from the positrons. Nano-indentation on the cross-sections showed a flat damage depth distribution profile. The nano-indentation hardness increased for increasing damage dose without any saturation up to 0.5dpa. This means that other defects such as dislocation loops and large sized voids seem to contribute.

Keywords: tungsten, ion irradiation, nano-indentation, positron annihilation spectroscopy

1. Introduction

Tungsten is one of the most promising options as the plasma facing material for a nuclear fusion reactor. Tungsten materials have the highest melting point, good thermal conductivity and low tritium retention. However, one of the drawbacks is the limited temperature window in which they can be operated. The lower temperatures are restricted by the ductile to brittle transition temperature (typical value) while the recrystallization temperature gives a limit for the higher temperatures (1200°C). As a plasma facing material, tungsten will be subjected to severe heat, particle and neutron loads [1]. Neutron irradiation leads to activation and to changes in the mechanical properties of the material. Reviews of the effects relevant for fusion applications are given in [2, 3]. The ITER design foresees a 0.3 MWa/m² ($\sim 2 \times 10^{21}$ n/cm²) fluency for the tungsten armor used in the divertor region [4]. This is equivalent to a damage of 0.30 to 0.50 dpa (displacements per atom) [3]. It is well known that the neutron irradiation leads to a displacement damage of the lattice structure creating vacancies and interstitials, and to the generation of transmutation products.

In order to expand the knowledge on the irradiation damage, the mechanical and microstructural properties of a pure tungsten product were investigated. The present work investigates the self-ion irradiation response of pure tungsten for dose levels from 0 up to 0.50 dpa at room temperature. The effect of self-ion implantation on the nano-indentation hardness of the materials is related to the changes in the microstructure and is investigated in detail by using positron annihilation spectroscopy. Similar investigations with positron annihilation spectroscopy, transmission electron microscopy and hardness are conducted at this moment on neutron irradiated materials in order to compare with this study. The final aim is to see if similar defects are created by self-ion damage and neutron irradiation and if not, what the main differences and especially implications are.

2. Material and testing procedure

Stress relieved pure tungsten samples from a hammered rod from Plansee with the surface perpendicular to the deformation direction were used. The surfaces were prepared in a similar way by grinding with SiC

paper and final polishing with diamond paste. Self-tungsten ion beam irradiation with 20MeV W^{6+} ions was performed at the IPP tandem laboratory setup TOF at room temperature. The average damage rate was around 1×10^{-4} dpa/s. The estimated maximum depth of the damaged layer is only $\sim 2 \mu\text{m}$ deep and the mean depth of the maximum damage is at $\sim 1.4 \mu\text{m}$ as predicted by SRIM2013 [5] (stopping range of ions in matter) for a displacement energy E_{disp} of 90eV [6] in tungsten. Conventional mechanical tests could not be used for such thin damage layers. Therefore, nano-indentation has been used to investigate the change of the hardness values of three damage levels (0.10dpa, 0.25dpa and 0.50dpa). For each measurement, a pre-calibration on a Si (fused Silica) target to determine the indenter area-function calibration was performed. Because of the hardness of tungsten, a Berkovich Diamond indenter was chosen. Since the exposed material depth was limited, the CMS (continuous stiffness) mode has been chosen with a strain rate at the target of 0.05s^{-1} and a harmonic frequency of 45Hz. Positron annihilation spectroscopy (PAS) gave some insight on the development and evolution of the microstructure, more specifically related to the vacancies, clusters and the dislocation and vacancy loops. The PAS experiments are performed with the Coincidence Doppler Broadening (CDB) and Positron Annihilation Lifetime Spectrometers (PALS). The CDB spectra are measured using two Ge detectors (for more details [7]). The CDB spectrum provides the momentum distribution of core electrons, which can be used to determine the chemical environment of positron-electron annihilation site. The higher the S-parameter, the higher the defect size and/or number density because the S-parameter is very sensitive to open volume defects. The W-parameter corresponds to annihilation of the positrons at the core electrons and gives insight in the chemical surrounding at the annihilation site. The PALS measurements, which provide the size and density of the vacancy type defects, are performed with a spectrometer working in a triple coincidence mode [8]. Lifetime measurements gives much more detailed results on the type of defects such as mono-vacancies, dislocation and/or vacancy loops or vacancy clusters.

3. Results

3.1. Positron annihilation spectroscopy

Positron annihilation spectroscopy is a very powerful tool to investigate small sized defects such as vacancies. The S-parameter corresponds to positron annihilation with the valence electrons, while the W-parameter is related to annihilation of the positrons with the core electrons. This ensures that the S-parameter is sensitive to open volume defects and the W-parameter to the chemical surrounding at the annihilation site. In table 1, a summary of the S- and W- parameters is given for all the samples. The reference sample underwent similar surface treatment procedures (related to grinding and polishing) as the other three samples that underwent ion beam exposures (0.10, 0.25 and 0.50dpa) at room temperature. In general for one type of material (in this case tungsten), a higher S-parameter means a decrease in the W-parameter. This is fulfilled for all measured samples. The self-damage induced by the self-tungsten ions gives a clear increase in the S-parameter which was expected since more and/or larger vacancy clusters are created. The damage created after only 0.10dpa is very large as compared to the reference sample, while the additional damage created when going from 0.10dpa to 0.25dpa is much smaller. In addition some saturation effect was found when the sample was further damaged up to 0.50dpa. This can mean several things. First of all, there is a limit in what the positrons in tungsten (or any other material) may observe. If the vacancy clusters become too large (~37 vacancies [9]), the positron cannot “see” the other side of the cluster and therefore it saturates even when the clusters are still growing. In addition, for CDB (Coincidence Doppler Broadening), there is an interplay of two things. Namely the amount (density) of vacancies and the size of the vacancy clusters. When clusters become larger than what the positrons are able to measure and at the same time the density remains constant or decreases due to clustering, it may seem that the S-parameter is saturating or even decreases.

In figure 1, the S-W parameter plot is shown for the reference and ion beam damaged samples. The reference material is shown at the top left of the graph. A red dotted line is drawn to guide the eye showing the clear effect of the ion beam damage. It is clearly visible in the plot that most of the damage creation occurs within the first 0.10dpa. The increase from 0.10dpa towards 0.25dpa is much less. Even a decrease was observed for the 0.50dpa damaged material, where the S-W parameters were within the

values of 0.10 and 0.25dpa. It is apparent that all values are following one S-W parameter line indicating that no special vacancy solute clusters are created in addition, which was of course expected for pure tungsten.

The PALS technique is less accurate (error is much larger) than the CDB one and takes a longer time to measure but the resolution is much better. It gives thus more detailed information on the evolution of the density and size of the vacancy clusters and the type of defects that are present. The lifetime results of the reference sample and the ion beam damaged samples can be seen in figure 2. The mean average lifetime is after subtraction of the source and resolution function of the PALS system. It can be seen that the average mean lifetime of the reference sample is $\tau_{\text{avg}} = 166.3\text{ps}$. The average lifetime is much higher than the bulk value of 105ps [10, 11] and the 150ps value for dislocation/vacancy loops [12] in tungsten. But is lower than the typical 180ps value for mono-vacancies [13]. Detailed analysis by de-convolution of the spectra gives results for the “short” lifetime value $\tau_1 = 138.8\text{ps}$ for the reference sample. This is clearly a mixture of the bulk and the dislocation/vacancy loop components. While the second “large” lifetime component τ_2 is related to vacancy clusters and voids ($\tau_2 = 486.1\text{ps}$). The intensity of the second component I_2 gives an idea on the density (number) of the vacancy clusters. After ion beam irradiation, the average lifetime increased significantly, in line with the increase of the S-parameter. In addition, not only the average lifetime increases, but also the “short” lifetime τ_1 gradually increased. The size of the clusters (lifetime τ_2) seemed to first decrease after ion beam irradiation of 0.1dpa, but the density increased (increase in I_2 intensity). Additional damage (from 0.1 to 0.25dpa) gave again an increase in the large cluster size, less in the density increase. So it seems that ion beam irradiation is destroying some large clusters in more numerous smaller clusters which grow again when increasing damage from 0.10dpa up to 0.25dpa. When increasing the dose even further to 0.50dpa, it was found that the average lifetime decreases slightly down to values in between the 0.10 and 0.25dpa values. This is in perfect agreement with the CDB results. After decomposition it is clear that some saturation was found for the “short” lifetime value at 0.50dpa which remains at $\tau_1 = 151\text{ps}$, while the larger vacancy clusters decrease in size to a lifetime of $\tau_2 = 435\text{ps}$. The

intensity I_2 (so density) of this second component after 0.50dpa decreased below values after 0.10dpa damage.

These results can mean several things. First of all, the large component τ_2 is close to saturation, if clusters become much larger they will not be measured as larger. If thus many smaller sized clusters are formed at 0.50dpa, it will seemingly reduce the lifetime of the second component τ_2 . In addition if the intensity of the first short component becomes larger, the intensity of the second component I_2 will seemingly decrease. And finally, the drop in the first short component τ_1 after 0.50dpa does not necessarily mean that less mono-vacancies are formed, but could be due to the fact that the dislocation/vacancy loops become much more important. Since this lifetime is below the one for mono-vacancies, a transformation of vacancies towards loops will give a reduction in the first lifetime component τ_1 while increasing the intensity I_1 . Further investigations should be conducted with Transmission Electron Microscopy (TEM) to determine the contribution of large size voids and loops to make more accurate conclusions.

Previous studies have shown that self-ion irradiation damage strongly enhances deuterium retention [14, 15, 16, 17, 18]. Tyburska et al [14] noticed that a linear increase of damage did not lead to a linear increase of deuterium stored in the sample but that saturation was observed at 0.30dpa independent of the implantation fluence. Similar experiments at much higher fluxes, showed already saturation of the deuterium retention at 0.22dpa [16, 18]. According to 't Hoen [18], the deuterium trapping is solely related to the self-ion tungsten irradiation damage. This would mean that indeed the deuterium is mainly stored at the vacancies and clusters when self-ion damaging is applied. In addition, the saturation seen in the PAS results is very similar to the deuterium retention saturation. This means that the change of defect types between 0.25 and 0.50 dpa leads to different and less efficient trapping of deuterium, thus leading to the saturation.

3.2. Nano-indentation

Nano-indentation was performed on both the top surface (figure 3a) as well as on the cross-sections embedded under a very shallow angle to obtain information on the damage depth distribution (figure 3b). Indentation was performed up to a maximum of 300nm depth, where the average values were taken in between 150 and 200nm to avoid the influence from the surface (roughening, oxide layer, ISE indentation size effect, ...). The contact depth has been calculated from the stiffness, the normal load and the indentation depth following the classical method proposed by Oliver&Pharr [19]. In figure 3a, the averaged nano-indentation hardness for the reference and measured over the ion beam loaded area is shown. For materials with a high modulus over yield strength ratio such as strain-hardened metals, pile-up behavior is expected. Hence, the real contact area is underestimated, which leads to an overestimation of the modulus and the hardness. Normally ion beam irradiation removes the pile-up behaviors. This can explain why the reference material seems much harder than the ion beam damaged samples. Further analysis to include the real contact area and to correct the nano-indentation hardness with the Gao and Nix method are underway but are outside the scope of this paper. Secondly some averaging was done over the ion beam loaded area, so probably some underestimation will be present for the ion beam irradiated samples. Because in the case that there is a damage depth profile, the error might change with indentation depth. The small arrows shows the expected trend related to the hardness values that more detailed analysis will induce. Anyway, irrespective of the absolute numbers, the trend remains clear, namely that there is a hardening for increasing dose level, so without any saturation as was found in the PALS and CDB measurements. This means that other defects such as (mainly) dislocation loops and large-sized voids seem to contribute significantly for the 0.50dpa case. In addition, a clear depth profile was found for the ion beam damaged samples. In figure 3b, an example of the cross-section profile of the highest damage dose at 0.50dpa is shown. The hardening profile from the surface (~4 μ m) into the depth (~15 μ m) seems to be much flatter than the dose damage profile. Taking into account the angle of embedding and polishing of the sample (see inset in figure 3b), the depth within the cross-section of 15 μ m agrees to the ~2 μ m maximum damage depth as was calculated with the SRIM2013 code with a displacement energy of 90eV in tungsten.

4. Discussions and conclusions

A very good correlation between the positron lifetime and Doppler Broadening parameters was seen. It was found that self-ion beam irradiation showed the formation of mono-vacancies and vacancy clusters. In the first irradiation step (0-0.10dpa) most of the damage occurred with some splitting up of large vacancy clusters that increased in density. By increasing the dose up to 0.25dpa, the clusters started to grow. At 0.50dpa it seems that a change in defect formation occurs apparently leading to a saturation in the lifetime signal obtained from the positrons. This PALS defect saturation is in agreement with the deuterium retention saturation seen in several studies. It can thus be concluded that the deuterium was mainly trapped in those studies at vacancies and clusters and that the loops and large sized voids did not induce additional trapping sites. Nano-indentation was performed on both the top surface as well as the cross-sections to obtain information on the damage depth distribution. Some pile-up occurred for the undamaged reference material that disappeared after ion beam irradiation of 0.10dpa and higher. This led to an overestimation of the reference hardness. A clear hardening depth profile was found, which was flatter than the damage dose profile. In addition, the hardness increased for increasing dose without any saturation. This means that other defects such as dislocation loops and/or large sized voids seem to contribute for the 0.50dpa case. From both the PALS as nano-indentation results it looks that the decrease of the vacancy induced damage is due to restructuring of those defects in other forms (types) of defects where the positrons are less sensitive for such as dislocation/vacancy loops and/or very large sized voids. In future more work will be conducted on estimates and calculations to determine the trapping rate (ns^{-1}) of the defects and estimated number of vacancies (ppm). In addition TEM analysis will be conducted to obtain information on the formed dislocations, voids and loops.

At this moment measurements are underway to characterize the defect damage created by neutron irradiation in tungsten. Similar studies as for the self-ion damage will be conducted in order to compare the defect creation between self-ion and neutron damage. It will give insight if similar defects are created and what the possible implications (on hardening, ...) can be.

Acknowledgements

This work was executed under EUROfusion **WP PFC**. This work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 under grant agreement No **633053**. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

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Table 1: S- and W-parameter for the reference and ion beam damaged materials at room temperature for different dose levels and Tungsten fluences. (Error S-parameter: ± 0.001)(Error W-parameter ± 0.0001)

Dose level [dpa]	Tungsten fluence [W/cm^2]	S-parameter	W-parameter
Reference sample	-	0.384	0.0085
0.10	1.78×10^{13}	0.393	0.0076
0.25	3.94×10^{13}	0.396	0.0074
0.50	7.87×10^{13}	0.394	0.0075

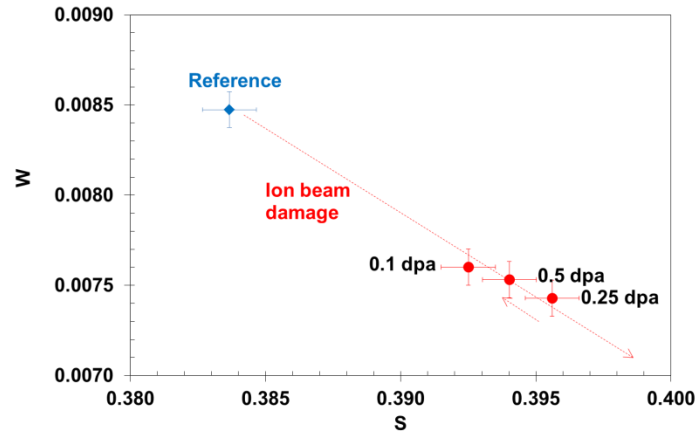


Figure 1: S-W parameter plot of the reference (blue diamond) and ion beam irradiated samples at RT (red circles). Red dotted arrows is added to guide the eye.

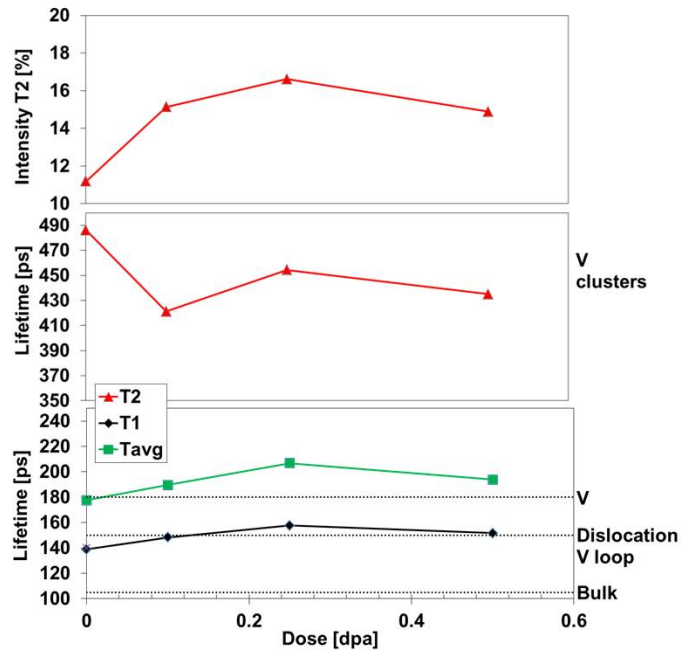


Figure 2: PALS lifetime values as a function of damage level dose (dpa) (green squares: mean lifetime τ_{avg} , black diamonds: short lifetime component τ_1 , red triangles: large lifetime component τ_2 and intensity I_2).

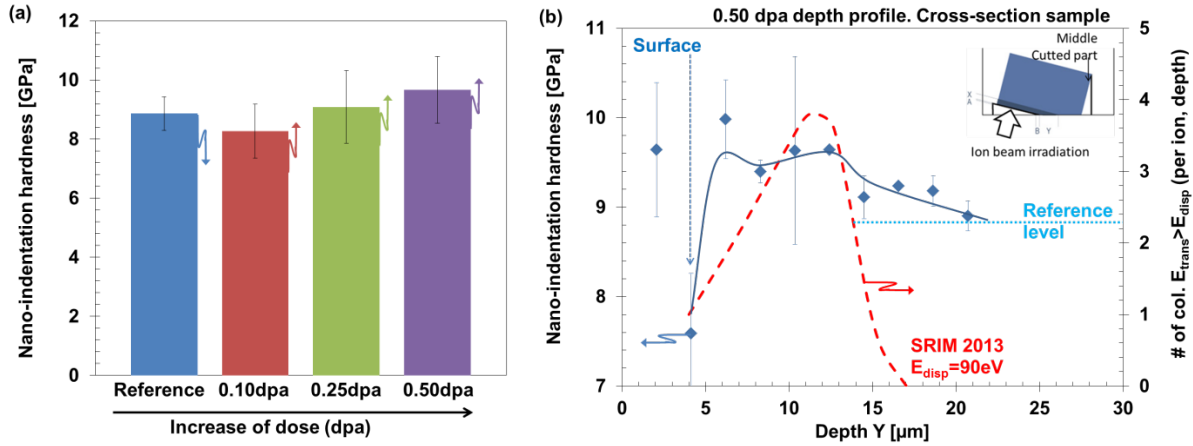


Figure 3: (a) Averaged nano-indentation hardness (GPa) for the reference and the ion beam irradiated tungsten samples, (b) Nano-indentation hardness values (blue diamonds) in depth on the cross-section sample after 0.50 dpa damage dose. Sample is embedded under a shallow angle (see inset) with the calculated dose profile from SRIM 2013 (red striped line) for a displacement energy of 90 eV in tungsten recalculated for the depth Y. The blue full line going through the data-points is added to guide the eye for the nano-indentation results and the light blue dotted line shows the reference level without any ion beam damaging.