EVALUATION OF TUNGSTEN AS DIVERTOR PLASMA-FACING MATERIAL: RESULTS FROM ION IRRADIATION EXPERIMENTS AND COMPUTER SIMULATIONS

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Abstract

The effect of Primary Knock-on Atom (PKA) spectrum on the damage creation and subsequent deuterium trapping at the defects has been investigated using computer simulations and surrogate ion-irradiation experiments. The PKA spectrum generated by light ions resembles close to 14 MeV neutrons. Ion-irradiation experiments have been carried out with ions of Au, W, He and D of energies ranging from 100 keV – 80 MeV for a fluence range of $1.3 \times 10^{14}$ ions-cm$^{-2}$ to $5 \times 10^{17}$ ions-cm$^{-2}$. While 80 MeV gold ions produced dense clusters of defects and dislocation loops, boron produced predominantly dislocation lines and dislocation loops. Molecular dynamics simulations show that at large PKA energies (>150 keV) the fragmentation of the cascade takes place which tend to limit the size of individual defect cluster. Transmission electron microscopy studies have shown that 80 MeV Au-irradiation which has significant fraction of PKA energies above fragmentation threshold has produced dense cluster of smaller defects (<12 nm). Deuterium irradiation experiments indicate that the defect produced during the irradiation might act as channels for diffusion leading to a significant shift in the implantation profiles.

Key words: PKA Spectrum, Fragmentation, Transmission Electron Microscopy, Positron Annihilation Spectroscopy

1. INTRODUCTION

Emergence of tungsten as a candidate plasma-facing material for the divertor in fusion devices makes it necessary to evaluate it from the point of view of neutron irradiation and its impact [1,2]. One of the unresolved issues in using tungsten is the difficulty in estimation of H-isotope trapping at the defects. This requires knowledge of various types of defects which are mainly determined by the Primary Knock-on Atom (PKA) spectrum and associated cascades created by fusion neutrons within the tungsten lattice [3].

Since the facilities for creating damage due to reactor-like fluxes of neutron (along with plasma & alpha particles) do not exist, one has to resort to indirect means like surrogate ion-irradiation. However, there will be key differences in PKA spectra, energy-loss and heating mechanisms and the rate of production of defects. Therefore, there is a need to understand how these differences impact the defect structure and conclusions on retention. In this paper, we address the effect of PKA spectra on the defects by using low and high mass ion irradiation experiments and computer simulations.
A reasonable estimate of neutron spectrum in a fusion reactor can be established with neutronic calculations by using ITER as an example for a future fusion-reactor. The PKA spectrum and the corresponding displacement per atom (dpa) due to the neutron flux are calculated at tungsten divertor by taking into account the neutron nuclear reaction cross-section. The PKA spectrum generated by different ions have been calculated using binary collision models and the experiments have been carried out using ions with varying mass and energy. The defect structure obtained due to neutron irradiation is simulated using MD simulations and compared with those obtained in surrogate ion experiments. The difference in the neutron and surrogate ion PKA and the consequences in the defect structure are discussed in this paper. The simulations and experimental methods are discussed in section 2. The results from both simulations and experiments and their analysis and extrapolation are given in Section 3. The conclusions of the present study are given in Section 4.

2. SIMULATIONS AND EXPERIMENTS

Neutronic calculations have been carried out to estimate the neutron flux at tungsten divertor in ITER using ATTILA simulation code [4] with ITER-B-lite geometry [5]. The divertor structure is divided into various elements and the detailed neutron flux is estimated at various mesh-points for a standard ITER shot of 500 MW power for 400 s duration [6]. The corresponding PKA spectrum at each mesh point is calculated using SPECTER [7] which takes into account cross-sections for various interactions between neutron and tungsten atoms that leads to a displacement of tungsten atom. The threshold displacement energy (E_d) for tungsten atom is chosen to be 90 eV as recommended by the American Society for Testing and Materials (ASTM). The dpa at each mesh is calculated using the Norgett-Robinson-Torrens (NRT) model [8].

Molecular Dynamics simulations have been carried out to study the collision cascades and the subsequent defect structure for an energy range of 500 eV to 215 keV using Parallel Cascade (ParCAS) code [9]. The simulations were carried out on single crystal tungsten samples with X-Y plane oriented along (110) with 3D periodic boundary conditions. A temperature scaling was enabled at the boundaries of the simulation cell for 4 angstroms along each direction to mimic the condition where the system is in contact with the surroundings at 300 K. No temperature scaling was used in the centre of the simulation cell. At least 50 non-cumulative bombardment studies were performed for each PKA energy. Depending on the PKA energy the number of atoms in the simulation cell varied from 0.5 to 35 million atoms and they were carried out on a 35 TF, 756 core HPC machines at IPR, Gandhinagar. In order to create a variety of PKA spectra to study its influence in damage creation experimentally, binary collision simulation have been carried out using SRIM [10] simulation package. Ions of different mass (H to Au) and energy (1 MeV to 400 MeV) are used to generate a PKA spectrum comparable to that of neutron spectrum by combination of ions.

Irradiation experiments have been carried out on tungsten foil samples of 8 x 8 x 0.1 mm^3 size recrystallized at 1838 K under 10^-3 mbar pressure with 100 mbar Ar+8% H2 gas for about one hour. These foils were irradiated with energetic ions of 100 keV D^+ (14 MeV neutron generator, IPR, Gandhinagar), 250 keV He^+ (Low Energy Ion Beam Facility, IUAC Delhi), 10 MeV B^3+ (3 MeV tandem accelerator, GGU, Bilaspur) and 80 MeV Au^7+15 MV pelletron facility, IUAC Delhi) for fluence values of 1.3 x 10^{14} ions-cm^{-2} (B^3+, Au^7+), 1 x 10^{15} ions-cm^{-2} (B^3+), 5 x 10^{15} ions-cm^{-2} (250 keV He^+) and 5 x 10^{17} ions-cm^{-2} (100 keV D^+). The corresponding dpa varied between 0.001 (1.3x10^{14} B^{3+}) to 0.85 (5 x 10^{17} D^+) for E_d = 90 eV. In order to study the effect of pre-damage and presence of helium in deuterium, D+ irradiation was carried out on samples that are pre-irradiated with B, Au and He and Au+He.

The microstructure and the multiple scales of defects that are formed in the irradiated samples are studied using XRD, low temperature resistivity measurements, microscopy techniques (FESEM, TEM) and positron annihilation lifetime measurements (light time measurements and Doppler broadening). The deuterium depth profile in the sample is measured using Secondary Ion Mass Spectroscopy (SIMS) and Elastic Recoil Detection Analysis (ERDA).

3. RESULTS AND DISCUSSIONS

The minimum and maximum of the energy resolved neutron spectrum generated by ATTILA for ITER-B-lite geometry for a 400 s ITER shot of 500 MW at the tungsten divertor is shown in FIG. 1a. The highest neutron flux is obtained at the outer vertical target of the divertor and the lowest is the center of the dome region. Corresponding PKA spectrum generated using SPECTER code is shown in FIG. 1b. The maximum PKA energy obtained due to the neutrons is 300 keV corresponding to the energy transfer limit of binary head-on collisions. Besides elastic collisions, the contributions from (n,n’), (n,p), (n,α), (n,2n) reactions are also taken into account to the estimation.
of recoil energy. While at higher neutron energies elastic and (n,2n) reactions dominate the recoil process, other reactions contribute significantly at lower and intermediate energies. The total interaction cross-section that leads to an energy transfer to the recoil atom more than $E_d$ is 6.09 barns.

![FIG.1](image)

FIG.1 (a) Minimum and maximum energy resolved neutron flux obtained at the tungsten divertor, (b) corresponding PKA energy distribution. Colours indicate various locations on the divertor.

The PKA spectrum obtained from the neutron flux is compared with that obtained from SRIM computations for a variety of ions is shown in FIG.2b. Five thousand trials were carried out with respective ions to calculate the PKA spectrum. An enlarged view of the PKA spectrum for energies less than 300 keV is shown FIG.2a. Simulations have been carried out with tungsten ions, light ions (Li, B, C) of 10 MeV each and 80 MeV Au ions. The low-mass, high-energy ions seem to produce a PKA spectrum closer to the energy range of what is produced by fusion neutrons. While neutrons transfer its energy entirely in elastic collisions and inelastic nuclear collisions, the ions lose their energy mainly via inelastic energy loss to electrons and elastic collisions. The inelastic loss is higher in the case of energetic light ions in contrast to heavy ions. Although the nature of decay of PKA distribution is similar in the case of heavy and light ions, quantitatively, the light ions agree with those produced by fusion neutrons. It is also to be noted that at very low energies (<1 keV) the PKA contribution from ions is significantly higher than that of neutrons. This may be due to the fact that lower energy neutrons have higher capture cross-sections than recoil.

![FIG.2](image)

FIG. 2. PKA spectrum from SRIM: PKA/ion (a) and PKA density for irradiated fluence (b), along with PKA from neutrons using ATTILA+SPECTER shown for 500 MW reactor for 0.4FPY

Experiments have been carried out using two extreme case of surrogate ions, namely, 80 MeV Au and 10 MeV B. The irradiation fluence in both the cases was $1.3 \times 10^{14}$ ions/cm$^2$. The range of both the ion species in tungsten sample were about 4.5 mm. This corresponds to 0.01 dpa in the case of B and 0.22 dpa in the case of Au. While neutrons generate PKA less than 300 keV, 10 MeV B produced PKA up to 1.5 MeV and 80 MeV Au ions created up to 58 MeV. However, the probability of high energy PKA was a factor at least $10^{-3}$ lower than of 300 keV.

The XRD studies had shown that none of the irradiated samples show any phase change and the 4-probe resistivity studies have shown that both Au and B irradiated W foils have shown a lower residual resistivity ratio (approximately 60) compared to the un-irradiated recrystallized foils (160). This indicates the formation of defects inside the sample after irradiation.
The positron annihilation lifetime measurements using 270 keV positrons from a $^{22}$Na source show that while comparing with un-irradiated samples, the intensity of the first lifetime component (107.3 ± 1.7 ps) of Au irradiated samples has reduced from 70 % to 18 %, which indicates the formation of defects throughout the positron range. The second lifetime component obtained by keeping the first lifetime component constant was found to be 154 ± 2.3 ps with intensity of 41.0 ± 7 %, which may be indicative of the presence of dislocations [11] throughout the implantation range. The third lifetime component is found to be about 259.6 ± 8.7 ps with 36.6 ± 3.9 % intensity, which corresponds to the annihilation from clusters of 6-7 vacancies [12]. In the case of B irradiated samples, the first component reduces to 46.5±3.27 % intensity indicating formation of fewer defects compared to Au irradiated samples. The intensity of this further reduces with fluence. The second component 212.3 ± 7.6 ps with 48.31 ± 7.5% intensity indicating vacancy clusters in the ion range.

Results from Doppler broadening measurements using a variable energy slow positron beam of energy up to 25 keV for both Au and B irradiated samples are shown in FIG. 3a. The high S-parameter for the un-irradiated sample is due to the positrons annihilating at or the near surface regions. As the energy increases, the percentage of positrons diffusing back to the surface reduces and thus shows a reduced S-parameter. At higher beam energies, the saturation of S-parameter indicates nearly defect-free bulk state of the annealed foils. In Au irradiated sample, the S-parameter shows the saturation at all beam energies. In the case of B irradiation, the higher values of S-parameter compared to un-irradiated ones indicate the formation of defects throughout and the values increases with boron fluence. However, it is to be noted that the values are significantly lower than Au irradiated samples indicating fewer number of defects produced in them. The contribution from the high (W-Parameter) and low (S-Parameter) momentum annihilation of the un-irradiated and irradiated foil samples is shown in FIG. 3b. The high momentum contribution is from the core electrons and indicative of the chemical environment within the sample [13]. All irradiated and un-irradiated samples fall on the same line indicating that the irradiation did not change the chemical environment of the sample.

FIG. 3. (a) S-E and (b) S-W plot obtained from PAS for Au and B irradiated W samples

Transmission electron micrographs 10 MeV B ion irradiation and 80 MeV Au ion irradiation are shown in FIG. 4a and FIG.4b at a depth of 2 μm from the surface. Isolated dislocation loops, clusters and dislocation lines are observed in the case of boron irradiated samples. Clusters together with vacancies (FIG. 4c) are observed in the case of Au irradiated samples. The cluster size in the case of Au irradiated sample varied between 2.2 – 12 nm with a cluster density was about 1.3x10^{12} cm^{-2}, which is the highest observed in any of the samples. While the dislocation line density for Au irradiated samples (6.1 x 10^8 cm^{-2}) were almost a factor of 30 lower than B irradiated samples (1.9 x 10^{10} cm^{-2}), the length of dislocation lines found to be larger in the case of Au. The loop and cluster density was two orders of magnitude higher for Au irradiated samples in comparison to B irradiated samples. For details of TEM analysis, see the contribution in this conference [14,15].
FIG. 4 TEM micrographs of 10 MeV B (a) and 80 MeV Au (c) irradiated samples, with expanded view in (b) and (c) respectively.

Molecular dynamics simulations have shown that for PKA energies less than 40 keV, point defects, mainly interstitial and vacancy clusters are of the dominant type. At room temperature irradiation events, the vacancies stayed at the center of the cascade whereas interstitials form clusters at the periphery of the cascade. However, at energies above 40 keV, spontaneous nucleation of dislocations were observed at the end of the cascade, which was typically several tens of pico-seconds. FIG. 5a shows a typical cascade event of a 100 keV PKA in single crystal tungsten lattice. The surviving vacancies (green) and interstitials (red) at the end of the cascade (at 500 ps) is shown along with the maximum extent of the thermal spike (blue background). The size of the vacancy and interstitial clusters increased with energy till 150 keV and the maximum cluster size observed in our simulations were about 970 atoms. The interstitial clusters spontaneously convert to dislocation loops and the dislocation analysis carried out using OVITO [16] software has revealed that the interstitials form dislocations predominantly of $\frac{1}{2} <111>$ type or $<100>$ type. Splitting and merging of $<111>$ dislocations to $<100>$ were also observed during the simulations. Formation of dislocation junctions of three or more line segments has also been observed. Four dislocation loops of $\frac{1}{2} <111>$ type formed at the end of the cascade is marked along with their Burgers vector in FIG. 5a. However, for PKA energies above 150 keV, it has been observed that a given PKA can produce more than one energetic Secondary Knock-on Atom (SKA) and each of them leads to a separate cascade called fragmentation of cascades. It has been observed that this tend to limit the maximum size of the vacancy and interstitial clusters produced in the cascade. A splitting of cascade event due to 315 keV PKA atom observed in the simulations is shown in FIG. 5b. The PKA is initialized at the center (blue) circle and three SKA were produced at 150 keV, 100 keV and 20 keV energies. Each of them produced independent cascades. During the simulation time, the vacancies remained in the centre of the individual cascades and the interstitials formed clusters, which turned into dislocation loops and partial dislocation lines.

FIG. 5 PKA driven cascades for 100 keV (a) and 315 keV (b)

Since a significant fraction of PKA energies produced by 80 MeV Au are above 150 keV, there is a possibility that the cascades were fragmented which led to small yet numerous interstitial dislocation loops trapped around the cascade core leading to clusters of defects. On average, each 80 MeV Au ion produces about 16 PKAs of energy above 150 keV and about 1200 PKA below it. The energetic PKA capable of producing large-scale clusters and dislocation loops were roughly about 45 per ion. The fragmented cascades lead to potentially overlapping temporal cascades. At room temperature, this can lead to the formation of cluster of small defects. Such observations are also supported by [17] for 20 MeV W$^{6+}$ irradiated tungsten. In the case of 10 MeV boron, the average number of PKA produced above 150 keV is roughly 0.015, which is roughly 3 orders of magnitude less than that of 80 MeV Au. Besides, the total number of PKA produced per ion is about 73 and almost all of them (72) were less than 10 keV. This lead to the formation of isolated Frenkel Pairs (FP), small vacancy and interstitial clusters. From MD simulations, the number of FP produced by 10 keV PKA is about 10.4 ± 2.9. The maximum cluster size observed was about 8 atoms interstitials and average defect cluster size was 3-4 atoms. Therefore, the interstitial clusters could have diffused at a longer time scale leading to the formation of small dislocation lines and isolated larger loops. The contribution from the high energy PKA, can lead to the clustering of defects, which is significantly lower than that observed in Au irradiated sample.

The S-E and S-W parameters measured using DB-positron beam measurements for tungsten samples irradiated with 250 keV He ions is shown in FIG. 6a. The irradiation fluence was $3 \times 10^{13}$ ions cm$^{-2}$ and the ion penetration range calculated from SRIM was about 460 nm. The S-parameter saturates from the surface similar to the Au irradiated sample, indicating the formation of defects all throughout the penetration range. The S-W parameter on the other hand indicates no significant variation of the chemical composition of the sample from the un-irradiated case which is not expected at such low fluence.
The TEM micrographs of He irradiated tungsten is shown in FIG. 6b indicate the formation of dislocation lines and clusters of defects. The dislocation line length varied between 40-1000 nm with a density of 1.05 x 10^9 cm^-2 and the cluster density is about 5.6 x 10^{10} cm^-2 with a size range of 5-15 nm. The maximum PKA energy obtained from SRIM was about 16 keV and the average number of PKA above 1 keV was about 0.9. Most of the PKA energies were below 5 keV. From MD simulations, the average number of surviving FP for 5 keV was about 7.2 ± 2.3 with 3-4 vacancy clusters. At room temperature, the interstitial diffusion can lead to the formation of dislocation lines.

![Image](image.png)

FIG. 6. 250 keV He irradiation: S-E curves (a) and TEM showing dislocation lines and clusters

The S-E and S-W parameters for 100 keV D irradiated tungsten are shown in FIG. 7. The S-parameter shows a broad peak between 10 to 20 keV positron energy. The peak in S-parameter usually indicates that a defect rich zone created due to irradiation. From the depth profile of the implanted positrons given by Makhovian [18] profile, this corresponds to 100 to 300 nm. The S-W curve for D irradiated sample shows a different slope compared to the un-irradiated samples. This might indicate that the annihilation is taking place from core electrons having a different angular momentum than tungsten, which could be an indication of deuterium rich defects in this depth.

![Image](image.png)

FIG.7. 100 keV Deuterium irradiation in W: S-E curves (a) and S-W curves (b)

The TEM micrographs of D irradiated sample shown in FIG.8a and FIG.8b, shows isolated and entangled dislocation lines and loops. The dislocation lines size varied between 120 to 1900 nm with a density of 9.68 x 10^8 cm^-2 and the loop size varied between 50 – 100 nm and the corresponding density was about 1.4 x 10^8 cm^-2. Although the flux and the subsequent dpa of the D irradiated samples is higher (0.85) than any of the above discussed cases, dislocation density observed is comparable or even lower than He irradiated samples. This could be attributed to the slightly higher surface temperature due to irradiation in the high current ion source (35 μA current). This might have led to a higher mobility of defects, which could also lead to the formation of bigger sized clear dislocation loops in contrast to other samples. The maximum PKA energy of 100 keV D in W calculated using SRIM was ~4 keV and on average each D atom produced 0.08 PKA of energy more than 1 keV. MD simulations show that the maximum number of surviving FP is about 6.9 ± 1.9 with the biggest cluster size of 3 vacancies. The spontaneous dislocation nucleation was not observed at this energy in the simulations. Therefore, the diffusion of interstitials might have led to the formation of dislocation lines and loops. The depth
profile of deuterium measured using SIMS and ERDA showed that most of the deuterium is trapped within 500 nm from the surface and the peak of the distribution is about 300 nm. This is in agreement with the observations of the S-W measurements using positron beam where it is observed a change in the chemical environment of the sample within this range.

![Dislocation networks (a) and loops (b)](image)

FIG. 8 100 keV Deuterium irradiation in W: dislocation networks (a) and loops (b)

A comparison of dislocation density from TEM analysis seems to show that the cluster size decreases with dpa whereas the cluster density increases. The line density seem to have no influence on the dose or on the ion mass as shown in FIG.9. The dpa is calculated for a 200 nm thick layer at the depth where the TEM was carried out.

![Dislocation line and cluster density from TEM for all the samples](image)

FIG. 9 Dislocation line and cluster density from TEM for all the samples

While comparing with neutron irradiated samples, the ion irradiated samples show dislocation lines and vacancy clusters in contrast to voids and loops. This could be because of the higher temperature or long-term diffusion of the surviving FP in the case of neutron irradiation [19,20]. It has been shown that in the case of neutron irradiated samples the higher dose rate leads to higher density of small clusters with fewer large voids [21]. However in our experiments, despite the difference in mass, energy and dose rate He and Au irradiated samples show similar cluster size (<15 nm) with a relatively high density of defects. The clusters formed after irradiation in our samples might relax to distinct loops and voids at higher temperature which is in agreement with the high temperature post-irradiation annealing experiments [17,22].

Tungsten samples pre-irradiated with Au, B and He were further irradiated with 100 keV D ions for a flux of $5 \times 10^{17}$ ions-cm$^{-2}$. The deuterium penetration and trapping in the samples have been investigated using SIMS and ERDA. The details of the experimental setup and the conditions are discussed in another contribution in the same proceedings [23]. The implantation range of 100 keV D in tungsten is roughly about 600 nm. However, the experiments indicate that most of the deuterium is within 500 nm and the observed profiles were broader than the implantation profiles. This indicates diffusion of deuterium in tungsten. From the profile we assume that the diffusion is predominantly towards the surface via defect channels created due to implantation. Besides, the surface might act as a strong trap for deuterium which has been shown in MD simulations separately [24]. In the case of Au irradiated tungsten the deuterium penetrated further into the material. This could be due to the fact that the defects produced in Au were further aiding the diffusion of D. However, the defects created due to D close to the surface was much higher (0.85 dpa) compared to Au (0.22 dpa). This might explain the pronounced diffusion towards surface. This might also be the reason for the observations of micron sized deuterium bubbles on the surface [25].
4. CONCLUSION

We have investigated the effect of PKA spectrum in damage structure using ions of different mass and energy. It has been observed that PKA spectrum has significant influence on the defect structure. In high-energy heavy ion irradiation, the energetic PKA cause fragmentation of the cascade that can limit the size of the interstitial/vacancy clusters. This can lead to small dense defect-clusters as observed in 80 MeV Au irradiated samples. However, light-ion irradiation with PKA energies lower than fragmentation threshold tend to form predominantly dislocation lines and distinct loops. The deuterium trapping studies indicate that defects can act as a channel for D diffusion after implantation and the trapping can be dependent on the type of defects created during irradiation. This might also imply that although heavy ions are effectively used for creating fast damage with distinctly different defect structures compared to neutrons, the light ion irradiation experiments might be important for estimating the deuterium trapping in the defects.

The relationship of PKA spectra to defect formation and of defects to trapping has been explored in this work for assessment of hydrogen isotope trapping within tungsten armor in reactor like conditions. Although it is clear that trapping is more in voids, for quantitative prediction a more work is required to simulate the neutron induced PKA spectra under the relevant temperature conditions.

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