

DEUTERIUM DEPTH PROFILE MEASUREMENT IN PRE AND POST IRRADIATED TUNGSTEN

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Abstract

Experiments have been carried out using D beam in pre and post irradiated polycrystalline tungsten foils. Tungsten foils were irradiated by high energy Au, B, D alone and sequentially Au followed by He to create defects. The samples were then exposed to D ion beam to study the trapping of D in irradiation induced defects. ERDA analysis show that in polycrystalline foils irradiated with Au, and B the D concentration increases. The depth profile of D shows that after exposure of helium in tungsten foil pre-irradiated with Au, the trapping of deuterium was reduced. The observed broader D profile suggest that defects enhances the diffusion of D predominantly towards the surface.

1. INTRODUCTION

Realization of fusion technology for electrical power production is one of the biggest challenges [1]. One of the important challenge towards this is the selection of appropriate Plasma Facing Materials (PFMs) [2], which can handle the high heat flux, energetic neutrons without degradation in its mechanical and chemical properties. High flux plasma exposure can lead to the permeation and trapping of hydrogen isotopes e.g. D and T. Tungsten has emerged as one of the most promising candidate for PFM and will be used in ITER due to its capability to handle high heat flux while having a low hydrogen (H) isotope affinity. However, in presence of fusion neutrons and alpha particles, tungsten can accumulate radiation damage, which might significantly enhance its H retention property.

In order to study the effect of radiation damage in tungsten, various experiments have been carried out using deuterium plasma, ion beams and gaseous ions in tungsten samples [3–5]. In order to create the damage, the tungsten samples were irradiated using both neutron & ion beams. The samples were analysed for D-depth profiles using NRA [5], SIMS & ERDA [6,7] and the total deuterium content was measured using thermal desorption experiments [3,7]. It has been observed that at low sample temperatures, most of the deuterium trapped at dislocations whereas, voids seem to be the favourite trapping sites for deuterium at high temperatures [8,9]. It has been observed that the defect structure in the sample depends on the rate of damage production and therefore it is possible that the subsequent estimation of trapping may also depend on the history of damage [10]. Available

experimental data generated using W-self ions and the heavy ions show the generation of cluster of defects with small dislocation loops. In order to extrapolate the D-trapping to neutron-like conditions, one need to study the effect on trapping using a variety of ions with different mass and energy. We have carried out experiments on deuterium trapping by creating defects in tungsten foils samples using high and low mass ions followed by exposure to D ion beam.

2. EXPERIMENTAL METHOD

Polycrystalline Tungsten (PCW) foil samples of size 8mmx8mmx0.1mm foils were mechanically polished and annealed at 1838 K to release the stress, to minimize the defects and recrystallize. These tungsten foils were irradiated by high energy Au⁷⁺ (80 MeV) ions at IUAC, Delhi [11], the beam current was varied between 1 to 3 pA. The beam was rastered uniformly over the sample surface for a fluence of 1×10^{14} cm⁻². Few foils were irradiated with 10 MeV boron ions (B³⁺) at Guru Ghasidas University at Bilaspur [12]. The irradiation chamber was evacuated to a base-pressure of 1×10^{-6} mbar and the foils were exposed to a beam current of 200 pA for two different fluences of 1.3×10^{14} ions-cm⁻² and 1×10^{15} ions-cm⁻².

In order to analyse the effect presence of Helium, on trapping of D in tungsten, helium-ions of 250 keV were implanted in samples which had been already irradiated by either gold or boron ions. This was done by using Low Energy Ion Beam Facility (LEIBF) at IUAC Delhi, for a fluence of 5×10^{15} ions-cm⁻².

The samples which had been irradiated by gold, boron and helium were further exposed to deuterium-ions of 100 keV, using a current of 20-100 μ A from the 14 MeV neutron generator at IPR. The sequence of irradiation followed was Au+D, Au+He+D, B+D, He+D, and D alone. The samples were exposed to a fluence of 5×10^{17} ions-cm⁻². The procedure is described in a flow chart as shown in FIG.1.

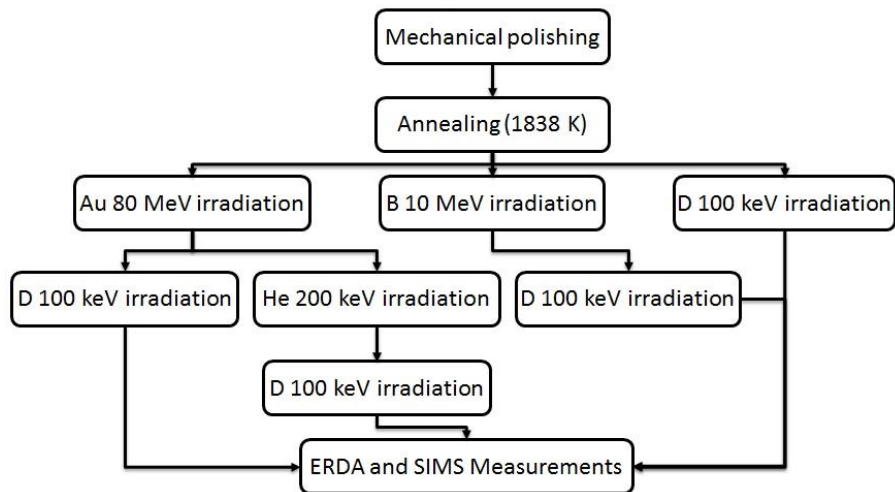


FIG.1: Flow chart for the irradiation and measurement experiments

The trapped D was measured using Elastic Recoil Detection Analysis (ERDA) and 6.4 Secondary Ion Mass Spectroscopy (SIMS). For ERDA A carbon beam (C⁴⁺) of 7.6 MeV energy was used yielding beam current of ~30 nA. Silicon surface barrier type detector having area 0.2 cm² was used and recoiled D atoms were detected at an angle of 85°. The detector window was covered with 6 μ m Al foil to stop forward scattered carbon atoms. The distance between detector and target is about 6 cm. To investigate the depth profiling the calibration was done by the measurement on the polyethylene–Mylar ERDA spectrum, afterwards fitted with SIMNRA code[13,14]. Finally D spectra obtained from Tungsten samples were fitted using SIMNRA code to get the D concentration (FIG.2).

The deuterium and helium depth profiles were measured using secondary ion mass spectroscopy. The depth distribution analyses of all the elements in all the specimens were carried out using magnetic sector based Cameca IMS-7f instrument equipped with both oxygen (O²⁺ and O⁻) and cesium (Cs⁺) primary ion beams. The depth distribution analyses were carried out using Cesium (Cs⁺) primary ion beam at an impact energy of 15 keV with negative secondary ion detection mode. Primary beam with beam current of 75 \pm 1 nA falls on the sample surface at an incidence angle of 23° w.r.t. surface normal. Primary ion beam was raster over an area of 250 μ m \times 250 μ m and secondary ions were collected from an analysis region of 63 μ m in diameter at the centre of raster area by

selecting the field aperture of 750 μm in order to remove the crater edge effects. Mass resolution (m/dm) of 400 was selected in all the analysis. The pressure in the analysis chamber was maintained $\sim 7 \times 10^{-9}$ mbar.

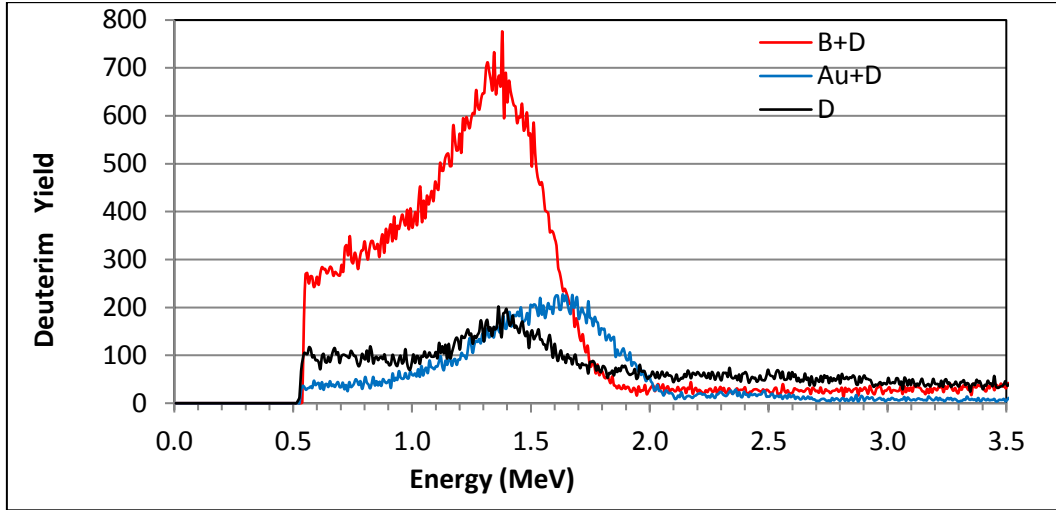


FIG.2: Deuterium yield as measured using ERDA

3. RESULTS AND DISCUSSION

The SRIM calculation for 80 MeV Au ion in tungsten shows that the defects created have a distribution around the range $\sim 4.5 \mu\text{m}$, which shows continuously increasing density of defects up to $\sim 4 \mu\text{m}$. The measured Au profile in W as measured using SIMS is shown in FIG.3, which is in agreement with SRIM.

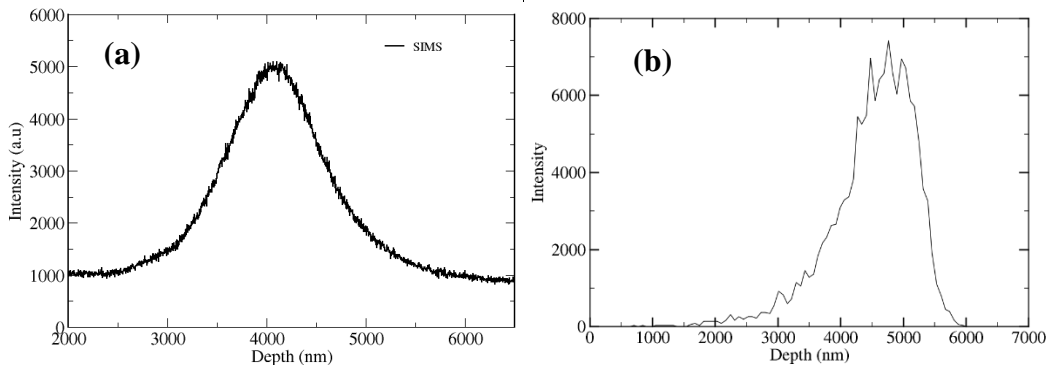


FIG.3: Au depth profile in W as measured using SIMS (a) and estimated from SRIM (b)

The range of both Au and B are roughly of the same order i.e., $\sim 4.5 \mu\text{m}$. The mean range of 100 keV D in tungsten calculated using binary SRIM was about 427 ± 139 nm and for 250 keV He is 429 ± 124 nm (FIG.4a and b). The results of ERDA measurements are shown in FIG.5, along with the fitted SIMNRA spectra. The results from ERDA analysis show that in polycrystalline foils irradiated with Au, the D concentration increases after 150 nm depth with peak in range 200 to 300 nm. The increase in the D concentration in pre-irradiated samples is larger as compared to the sample irradiated with D only case. The D profile in B irradiated samples is more homogeneous (FIG.5 blue color) as compared to Au (black color). This may be due to the qualitative difference in the defects (more line defects in B as compared to Au) as shown by TEM analysis by Prashant et al. 2018 [17]. Whether D trapping is preferentially higher at dislocation lines needs further investigation. The depth profile of D shows that after exposure of helium in pre-irradiated tungsten foils, the trapping of deuterium was reduced significantly. This might be because the defects created by ion irradiation are preoccupied by He and less number of vacancies are available to be filled by D leading to the lowering of D concentration in He implanted samples.

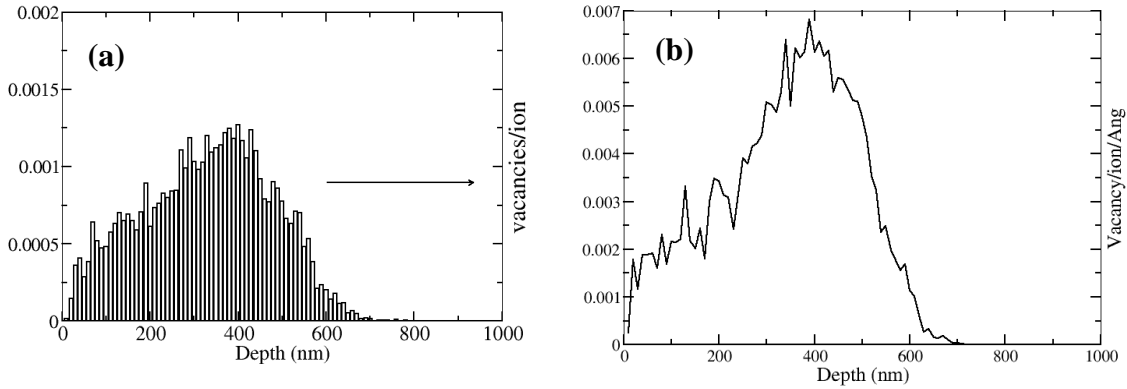


FIG.4 SRIM calculated Range profiles for (a) 100 keV D and (b) 250 keV He

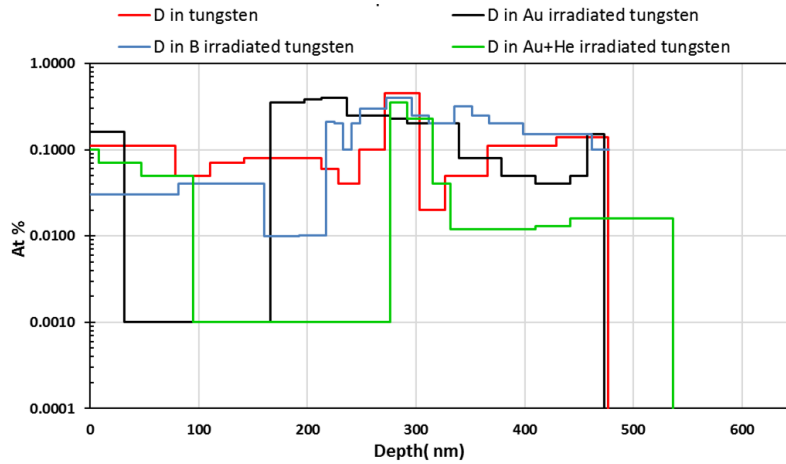


FIG.5: SIMNRA calculated D depth profiles implanted for different ion-irradiations in Tungsten foils

The ERDA results for bulk tungsten alloy (1% La_2O_3) are shown in FIG.6. The D concentration and peak does not show any significant change in with and without pre-irradiated samples. It is proposed in case of tungsten alloy that the D trapping is largely governed [18] by the impurity present in the tungsten and have least effect of irradiation. However, it is quiet possible that larger fluence of ion-irradiation or D may alter the results. However, under the present constraints of experiments, ERDA results show that deuterium trapping is not pre-damage/defect dependent. This implies that the impurity plays an important role in D trapping in tungsten lattice and therefore, a different mechanism of D trapping is involved as compared to pure tungsten.

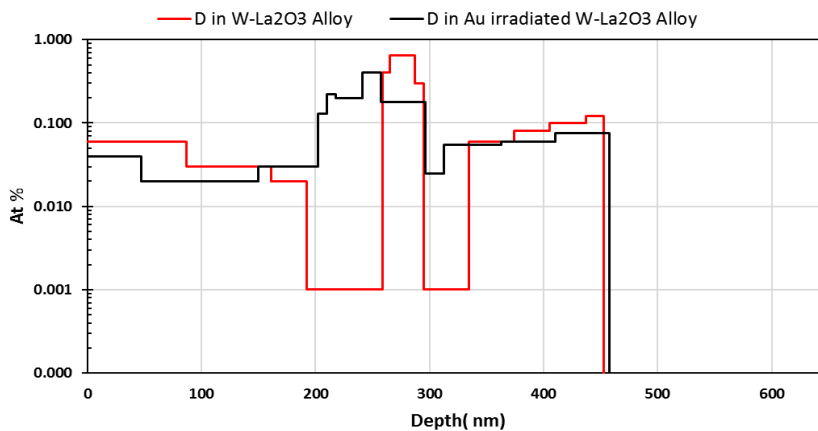


FIG.6: SIMNRA calculated D depth profiles implanted for different ion-irradiations in Tungsten Alloy

The SIMS data shown in FIG.7 show large surface concentration of D atoms in all the trials. A bump in the concentration profile is observed between 250 nm to 400 nm. The surface peak is shifted in the depth for the foils pre-damaged with 80 MeV Au ions. The deuterium depth profiles in the case of foils pre-damaged with 10 MeV boron shows a rather broad peak towards depth of the sample. The surface as well as the total deuterium content is found to be higher in the case of gold irradiation.

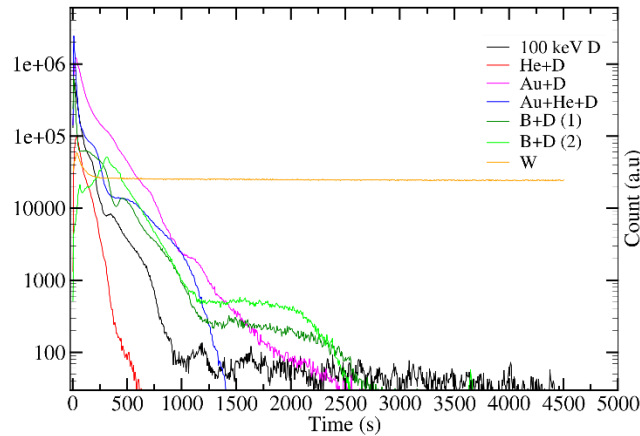


FIG.7: Deuterium profile (SIMS) in un-damaged W foil, foils damaged with 10 MeV B and 80 MeV Au ions (0.5 nm/s to be used for the conversion of time into depth profile) .

A surface peak concentration is visible in unirradiated and irradiated samples. The profile is shifted towards inside for the foils pre-damaged with 80 MeV Au ions. The D range as obtained from SIMS and ERDA is smaller as compared to estimate from SRIM calculations. The observed profiles from experiments were broader than the implantation profiles. This indicates diffusion of deuterium in tungsten. MD simulations [19] showed that surface can also act as trapping site for deuterium. Besides, there are defects available from surface to the D implantation range, which might work as pathways for diffusion of D from implanted range towards surface making the D profile broader. This indicates that the D diffusion is mostly towards surface. The defects created due to D close to the surface was much higher (0.85 dpa) compared to Au (0.22 dpa) [16]. This might also explain the pronounced diffusion towards surface.

4. CONCLUSION

D trapping in pure W with and without Au and B ion irradiation was investigated. The major outcome of the present study are

- (a) The D trapping is enhanced by the ion irradiation in PCW foils. The D profile seems broader and peaking towards the surface, which is puzzling as one expects it to peak around the implantation depth.
- (b) ERDA and SIMS observation are in agreement as far as the total range of D.
- (c) Sequential irradiation of pre-irradiated (with Au) foils with He and D indicate that D trapping is significantly suppressed by the presence of He.
- (d) Noting the uniformity of the defects over the D implantation range created by ion irradiations, it is proposed that the diffusion of D may be present and responsible for broader D profile.

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REFERENCES

- [1]. Zinkle, S. J. Fusion materials science: Overview of challenges and recent progress. *Phys. Plasmas* **2005**, *12*, 1–8, doi:10.1063/1.1880013.
- [2]. Kaufmann, M.; Neu, R. Tungsten as first wall material in fusion devices. *Fusion Eng. Des.* **2007**, *82*, 521–527, doi:10.1016/j.fusengdes.2007.03.045.

- [3]. Schwarz-Selinger, T. Deuterium retention in MeV self-implanted tungsten: Influence of damaging dose rate. *Nucl. Mater. Energy* **2017**, *12*, 683–688, doi:10.1016/j.nme.2017.02.003.
- [4]. Hasegawa, A.; Fukuda, M.; Nogami, S.; Yabuuchi, K. Neutron irradiation effects on tungsten materials. *Fusion Eng. Des.* **2014**, *89*, 1568–1572, doi:10.1016/j.fusengdes.2014.04.035.
- [5]. Ogorodnikova, O. V.; Gann, V. Simulation of neutron-induced damage in tungsten by irradiation with energetic self-ions. *J. Nucl. Mater.* **2015**, *460*, doi:10.1016/j.jnucmat.2015.02.004.
- [6]. Markelj, S.; Schwarz-Selinger, T.; Založnik, A. Hydrogen isotope accumulation in the helium implantation zone in tungsten. *Nucl. Fusion* **2017**, *57*, doi:10.1088/1741-4326/aa6b27.
- [7]. Gasparyan, Y.; Rasinski, M.; Mayer, M.; Pisarev, A.; Roth, J. Deuterium ion-driven permeation and bulk retention in tungsten. *J. Nucl. Mater.* **2011**, *417*, 540–544, doi:10.1016/j.jnucmat.2010.12.119.
- [8]. Kobayashi, M.; Shimada, M.; Hatano, Y.; Oda, T.; Merrill, B.; Oya, Y.; Okuno, K. Deuterium trapping by irradiation damage in tungsten induced by different displacement processes. *Fusion Eng. Des.* **2013**, *88*, 1749–1752, doi:10.1016/j.fusengdes.2013.04.009.
- [9]. 'T Hoen, M. H. J.; Dellasega, D.; Pezzoli, A.; Passoni, M.; Kleyn, A. W.; Zeijlmans Van Emmichoven, P. A. Deuterium retention and surface modifications of nanocrystalline tungsten films exposed to high-flux plasma. *J. Nucl. Mater.* **2015**, *463*, doi:10.1016/j.jnucmat.2014.11.025.
- [10]. Tanno, T.; Fukuda, M.; Nogami, S.; Hasegawa, A. Microstructure Development in Neutron Irradiated Tungsten Alloys. *Mater. Trans.* **2011**, *52*, 1447–1451, doi:10.2320/matertrans.MBW201025.
- [11]. Kanjilal, D.; Chopra, S.; Narayanan, M. M.; Iyer, I. S.; Jha, V.; Joshi, R.; Datta, S. K. Testing and operation of the 15UD Pelletron at NSC. *Nucl. Inst. Methods Phys. Res. A* **1993**, *328*, 97–100, doi:10.1016/0168-9002(93)90610-T.
- [12]. Trivedi, T.; Patel, S. P.; Chandra, P.; Bajpai, P. K. Ion Beam Facilities at the National Centre for Accelerator based Research using a 3 MV Pelletron Accelerator. *Phys. Procedia* **2017**, *90*, 100–106, doi:10.1016/j.phpro.2017.09.032.
- [13]. M. Mayer *SIMNRA User's Guide*; Garching, Germany, 1997;
- [14]. Mayer, M. SIMNRA, a simulation program for the analysis of NRA, RBS and ERDA. *AIP Conf. Proc.* **1999**, *475*, 541–544, doi:10.1063/1.59188.
- [15]. James F Ziegler, MD Ziegler, J. B. James F Ziegler, MD Ziegler, JP Biersack. *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms* **2010**, *268*, 18181823.
- [16]. Maya, P. N.; Sharma, P.; Raole, P. .; Vala, S. S.; Satyaprasad, A.; Mukherjee, S.; Pujari, P. K.; Kulriya, P.; Bajpai, P. K.; Attri, A.; Tyagi, A. K.; Warriar, M.; Subhash, P. V.; Kikani, P.; Rayjada, P.; David, C.; Lakhani, A.; Karki, V.; Singh, M.; Kumar, R.; Abhangi, M. R.; Devrani, K.; Kedarmal; Patel, S. P.; Trivedi, T.; Saravanan, K.; Kannan, S.; Mishra, S.; Khan, K. B.; Nandi, P.; Khirwadkar, S.; Deshpande, S. P. Evaluation of tungsten as divertor plasma-facing material: results from ion irradiation experiments and computer simulations. In *FEC 2018 MPT/2-3*; Ahmedabad, 2018.
- [17]. Sharma, P.; Satyaprasad, A.; Maya, P. N.; Raole, P. M.; Attri, A.; Tyagi, K.; Kulriya, P. K.; Bajpai, P. K.; Mishra, S.; Trivedi, T.; Khan, K. B.; Deshpande, S. P. A TRANSMISSION ELECTRON MICROSCOPY INVESTIGATION OF DEFECTS INDUCED IN TUNGSTEN FOILS BY GOLD (Au) AND BORON (B) ION IRRADIATION. In *FEC 2018 EX/P2-10*; 2018; p. 8.
- [18]. Fukumoto, M.; Nakano, T.; Ueda, Y.; Itami, K.; Kubo, H. Deuterium retention in tungsten coating layers irradiated with deuterium and carbon ions. *J. Nucl. Mater.* **2015**, *462*, 354–359, doi:10.1016/j.jnucmat.2015.02.041.
- [19]. Maya, P. N. Molecular dynamics studies of sticking and reflection of low-energy deuterium on single crystal tungsten. *J. Nucl. Mater.* **2016**, *480*, 411–419, doi:10.1016/j.jnucmat.2016.08.007.