Nuclear Materials and Energy 16 (2018) 217–220

Contents lists available at ScienceDirect

Nuclear Materials and Energy

journal homepage: www.elsevier.com/locate/nme

Surface or bulk He existence effect on deuterium retention in Fe ion damaged W

Yasuhisa Oya[a,*], Shodai Sakurada[a], Keisuke Azuma[a], Qilai Zhou[a], Akihiro Togari[a], Sosuke Kondo[b], Tatsuya Hinoki[b], Naoaki Yoshida[c], Dean Buchenauer[d], Robert Kolasinski[d], Masashi Shimada[e], Chase N. Taylor[e], Takumi Chikada[a], Yuji Hatano[f]

[a] Graduate School of Science and Technology, Shizuoka University, Shizuoka 422-8529, Japan
[b] Institute of Advanced Energy, Kyoto University, Uji, Kyoto 611-0011, Japan
[c] Research Institute for Applied Mechanics, Kyushu University, Kasuga, Fukuoka 816-8580, Japan
[d] Energy Innovation Department, Sandia National Laboratories, Livermore, CA 94550, USA
[e] Fusion Safety Program, Idaho National Laboratory, Idaho Falls, ID 83415, USA
[f] Hydrogen Isotope Research Center, University of Toyama, Toyama 930-8555, Japan

ARTICLE INFO

Keywords:
He in damaged W
Hydrogen isotope retention behavior
Damaged W
Fusion

ABSTRACT

To evaluate Helium (He) effect on hydrogen isotope retention in tungsten (W), He^+ was introduced into W bulk by 201 – 1000 keV He^+, or W surface by 3 keV He^+ for 6 MeV Fe ion damaged W at room temperature. The deuterium (D) retention behavior was evaluated by thermal desorption spectroscopy (TDS). In addition, the amount of tritium (T) at surface and bulk were separately evaluated by beta-ray induced X-ray spectroscopy (BIXS). The experimental results indicated that the formation of He-void complexes reduced the D trapping in vacancies and voids which have higher trapping energy by the bulk He retention. The BIXS measurement also supported the He enhanced the D reduction in the W bulk region. On the other hand, the He ion irradiation near the surface region enhanced D trapping by dislocation loops or surface, indicating the existence of He near surface interfered the D diffusion toward the bulk. It was concluded that the He existence in bulk or surface will significantly change the D trapping and diffusion behavior in damaged W.

1. Introduction

Evaluation of fuel retention in plasma facing materials (PFM), tungsten (W), is one of the important issues to sustain safe fusion operation with sufficient supply of deuterium(D)/tritium(T). 14 MeV neutrons will introduce various types of irradiation damages like dislocation loops, mono vacancies, vacancy clusters and voids, which will be potential stable hydrogen isotope trapping sites in W [1–5]. Therefore, the additional desorption stages were found at higher temperature side in TDS spectra by the accumulation of irradiation damages [3]. The desorption behavior is also controlled by the damage distribution in W. Therefore, the evaluation of D retention and desorption characteristics, the damage profile and concentration are quite important information for precise evaluation of tritium recycling.

Introduction of helium (He) will form He bubbles and/or occupy hydrogen isotope trapping sites, which will make a large impact on hydrogen isotope retention characteristics [6–15]. There are many studies dedicated to He irradiation effect on hydrogen isotope retention in W. It is reported that, by low energy He plasma exposure, the presence of He reduces D diffusion toward bulk and He + D mixed plasma makes significant reduction of D retention in wide temperature range of 350–700 K with suppression of blistering.

These backgrounds indicate that both of irradiation damages and He distribution should be taken into account for tritium retention estimation. In DT fusion reaction, He with lower energy and high flux will be implanted and accumulated near the surface region. However, the disintegration of radionuclide and tritium produces both of ^{4}He and ^{3}He. Therefore, low concentration He will exist throughout the PFMs with a concentration of ~1 appm/He [16]. Therefore, the synergetic effect of both He and damage introduction on hydrogen isotope retention near surface (~50 nm) and/or bulk (~2 μm) region in W should be evaluated.

2. Experimental setup

The mirror finished disk-type tungsten samples (diameter 10 mm × thickness 0.5 mm) were prepared by cutting polycrystalline

* Corresponding author.
E-mail address: oya.yasuhisa@shizuoka.ac.jp (Y. Oya).

https://doi.org/10.1016/j.nme.2018.06.018
Received 29 November 2017; Received in revised form 22 June 2018; Accepted 26 June 2018
Available online 20 July 2018

tungsten rods manufactured by Allied Material Corp., Japan. All the samples were annealed at 1173 K for 30 min in vacuum to remove residual hydrogen and damage introduced during the fabrication processes. Two different experimental procedures are described. For the preparation of bulk He irradiated samples, these samples were installed into Dual-Beam Facility for Energy Science and Technology (DuET) facility at Kyoto University. A 1.7 MV tandem accelerator and 1 MV single end accelerator were used to introduce 6.4 MeV Fe^{3+} and, 201, 467, 737 and 1000 keV He^{+} simultaneously at room temperature. A total of four He ion energies were used in order to obtain a deeper He distribution in the bulk. To clarify the clear bulk He existence effect on hydrogen isotope retention in the damaged W, the ratio of He to damage concentration was set to be 10 appm He / dpa on average, in this study. The total introduced damages were 0.1 or 0.3 dpa calculated by SRIM code with the fluence of 7.3×10^{18} – 2.2×10^{18} Fe^{2+} m^{-2} and He fluence was evaluated to be 2.2 – 6.6×10^{18} He^{+} m^{-2} using the displacement energy of 50 eV. Fig. 1 shows the depth profiles of damage and He distribution within the depth of 2 μm evaluated by integration of SRIM results with all the He species. 1 keV D_2^{+} irradiation was performed with the ion flux of 1.0×10^{18} D^{+} m^{-2} s^{-1} up to the fluence of 1.0×10^{22} D^{+} m^{-2} at room temperature by triple ion implantation system at Shizuoka University after the simultaneous Fe + He ion irradiation [17]. Thereafter thermal desorption spectroscopy (TDS) was applied from room temperature up to 1173 K with the heating rate of 0.5 K s^{-1} to evaluate the D desorption behavior using a high-resolution mass spectrometer (SPECTRA Microvision Plus) purchased by SPECTRA (present MKS). Before starting the experiment, the mass sensitivity was calibrated using two D_2 standard leaks with different leak rate. The separation of D_2 and He was confirmed using 1:1 of D_2 and He mixture gas.

For the preparation of surface He irradiated sample, 3 MV tandem accelerator at Takasaki Ion Accelerators for Advanced Radiation Application (TIARA) in National Institutes for Quantum and Radiological Science and Technology (QST) was used to introduce irradiation damage of 0.1 dpa in average by 6 MeV Fe^{2+} irradiation at room temperature. Thereafter, 3 keV He^{+} irradiation was performed with the ion flux of 5.0×10^{17} He^{+} m^{-2} s^{-1} up to the fluence of 5.0×10^{19} - 5.0×10^{20} He^{+} m^{-2} at room temperature, which was higher than the fluence for bulk He implanted sample. The implantation depth of He was ~50 nm. The 1 keV D_2^{+} was also irradiated into the sample with 1.0×10^{18} D^{+} m^{-2} s^{-1} up to the fluence of 0.5×10^{22} - 1.0×10^{22} D^{+} m^{-2}at room temperature. The same TDS system mentioned above was used.

Transmission Electron Microscope (TEM) observation was also performed at Research Institute of Applied Mechanics, Kyushu University using the small thin sample with the diameter of ϕ 3 mm.

Tritium gas exposure with the pressure of 2 kPa for 4 h at 573 K for both samples was also performed to clarify tritium diffusion behavior for only Fe^{2+} irradiated W and simultaneous Fe + He ion irradiated W. After the gas exposure, beta-ray induced X-ray spectroscopy (BIXS) was performed to evaluate the T depth profile using the analyses of characteristic X-ray of Ar (Kα) and W (Mα) with the measurement time of 19 h. The surface T distribution was analyzed by tritium imaging plate (IP) technique with exposure time of 1 h, too.

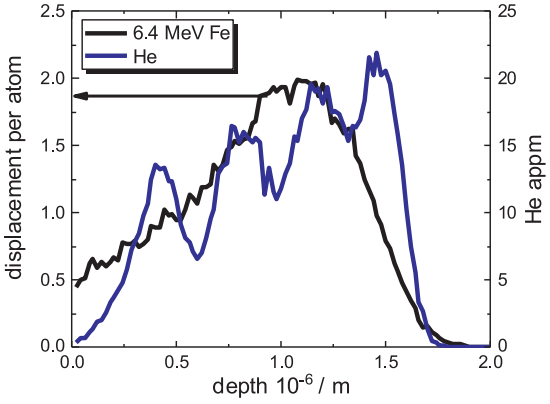

Fig. 1. Distribution of irradiation damages and He in W by simultaneous Fe and He ion irradiation at DuET evaluated by SRIM.

3. Results and discussion

Fig. 2 shows the D_2 and He TDS spectra for bulk He irradiated samples with 0.1 or 0.3 dpa and surface He irradiated samples with 0.3 dpa. For the comparison, the TDS spectrum for only 0.1 dpa Fe ion irradiated W was also shown. For all the samples, The D_2^{+} was irradiated with the fluence of 1.0×10^{22} D^{+} m^{-2}. By comparing these TDS spectra for 0.1 dpa bulk He irradiated W and only 0.1 dpa Fe ion irradiated W, it was clear that the D desorption at higher temperature above 800 K was completely reduced by the He co-implantation (see black and red plots). In addition, the D desorption peak was slightly shifted to lower temperature by simultaneous implantation. In our previous studies, the desorption stage at higher temperature was assigned to be the desorption of D trapped by voids [3,4]. Therefore, some of He migrated toward the bulk, which would be trapped by vacancies, vacancy clusters and voids, like He-vacancy complexes which reduced D trapping energy of vacancies or voids [18]. As the damage concentration has increased to 0.3 dpa, D desorption at 800 K was also observed due to introduction of dense damages by Fe ion irradiation, which is higher than that for only 0.1 dpa Fe ion irradiated W. In addition, D desorption rate at lower temperature of 400 K was also slightly decreased, leading to the reduction of D trapping sites by dislocation loops in the surface due to the He occupation. In contrast, the D_2 TDS spectrum for 0.3 dpa surface He irradiated W (5.0×10^{20} He^{+} m^{-2}) was quite different. Large desorption peak was found at lower temperature at 400 K and the desorption of D at 800 K was limited, indicating that the damages were concentrated near the surface by 3 keV He ion implantation and D diffusion to the bulk was suppressed by the existence of He. Fig. 3 shows TEM images for bulk He-Fe ion irradiated W and only Fe ion irradiated W. The density of dislocation loops was increased by the simultaneous irradiation of Fe^{2+} and He^{+} irradiation. In contrast, TDS result at lower desorption peak was not make a large difference between simultaneous (bulk) Fe - He ion irradiation and only Fe ion irradiation. These facts indicate that the large amount of damages was introduced in the bulk by Fe and He simultaneous implantation with broad damage introduction up to 2 μm. It can be said

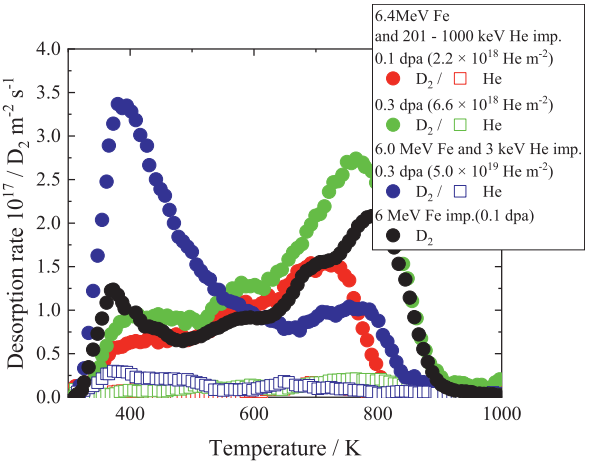

Fig. 2. D_2 and He TDS spectra for bulk He irradiated samples with 0.1 or 0.3 dpa, surface He irradiated samples with 0.3 dpa and only 0.1 dpa Fe_2^{+} irradiated W.

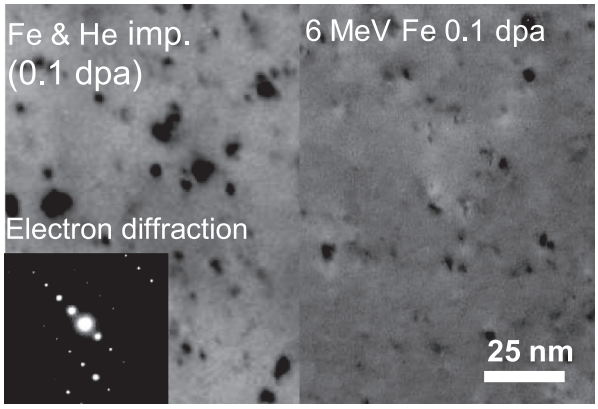

Fig. 3. TEM images for bulk He^+ irradiated W (simultaneous Fe - He ion irradiation) and only Fe ion irradiated W.

that He effect for the retention of D trapped by dislocation loops near the surface was smaller than that in sequential Fe – 3 keV He ion implantation. In Fig. 2, He TDS spectra were also shown, but the amount of desorbed He was quite limited due to lower He fluence. The other important consideration is that the heating temperature is not sufficient to desorb He from W due to much more stable trapping of He compared with that of D [7].

More detailed analyses for the surface He irradiated W were made with changing the He fluence from 5.0×10^{19} - 5.0×10^{20} He^+ m^{-2} at room temperature or 5.0×10^{20} He^+ m^{-2} after Fe irradiation at 1173 K. Fig. 4 summarized their D_2 and He TDS spectra. Three major D_2 desorption stages were found at the temperature of ~400, ~650 and ~800 K. In lower He fluence, the D_2 desorption was concentrated on the higher temperature side, but the He fluence was increased all the three desorption stages were confirmed. However, when Fe ion irradiated at high temperature such as 1173 K, no so much D_2 desorption was observed at higher temperature above 600 K caused by annealing which recover the defects [19]. It was found that the D trapping with lower energy would be controlled by the concentration of dislocation loops and/or surface damages introduced by He ion irradiation at room temperature. The D desorption at higher temperature of 600 ~800 K would be mainly controlled by the damages introduced by Fe ion irradiation. The D retention for bulk He ion irradiated W and surface He ion irradiated W was summarized in Table 1. The accumulation of damages from 0.1 dpa to 0.3 dpa (bulk Fe-He irradiated W) enhanced D

Table 1
Summary of D retentions for bulk He ion irradiated W and surface He ion irradiated W.

Sample	D retention /m^{-2}
0.3 dpa (Bulk He / 6.6×10^{18} m^{-2})	3.26×10^{20}
0.3 dpa (Surface He / 5.0×10^{20} m^{-2})	2.68×10^{20}
0.1 dpa (Bulk He / 2.2×10^{18} m^{-2})	2.07×10^{20}
0.1 dpa (Only Fe imp.)	2.57×10^{20}
0.3 dpa (Surface He / 5.0×10^{19} m^{-2})	1.29×10^{20}
0.1 dpa (Surface He / 5.0×10^{20} m^{-2}) @ 1173 K	1.64×10^{20}

retention in W. However, the formation of He-vacancy complexes may lead to suppressing D retention in W by comparing 0.1 dpa bulk He^+-Fe^{3+} irradiated W and only Fe ion irradiated W. Therefore, the total D retention would be determined by the competition of these two processes. For the surface He irradiation, the fluence of 3 keV He^+ was higher than that for bulk He^+ irradiation, which may lead to the high D retention. However, even lower He ion fluence of 2.2×10^{18} He^+ m^{-2} and He concentration of 10 appm He/dpa, the existence of He in bulk region clearly change the D desorption behavior and it can be said that He interacts with voids to form He - vacancy complexes. The growth of He bubbles refrain D trapping, which is consistent with the previous report [9,23].

To confirm the hydrogen isotope trapping efficiency on the surface of damaged W, tritium gas (HTO) exposure was performed. Fig. 5 shows the tritium IP images for 0.1 dpa Fe^{2+} damaged W and 0.1 or 0.3 dpa bulk Fe + He ion irradiated W. The results showed that tritium concentrations on the surface for bulk Fe + He ion irradiated W were lower than that for only Fe ion damaged W, indicating the existence of He would prevent the accumulation of tritium near surface region.

To confirm the tritium surface and bulk retention, BIXS analysis was applied [20–22]. The characteristic X-ray of Ar (Kα) gives the tritium retention within the depth of 50 nm, however the escape depths of W (Mα) and W(Lα) characteristic X-rays are extended to 1 μm and more than 5 μm. Therefore, the comparison of both intensities gives the tritium migration in the damaged W as shown in Fig. 6. There was small peak for W(Lα), indicating tritium migration was limited within the depth of 5 μm by tritium gas exposure with 2 kPa at 573 K for 4 h. Both of intensities of W(Mα) and Ar(Kα) for only 0.1 dpa Fe ion irradiated W were higher than that for simultaneous Fe - He ion irradiated W with 0.1 dpa or 0.3 dpa, showing that the Fe - He ion irradiation refrains D trapping on the surface. It can be clearly said that the distribution of He from the surface to deeper bulk region of 2 μm reduces tritium diffusion and trapping due to the existence of He with forming vacancy complex.

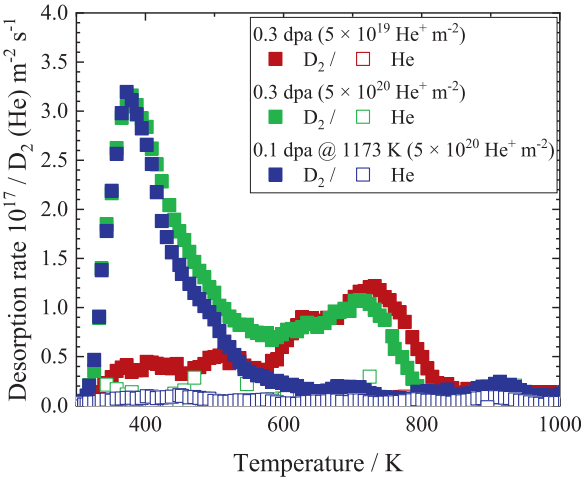

Fig. 4. D_2 and He TDS spectra for 6 MeV Fe_2^+ - 3 keV He^+ sequential irradiated W with various damage concentrations and He fluences.

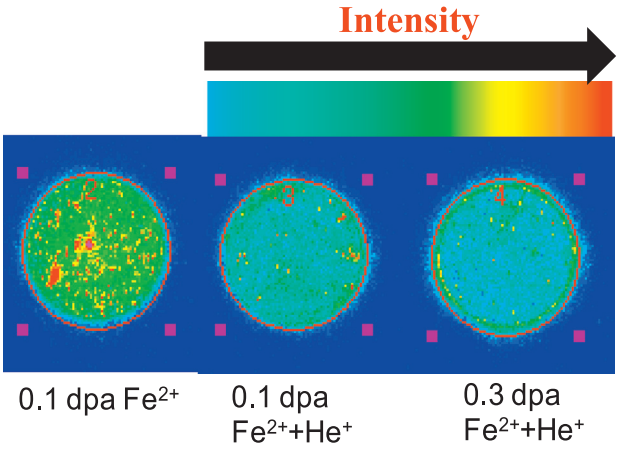

Fig. 5. Tritium IP images for only Fe ion irradiated W and bulk He ion irradiated W (simultaneous Fe-He ion irradiation).

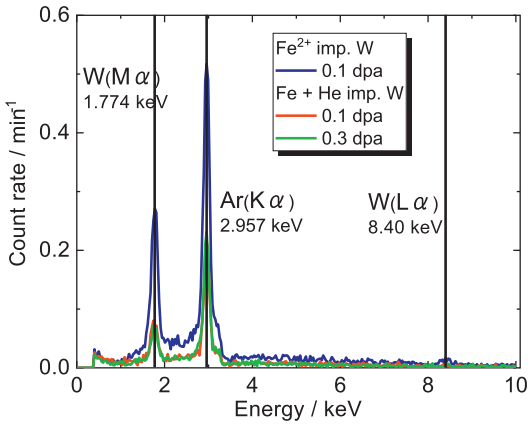

Fig. 6. BIXS spectra for only Fe ion irradiated W and bulk He ion irradiated W (simultaneous Fe-He ion irradiation).

4. Conclusions

The effect of He existence and damage introduction in W was studied. He was implanted into bulk or surface region for undamaged W and Fe ion damaged W and their D retention behavior was studied by TDS. It was found that the D retention was reduced by the He existence in the bulk region even if the irradiation damages were introduced. The formation of He-voids complex reduced the amount of D trapped in the voids. In addition, D trapping energy was decreased by simultaneous Fe - He ion implantation in comparison to that by only Fe implantation. In contrast, the He irradiation near surface region enhances and controls D trapping with lower energy and large D desorption was initiated at lower temperature less than 500 K, indicating the existence of He near surface would interfere D diffusion toward bulk. It was concluded that the He existence in bulk or surface will significantly change the D trapping and diffusion behavior in damaged W.

Acknowledgments

This study was supported by Japan-US collaboration program, PHENIX, the Joint Usage/Research Program on Zero-Emission Energy Research, Institute of Advanced Energy, Kyoto University (ZE29A-8), by the Collaborative Research Program of Research Institute for Applied Mechanics, Kyushu University, University of Toyama Collaboration Program (NIFS16KUHR035) and Inter-University Laboratory for the Common Use of Nuclear Facilities, Kaihoken. This material is based upon work supported by the U.S. Department of Energy Office of Science, Office of Fusion Energy Sciences, under the DOE Idaho Operations Office contract number DE-AC07-05ID14517. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.

References

[1] Y. Hatano, M. Shimada, T. Otsuka, et al., Deuterium trapping at defects created with neutron and ion irradiations in tungsten, Nucl. Fusion 53 (2013) 073006.
[2] Y. Oya, M. Shimada, M. Kobayashi, et al., Comparison of deuterium retention for ion-irradiated and neutron-irradiated tungsten, Phy. Scr. T 145 (2011) 014050.
[3] Y. Oya, X. Li, M. Sato, et al., Thermal desorption behavior of deuterium for 6 MeV Fe ion irradiated W with various damage concentrations, J. Nucl. Mater. 461 (2015) 336–340.
[4] H. Fujita, K. Yuyama, X. Li, et al., Effect of neutron energy and fluence on deuterium retention behaviour in neutron irradiated tungsten, Phys. Scr. T 167 (2016) 014068.
[5] M. Shimada, G. Cao, T. Otsuka, et al., Irradiation effect on deuterium behaviour in low-dose HFIR neutron-irradiated tungsten, Nucl. Fusion 55 (2015) 013008.
[6] Y. Ueda, H.Y. Peng, H.T. Lee, et al., Helium effects on tungsten surface morphology and deuterium retention, J. Nucl. Mater. 442 (2013) S267-272.
[7] S. Möller, D. Matveev, Y. Martynova, et al., Dynamic outgassing of deuterium, helium and nitrogen from plasma-facing materials under DEMO relevant conditions, Nucl. Fusion 57 (2017) 016020.
[8] Y. Uemura, S. Sakurada, H. Fujita, et al., Effect of helium irradiation on deuterium permeation behavior in tungsten, J. Nucl. Mater. 490 (2017) 242–246.
[9] Q. Zhou, K. Azuma, A. Togari, et al., Helium retention behavior in simultaneous He^+-H_2^+ implanted tungsten, J. Nucl. Mater. 502 (2018) 289–294.
[10] H. Iwakiri, et al., Effects of helium bombardment on the deuterium behavior in tungsten, J. Nucl. Mater. 307–311 (2002) 135–138.
[11] S. Nagata, et al., Helium and hydrogen trapping in W and Mo single-crystals irradiated by He ions, J. Nucl. Mater. 307–311 (2002) 1513–1516.
[12] H.T. Lee, et al., Hydrogen and helium trapping in tungsten under simultaneous irradiations, J. Nucl. Mater. 363–365 (2007) 898–903.
[13] O.V. Ogorodnikova, et al., The influence of radiation damage on the plasma-induced deuterium retention in self-implanted tungsten, J. Nucl. Mater. 415 (2011) S661–S666.
[14] O.V. Ogorodnikova, et al., Deuterium retention in tungsten exposed to low-energy pure and helium-seeded deuterium plasmas, J. Appl. Phys. 109 (2011) 013309.
[15] M.J. Baldwin, R.P. Doerner, Helium induced nanoscopic morphology on tungsten under fusion relevant plasma conditions, Nucl. Fusion 48 (2008) 035001.
[16] M.E. Sawan, Structural Materials Damage in Fusion and Fission Systems, University of Wisconsin Fusion Design Memos (UWFDM), 2011 UWFDM-1394 http://fti.neep.wisc.edu/pubs/uwfdm.
[17] Y. Oya, M. Kobayashi, T. Fujishima, et al., Behaviors of deuterium retention and microstructure change of tungsten simultaneously implanted with carbon and/or helium ions, Mater. Trans. 54 (2013) 430–436.
[18] O.V. Ogorodnikova, et al., Deuterium retention in tungsten in dependence of the surface conditions, J. Nucl. Mater. 313-316 (2003) 469–477.
[19] S. Sakurada, Annealing effects on deuterium retention behavior in damaged tungsten, Nucl. Mater. Energy 9 (2016) 141–144.
[20] M. Matsuyama, M. Murai, K. Watanabe, Quantitative measurement of surface tritium by beta-ray-induced X-ray spectrometry (BIXS), Fusion Sci. Tech. 41 (2002) 505–509.
[21] M. Matsuyama, T. Tanabe, N. Noda, et al., Nondestructive measurement of surface tritium by beta-ray induced X-ray spectrometry (BIXS), J. Nucl. Mater. 290 (2001) 437–442.
[22] H. Fujita, Y. Uemura, S. Sakurada, et al., The damage depth profile effect on hydrogen isotope retention behavior in heavy ion irradiated tungsten, Fusion Eng. Des. 125 (2017) 468–472.
[23] M. Sato, K. Yuyama, X. Li, et al., Effect of heating temperature on deuterium retention behavior for helium/carbon implanted tungsten, Fusion Sci. Technol. 68 (2015) 531–534.