HYDROGEN AND HELIUM RETENTION IN TUNGSTEN UNDER ION IRRADIATION

Yu. Gasparyan, M. Zibrov, V. Efimov, S. Ryabtsev, O. Ogorodnikova and A. Pisarev

National Research Nuclear University "MEPhI" (Moscow Engineering Physics Institute),
Moscow, Russia, e-mail: YMGasparyan@mephi.ru.

Interaction of helium and hydrogen ions with tungsten is intensively investigated during last decades in relation to construction of fusion reactor. Tungsten has the high melting temperature and the energy threshold for sputtering and, therefore, is considered as plasma facing material (PFM) in fusion devices in the area of largest heat loads and small energies of ions (divertor area). In particular, tungsten will be used in the international experimental reactor ITER, which is now under construction.

Hydrogen isotope behaviour in PFMs should be well understood from different reasons. Unlike current fusion devices that work with deuterium, ITER and future reactors will use D-T mixture as a fuel, and the total tritium amount in the machine should be controlled below the safety limit (1 kg for ITER [1]), as well as tritium permeation into the cooling system. On the other hand, hydrogen release from the wall should be controlled, since it may significantly affect the plasma operation, especially, during spontaneous local heating of the wall [2].

Helium appears in fusion device as a result of fusion reaction and radioactive decay of tritium, which can lead to accumulation of a serious amount of helium in reactor materials. Presence of helium in the lattice can significantly affect hydrogen isotope behavior thermal-mechanical properties of PFM.

This work discusses some recent experimental and theoretical results on hydrogen and helium retention in tungsten and their mutual influence.

Hydrogen retention

Hydrogen solubility in tungsten is very low (the heat of solution E_S^H=1.04 eV [3]). However, the high heat of solution correlates usually with the high binding energy with radiation defects, and a significant amount of hydrogen can be accumulated in tungsten due to presence of defects in the lattice. Therefore, quantitative characteristics of trapping/detrapping of hydrogen in defects should be well known to predict hydrogen isotope retention in the wide range of parameters. In spite of a large number of experiments on hydrogen retention in tungsten, there is no general agreement about these values.

Experimental investigations of hydrogen-defect interaction are often performed on the base of thermal desorption spectroscopy (TDS), and characteristics of interaction are obtained by a fitting procedure with a number of parameters, which are not well known. Therefore, even the detrapping energy from a single vacancy in W varies among different works in the wide range of 1.3-1.8 eV [4].

Under conditions of a high recombination rate at the surface, the H detrapping energy (E_{dt}) can be directly determined from the shift of the desorption maximum (T_m) in a series of TDS measurements performed with identical samples, but with different heating rates (β), using the following equation:

$$\ln\left(\frac{\beta}{T_{\mathrm{m}}^2}\right)=\ln\left(A\frac{k}{E_{dt}}\right)-\frac{E_{dt}}{k}\frac{1}{T_{\mathrm{m}}},$$

where A – a constant depending on parameters of the material and trapping sites, k – Boltzmann constant.

This method was used for investigation of D interaction with single vacancies and small vacancy clusters in tungsten [4,5]. Vacancies were produced by 10 keV D^+ ion beam with a low fluence. Subsequent annealing at 800 K was used for vacancy cluster production. On the base of series of experiments with different heating rates, the detrapping energy of D from vacancies was calculated to be (1.56±0.06) eV and vacancy clusters – (2.1±0.02) eV (see [4,5] for details). An example of application of this technique is given in figure 1.

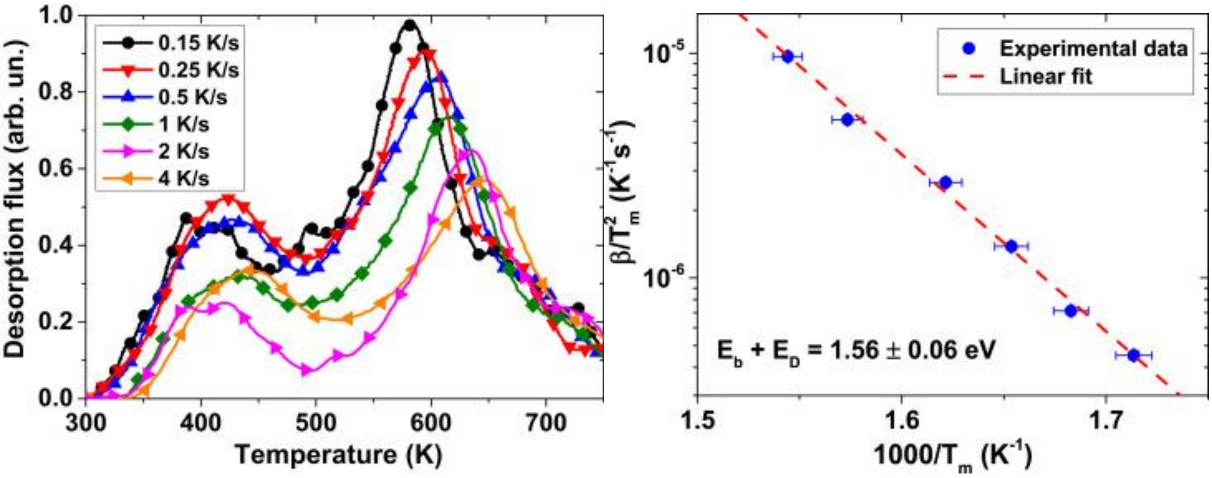

Fig.1. a) TDS spectra of D_2 from W irradiated by 10 keV/D ions to the fluence of 3×10^{19} D/m^2, subsequently annealed at 550 K for 5 min, and then implanted with 0.67 keV/D ions to the fluence of 1×10^{19} D/m^2. b) Semilogarithmic plot of β/T_m^2 versus $1/T_m$ for the peak corresponding to the D release from vacancies in W [4]

In spite of rather high detrapping energies, deuterium release from radiation defects in tungsten is observed even at room temperature during air exposure [6] or during exposure to H plasma [7], due to substitution of D atoms by H atoms (isotope exchange). This effect can be explained by the ability of defects to accumulate several H atoms (up to 6 for single vacancy [8,9]) and reduction of the detrapping energy with the increase of the number of H atoms in the trap. There is no significant impact of this process in the case of experiments with one isotope, since it is very difficult to see the difference between two independent traps and one multiple site trap. However, in the case of several isotopes, this effect can play an important role. For example, DFT calculations [9] predict reduction the detrapping energy of D atom from the single vacancy by ~1 eV in the presence of five additional H (or D) atoms in the trap, that makes D release possible at low temperatures.

Plasma facing materials in future fusion reactors will be exposed not only to low energy ions, but also to 14 MeV neutrons produced in DT- reaction. Neutrons produce a very deep damage profile and dense collision cascades, which lead to formation of complex defects in the whole bulk of PFMs. There is a limited number of researches on neutron damage in tungsten, but some groups investigated the radiation damage produced by MeV ions, which is expected to be a good proxy. One of the most adequate ways was implemented in IPP (Garching, Germany), where 20 MeV W^{6+} ions were used for damage production [10]. Heavy W ions do not contaminate the sample and produce also a dense collision cascade.

Figure 2 demonstrates experimental TDS spectra for polycrystalline tungsten damaged by 20 MeV W^{6+} ions (damage level – 0.9 dpa) and filled by D atoms at different temperatures [11].

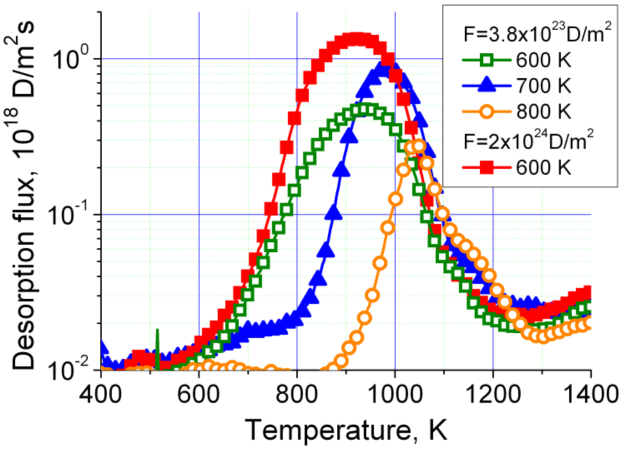

Fig.2. TDS spectra of D2 molecules for four damaged samples (0.9 dpa) exposed to D atomic flux at various conditions: 1- 600 K, 2×10^{24} D/m^2; 2- 600 K, 3.8×10^{23} D/m^2; 3- 700 K, 3.8×10^{23} D/m^2; 4- 800 K, 3.8×10^{23} D/m^2 [11]

A remarkable amount of deuterium was trapped in defects even at the highest temperature of 800 K, peak positions are also at much higher temperatures than in the case of keV ions implantation. However, the detrapping energy of D from defects responsible for trapping at elevated temperatures was determined using computer simulations to be 1.7-2.0 eV. This numbers are similar to that for the radiation damage produced by keV ions, and the differences in H behavior are explained by much deeper damage profile (~ 2μm for 20 MeV W ions) and re-trapping processes.

Similar conclusions have been done in experiments with neutrons [12]. Deuterium retention in n-damaged samples was much higher at elevated exposure temperatures due to a very deep damage profile. However, the detrapping energy was again calculated to be very similar (1.8 eV).

Helium retention

The solubility of helium in tungsten is almost zero (for helium: $E_S^{He} \approx 5.5$ eV [13]), and the detrapping energy of He from radiation defects is also very high. Helium atoms are known also to agglomerate efficiently into clusters around initially point defects, that lead finally to formation of bubbles.

Interaction of He with point defects formed by keV ion irradiation was investigated in details in [14]. TDS spectra have a multipeak structure (in the range of 1200-1800 K) that is explained by trapping of several atoms in one trap and different detrapping energies for different number of atoms in the trap.

In the case of MeV ion irradiation damage, He desorption again shifts to higher temperatures. This was observed in our recent experiments on ^{3}He desorption from the radiation damage created by 20 MeV W ions. The amount of He in the sample was controlled by nuclear reaction analysis (fig.3).

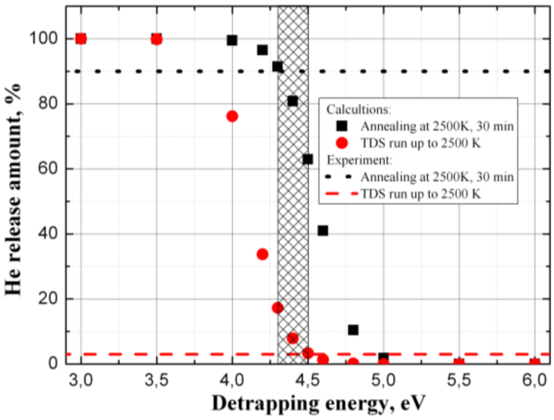

Fig.3. Comparison of experimental values of He desorption from W damaged by 20 MeV W ions and simulations using the flat profile of traps with the energy varied in the range of 3.0-6.0 eV

Only the minor part (below 5%) of implanted He desorbed during the TDS run (2 K/s) up to 2500 K, but the significant part (~90%) was released after 30 minutes annealing at 2500 K. Simulation of these experimental data using 1-D diffusion code with a flat profile of traps gave the best agreement in the case of the detrapping energy of 4.3 eV. This is again in a good agreement with keV ion experiments.

So high temperatures of helium release lead also to significant influence on annealing of radiation defects. In the case of deep radiation damage helium may stay in tungsten well above the recrystallization temperature (~1300 °C). In [15], the grain size was not changed in tungsten irradiated subsequently by W and He MeV ions even after annealing at 2000 K in contrast to the identical sample irradiated only by W MeV ions.

Opposite situation is observed in the case of high fluence plasma irradiation, where He release starts near the room temperature and the major part of He releases below 1200 K. This occurs even in the case of irradiation at very high temperatures [16]. Deuterium release from tungsten below the irradiation temperature is not observed usually. Mechanisms of this process are not explained.

Helium influence on deuterium retention

Mutual influence of helium and hydrogen isotopes is weakly investigated. One can expect that helium and deuterium will compete among themselves for places in defects, presence of helium should also lead to modification of the defect structure in materials, which may influence transport of deuterium, its retention, and release. However, some effects were already observed. The most remarkable effect is reduction of total deuterium retention [17] and ion-driven permeation flux [18] in presence of a small percentage of helium in the incident ion beam. Comparison of total deuterium inventory in tungsten after pure deuterium plasma exposure and helium (or nitrogen) seeded plasma is given in figure 7.

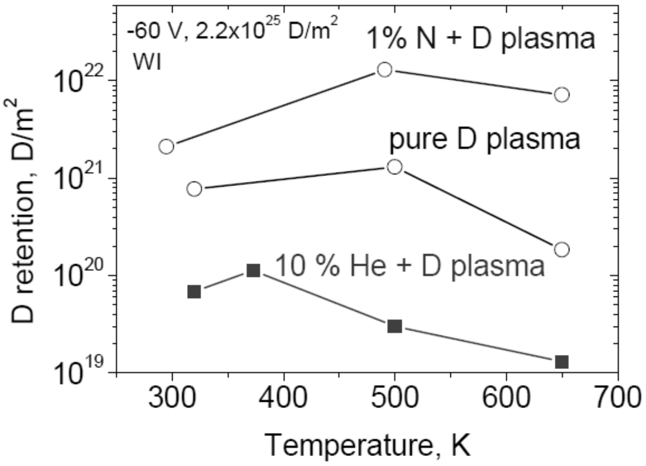

Fig.7. Deuterium retention in tungsten after deuterium plasma exposure with the fluence of 2.2×10^{25} D/m^2 without impurities and with addition of helium or nitrogen[17]

The effect of helium impurities is opposite to other impurities like nitrogen or carbon. Both effects can be explained by formation of a diffusion barrier, but at different depth. Carbon and nitrogen are expected to form a barrier at the top surface and reduce the desorption flux from the surface. In the case of helium, bubble formation behind the implantation zone of deuterium can reduce deuterium transport in the bulk.

References

1. M. Shimada, R.A. Pitts, S. Ciattaglia et al. J. Nucl. Mater., 438 (2013) S996.
2. E. D. Marenkov, S. I. Krasheninnikov, et al. Physics of Plasmas, 19, 092501 (2012).
3. R. Frauenfelder, J. Vac. Sci. Technol., 6 (1969) 388.
4. M. Zibrov, S. Ryabtsev, Yu. Gasparyan et al. J. Nucl. Mater., 477 (2016) 292.
5. S. Ryabtsev, Yu. Gasparyan, M. Zibrov, et al. Nucl. Instrum. Methods Phys. Res. B, 382 (2016) 101.
6. K.A. Moshkunov, K. Schmid, M. Mayer, et al., J. Nucl. Mater. 404 (2010) 174.
7. J. Roth, T. Schwarz-Selinger, V.Kh. Alimov, E. Markina. J. Nucl. Mater.,432 (2013) 341.
8. D. Johnson, E. Carter. Journal of Materials Research, 25 (2010) 315.
9. K. Heinola, T. Ahlgren, K. Nordlund, J. Keinonen, Physical Review B, 82 (2010) 094102.
10. B. Tyburska, V.Kh. Alimov, O.V. Ogorodnikova, et al., J. Nucl. Mater., 395 (2009) 150.
11. Yu. Gasparyan, O. Ogorodnikova, V. Efimov, et al. J. Nucl. Mater., 463 (2015) 1013.
12. Y. Hatano, M. Shimada, V.Kh. Alimov, et al. J. Nucl. Mater. 438 (2013) S114.
13. W. D. Wilson, C. L. Bisson. Radiation Effects, 19 (1973) 53.
14. E.V. Kornelsen, A.A. Van Gorkum, J. Nucl. Mater., 92 (1980) 79.
15. M. Mayer, 14th Conference on Plasma Facing Materials and Components for fusion applications, May 2013, Aachen, Germany.
16. Yu. Gasparyan, V. Efimov and K. Bystrov. Nucl. Fusion, 56 (2016) 054002.
17. O.V. Ogorodnikova, K. Sugiyama, A. Markin, et al., Phys. Scr. T145 (2011) 014034.
18. H.T. Lee, H. Tanaka, Y. Ohtsuka, Y. Ueda, J. Nucl. Mater., 415 (2011) S696.