P2-057

Influence of helium on hydrogen isotope exchange in tungsten at sequential exposures to deuterium and helium-protium plasmas

N.P. Bobyr[a*], V.Kh. Alimov[b,c], B.I. Khripunov[a], A.V. Spitsyn[a], M. Mayer[b], Y. Hatano[c], A.V. Golubeva[a], V.B. Petrov[a]

[a] *NRC "Kurcharov Institute", Ac. Kurcharov sq., 1/1, Moscow 123182, Russia*

[b] *Max-Planck-Institut für Plasmaphysik, D-85748 Garching, Germany,*

[c] *Hydrogen Isotope Research Center, University of Toyama, Toyama, 930-8555 Japan*

Abstract

Hydrogen isotopes exchange in tungsten was investigated after sequential exposures to low energy deuterium (D) and helium-seeded protium (He-seeded H) plasmas at sample temperatures of 403 and 533 K. Deuterium depth profiles were measured by the D(^{3}He, p)^{4}He nuclear reaction with $^3He^+$ energies between 0.69 and 4.5 MeV allowing determination of the D concentration up to a depth of 8 μm. It was found that a significant part of the deuterium initially retained in tungsten after D plasma exposure was released during sequential exposure to a protium plasma. However, exposure of the D-plasma-exposed W samples to the He-seeded H plasma reduces the amount of released deuterium as compared to pure H plasma exposure.

PACS: 52.40.Hf, 61.80.Jh

PSI-21 keywords: tungsten, detritiation, deuterium plasma, helium-protium plasma.

*Corresponding author address: Ac. Kurchatov sq. 1/1, Moscow, Russia.

*Corresponding author E-mail: NPBobyr@gmail.com

Presenting author: Nikolay Pavlovich Bobyr

Presenting author e-mail: NPBobyr@gmail.com

1. Introduction

Due to its favorable physical properties, such as low erosion yield and high melting temperature, tungsten (W) is a candidate material for plasma-facing high heat flux structures in future fusion reactors. A low tritium accumulation in fusion reactor materials is one of the main requirements for materials contacted with a fusion plasma [1, 2]. In the process of a burning fusion plasma, W will be irradiated by intense fluxes of deuterium (D) and tritium (T), as well as by helium (He) ions and by 14 MeV neutrons, arising from the D-T fusion reaction. Available data [3, 4] have shown that the hydrogen isotope retention in W materials exposed to high flux hydrogen plasmas differs from that after low flux ion implantation and can reach relatively high values (~10^{22} atoms/m^2) at temperatures around 500 K. This T inventory in the W materials would get close to the tritium safety limit in ITER (700 g) and would require removal of the retained tritium. One of the possible ways to recover the retained tritium might be hydrogen isotope exchange.

As reported recently in studies on hydrogen isotopes exchange in W exposed initially to a D plasma and then to a protium (H) plasma at temperatures close to room temperature [5, 6], under subsequent H plasma exposure deuterium is released from layers located at depths of 1-2 μm. However, in the case of sequential exposures to D and H plasmas at temperatures above 430 K, deuterium is released from depths up to 6 μm and even deeper. As this takes place, H is accumulated in defects generated in the W matrix under initial D plasma exposure. The rate of H accumulation in existing defects is much higher than the rate of D accumulation during the initial irradiation.

This fact raise doubts on the possibility of efficiently tritium removing to an acceptable level by isotopes exchange, as after the first cycle of replacement of tritium by some other

hydrogen isotope and subsequent irradiation by D-T plasma the W target will quickly accumulate tritium again [6].

One factor affecting the accumulation of hydrogen isotopes in tungsten during plasma exposure is the presence of He ions in the plasma. It has been shown that the addition of He in a D plasma substantially reduces the accumulation of deuterium in W at high exposure temperatures (440-650 K), compared with pure D plasma exposure [7, 8]. In W beforehand irradiated with MeV-range W ions to create radiation-induced defects, following exposure to He-seeded D plasma at elevated temperatures leads to a decreased accumulation of deuterium in the radiation damage area compared to pure D plasma exposure [9]. It was suggested that addition of He into a D plasma leads to the formation of open porosity in the surface layer. This porosity increases the deuterium re-emission reducing thus the flux of D atoms migrating into the bulk [7, 9]. Consequently, the concentration of solute D atoms decreases. Assuming that the probability of isotopic exchange depends on the concentration of the replacing isotope in the solute state [10], we can expect that seeding of He particles in tritium plasma will significantly reduce the probability of replacement of stable hydrogen isotope used in the previous detritiation cycle by radioactive tritium. In other words, it can be expected that the presence of He ions in a T plasma will significantly reduce the rate of tritium accumulation in W beforehand damaged due to high ion flux plasma exposure [11] and neutron irradiation [10].

The aim of this work was to study the processes of isotope exchange in tungsten in the case of sequential exposures to D and mixed H-He plasmas.

2. Experimental

Plates of polycrystalline tungsten (A.L.M.T. Corp., Japan), 10×10×2 mm^3 in size, recrystallized at 2073 K by annealing in hydrogen atmosphere after cutting and polishing to a

mirror surface, with a purity of 99.99 wt%, were used in this work. The grain size of the recrystallized W ranged from 20 to 200 μm.

The W samples were exposed to D, H and He-seeded H plasmas in the LENTA linear plasma divertor simulator [16]. The device is operated by a so called beam-plasma discharge driven by an electron beam. Two samples were placed on the sample holder positioned along the axis of the plasma column about 2 cm in diameter so that the plasma flux fell on the sample surfaces across the magnetic field. Bias voltages were applied to the W samples defining thus the energy of ions bombarding the surface. The -94 V bias was used for D plasma exposure and for subsequent H and He-seeded H plasmas exposure the -52 V bias was used. The flux of ions incident on the sample surface was determined from the ion current density which was 3-8 mA/cm^2. This ion current corresponds to the flux of (1.8-4.8)×10^{20} ions/m^{2}s, under the assumption that the ion flow consists mainly of the D^+ ions. The temperature of the samples during irradiation was determined by the ion flux and energy released at the target, and the temperature was measured with a thermocouple pressed in the holder and contacted with the reverse side of the sample. Just to maintain the required exposure temperature, the ion flux was adjusted by changing plasma density. The samples were exposed to the plasmas at temperatures of 403 and 533 K. For each sample exposed at 403 K, the ion flux and fluence were fixed at 1.8×10^{20} ions/m^{2}s and 3.3×10^{24} ions/m^2, respectively. At exposure temperature of 533 K, the flux and fluence were 4.8×10^{20} ions/m^{2}s and 4.1×10^{24} ions/m^2.

To generate D, H, and He-seeded H plasmas, D_2, H_2, and H_2+10%He gas mixtures were used, respectively. H_2+10%He gas mixture was prepared taking into account the ability of ionization for each gas. So, we believe that 10% of ions in He-seeded H plasma are He ions, but this is not known exactly.

The deuterium depth profiles in the plasma-exposed samples were determined by nuclear reaction analysis (NRA). The D(^{3}He, p)^{4}He reaction was utilized, and both the α particles and protons were analyzed. To determine the D concentration at larger depths, the energy of the analyzing beam of ^{3}He ions was varied from 0.69 to 4.5 MeV (0.69; 0.9; 1.2; 1.8; 2.4; 3.2; 4.5 MeV). The α-spectrum (for 0.69 MeV ^{3}He ions energy) and proton energy spectra measured at different ^{3}He ion energies were transformed into a D depth profiles up to 8 μm using the SIMNRA [12] and NRA-DC programs [13].

3. Results and discussion

Deuterium depth profiles give detailed information about depths of the deuterium depletion under sequential exposure to the D and H plasmas. From a comparison of the D profiles in the recrystallized W after D plasma exposure only (-94 V bias) and after subsequent exposures to the D and H plasmas (-52 V bias) at the same temperature T_{exp} = 403 K, the same ion flux of 1.8×10^{20} ions/m^{2}s and the same fluence of 3.3×10^{24} D(H)/m^2 (Fig. 1), it is apparent that at this exposure temperature the deuterium depletion occurs mainly at depths up to 3 μm. An insignificant difference in the D concentration at greater depths (≥ 3 μm) before and after H plasma exposures can be explained by the uncertainty in the determination of the D concentration.

However, after subsequent exposures to D and H plasmas at T_{exp} = 533 K, the same ion flux of 4.8×10^{20} ions/m^{2}s and the same fluence of 4.1×10^{24} D(H)/m^2, the deuterium depletion takes place across the whole analyzable depth, and the D concentration decreases by a factor of 4-5 (Fig. 2).

Seeding 10% of He into the H plasma at sequential exposures to D and H plasmas at T_{exp} = 403 K leads to a slight decrease in the deuterium depletion as compared with that for

sequential exposure to pure H plasma (Fig. 1). However, at T_{exp} = 533 K sequential exposures to D and He-seeded H plasmas don't cause significant release of deuterium (Fig. 2).

The total amount of deuterium retained in the recrystallized W at depths of up to 8 μm after exposures to D plasma only, after sequential exposures to D and H plasmas, and after sequential exposures to D and He-seeded H plasmas at temperatures of 403 and 533 K is shown in Fig. 3.

The fraction of deuterium released from the D-plasma-pre-exposed W under subsequent exposure to the H plasma is about 60% at T_{exp} = 403 K and about 75% at T_{exp} = 533 K. Seeding of He ions into the H plasma reduces the portion of released deuterium to about 53% at T_{exp} = 403 K and about 15% at T_{exp} = 533 K. Thus, the seeding of He ions reduces the probability of isotopic exchange at T_{irr} = 403 K insignificantly, but at T_{irr} = 533 K the probability of isotopic substitution is significantly reduced.

It may be suggested that in the case of subsequent exposures to D and H plasmas, the probability of replacement of initially retained deuterium by protium depends on the detrapping energies for defects responsible for retention of hydrogen isotopes, exposure temperature, and concentration of replacing isotopes in the solute state [10].

As mentioned above, in W irradiated with low-energy He ions, He platelets and bubbles are formed in the implantation zone [14]. Based on results obtained for simultaneous irradiation of W with helium and hydrogen ions, Ueda et al. [15] have concluded that a dynamic mechanism of nano-scale helium bubble formation can lead to development of an open porosity in the near-surface layer and can create short-circuit paths to the surface thus enhancing the H re-emission and limiting the H diffusion into the bulk. In [17] was observed that He is trapped within 30 nm of the surface. It is postulated that the incident He^+ forms He–vacancy complexes that produce secondary defects surrounding them. As this takes place, the diffusion deeper into the sample is suppressed. Additionally, the porous near-surface

structure may serve as a damper layer to dissipate the compressive stress induced by the local hydrogen super-saturation [7]. Evidently, that formation of He bubbles and interconnected porosity in W under exposure to the He-seeded H plasma depends on the exposure temperature.

In the case of subsequent exposures to the D and H plasmas at T_{exp} = 533 K, the deuterium depletion takes place across the whole analyzable depth (Fig. 2). It may be safely suggested that this exposure temperature is sufficiently high to activate the detrapping process for deuterium. As reported in [10], the probability of isotopic D-H exchange depends on the concentration of the replacing H atoms in the solution state, i.e., the higher H concentration in the solution state, the larger the deuterium depletion. Seeding of He ions into the H plasma causes formation of open porosity, that in its turn increases the H re-emission flux and decreases significantly the solute H concentration. At these conditions, the deuterium depletion from the W bulk (≥ 0.5 μm) is negligible (Fig. 2).

4. Summary

Hydrogen isotope exchange has been studied in recrystallized tungsten after (i) sequential exposure to low-energy (52-94 eV) deuterium and protium plasma and (ii) sequential to low-energy deuterium and helium-seeded protium plasmas to fluences of 3.3×10^{24} and 4.1×10^{24} D(H)/m^2 at temperatures of 403 and 533 K. A major portion of deuterium initially accumulated in W is released under subsequent exposure to the H plasma. At T_{exp} = 403 K, the deuterium depletion is found at depths up to 3 μm, whereas at 533 K release of deuterium occurs at depths up to 8 μm. Seeding of He ions into the H plasma at T_{exp} = 403 K reduces slightly the deuterium depletion, while at T_{exp} = 533 K the reduction in deuterium depletion becomes negligible.

Acknowledgements

The authors would like to thank J. Dorner and M. Fußeder (IPP Garching, Germany) for technical assistance with the ^{3}He beam analyses, G. Matern for scanning electron microscopy, and Dr. K. Isobe (JAEA Tokai, Japan) for providing W samples.

References

[1] G. Federici, C.H. Skinner, J.N. Brooks, J.P. Coad, C. Grisolia, A.A. Haasz, A. Hassanein, V. Philipps, C.S. Pitcher, J. Roth, W.R. Wampler, D.G. Whyte, Nucl. Fusion 41 (2001) 1967.

[2] K. Tobita, S. Nishio, M. Enoeda, M. Sato, T. Isono, S. Sakurai, H. Nakamura, S.Sato, S. Suzuki, M. Ando, K. Ezato, T. Hayashi, T. Hayashi, T. Hirose , T. Inoue, Y. Kawamura, N. Koizumi, Y. Kudo, R. Kurihara, T. Kuroda, M. Matsukawa, K. Mouri, Y. Nakamura, M. Nishi, Y. Nomoto, J. Ohmori, N. Oyama, K. Sakamoto, T. Suzuki, M. Takechi, H. Tanigawa, K. Tsuchiya, D. Tsuru, Fusion Eng. Des. 81 (2006) 1151.

[3] C.H. Skinner, A.A. Haasz, V.Kh. Alimov, N. Bekris, R.A. Causey, R.E.H. Clark, J.P. Coad, J.W. Davis, R.P. Doerner, M. Mayer, A. Pisarev, J. Roth, T. Tanabe, Fusion Sci. Technol. 54 (2008) 891.

[4] V.Kh. Alimov, W.M. Shu, J. Roth, S. Lindig, M. Balden, K. Isobe, T. Yamanishi, J. Nucl. Mater. 417 (2011) 572.

[5] V.Kh. Alimov, B. Tyburska-Püschel, M.H.J. ’t Hoen, J. Roth, Y. Hatano, K. Isobe, M. Matsuyama, T. Yamanishi, Phys. Scr. T 145 (2011) 014037.

[6] J. Roth, T. Schwarz-Selinger, V.Kh. Alimov, E.A. Markina, J. Nucl. Mater. 432 (2013) 341.

[7] V.Kh. Alimov, W.M. Shu, J. Roth, K. Sugiyama, S. Lindig, M. Balden, K. Isobe, T. Yamanishi, Physica Scripta, T138 (2009) 014048.

[8] M.J. Baldwin, R.P. Doerner, W.R. Wampler, D. Nishijima, T. Lynch and M. Miyamoto, Nucl. Fusion 51 (2011) 103021

[9] V.Kh. Alimov, B. Tyburska-Püschel, Y. Hatano, J. Roth, K. Isobe, M. Matsuyama, T. Yamanishi, J. Nucl. Mater. 420 (2012) 370.

[10] Y. Hatano, M. Shimada, V.Kh. Alimov, J. Shi, M. Hara, T. Nozaki, Y. Oya, M. Kobayashi, K. Okuno, T. Oda, G. Cao, N. Yoshida, N. Futagami, K. Sugiyama, J. Roth, B. Tyburska-Püschel, J. Dorner, I. Takagi, M. Hatakeyama, H. Kurishita, M.A. Sokolov, J. Nucl. Mater. 438 (2013) S114

[11] V.Kh. Alimov, B. Tyburska-Püschel, S. Lindig, Y. Hatano, M. Balden, J. Roth, K. Isobe, M. Matsuyama, T. Yamanishi, J. Nucl. Mater. 420 (2012) 519.

[12] M. Mayer, SIMNRA User's Guide, Tech. Rep. IPP 9/113, Max-Planck-Institut für Plasmaphysik, Garching, 1997

[13] K. Schmid and U. von Toussaint, Nucl. Instr. Meth. B 281 (2012) 64.

[14] H. Iwakiri, K. Yasunaga, K. Morishita, N. Yoshida, J. Nucl. Mater. 283-287 (2000) 1134.

[15] Y. Ueda, M. Fukumoto, J. Yoshida, Y. Ohtsuka, R. Akiyoshi, H. Iwakiri, N. Yoshida, J. Nucl. Mater. 386-388 (2009) 725.

[16] B.I. Khripunov, V.V. Shapkin, V.B. Petrov, N.V. Antonov, Fusion Technology 1994, Pages 243–246 (1995)

[17] Lee, H. T. Haasz, A. A. Davis, J. W. Macaulay-Newcombe, R. G. Whyte, D. G. Wright, G. M. J. Nucl. Mater. 898-903 (2007) 363

Figure captures

Figure 1. Depth profiles of deuterium retained in recrystallized W exposed at 403 K to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with the same fluence of 3.3×10^{24} D(H)/m^2.

Figure 2. Depth profiles of deuterium retained in recrystallized W exposed at 553 K to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with the same fluence of 4.1×10^{24} D(H)/m^2.

Figure 3. Amount of deuterium retained at depths of up to 8 μm in recrystallized W exposed at the same temperature to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with fluences of 3.3×10^{24} D(H)/m^2 (T_{exp} = 403 K) and 4.1×10^{24} D(H)/m^2 (T_{exp} = 533 K), as a function of the exposure temperature.

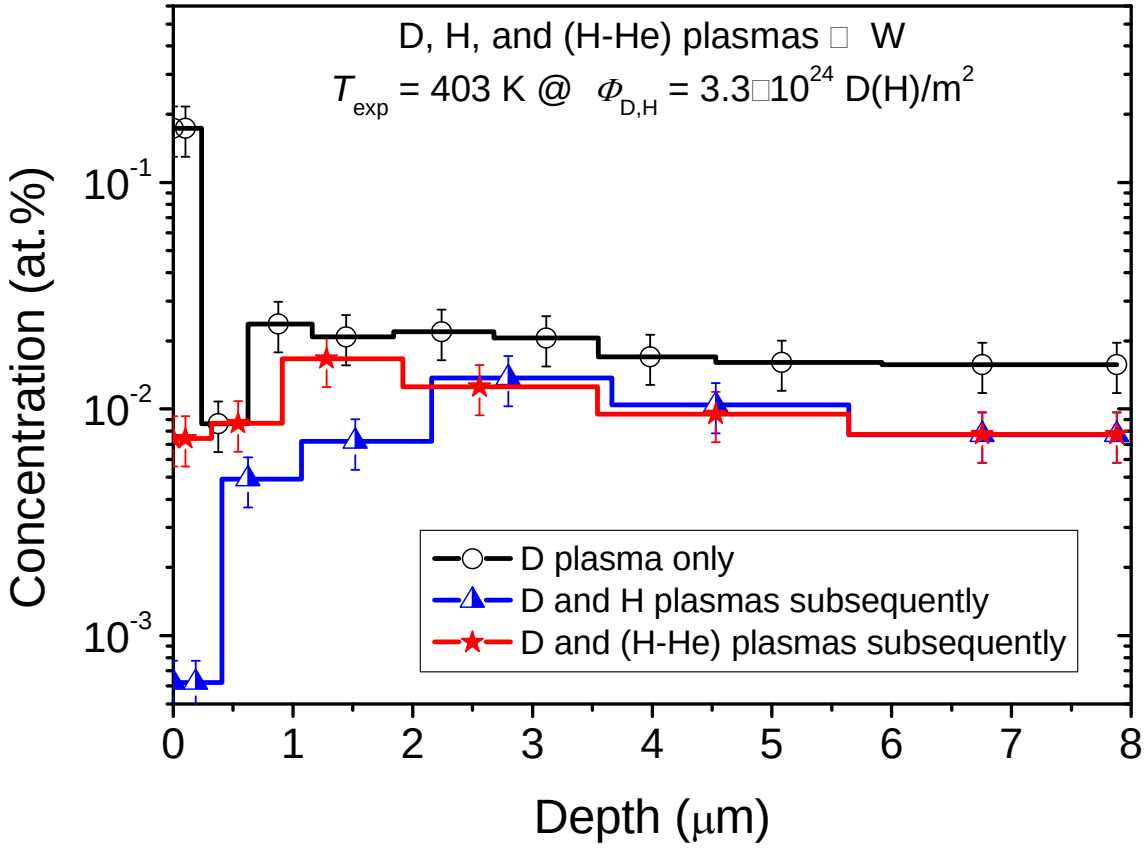

Figure 1. Depth profiles of deuterium retained in recrystallized W exposed at 403 K to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with the same fluence of 3.3×10^{24} D(H)/m^2.

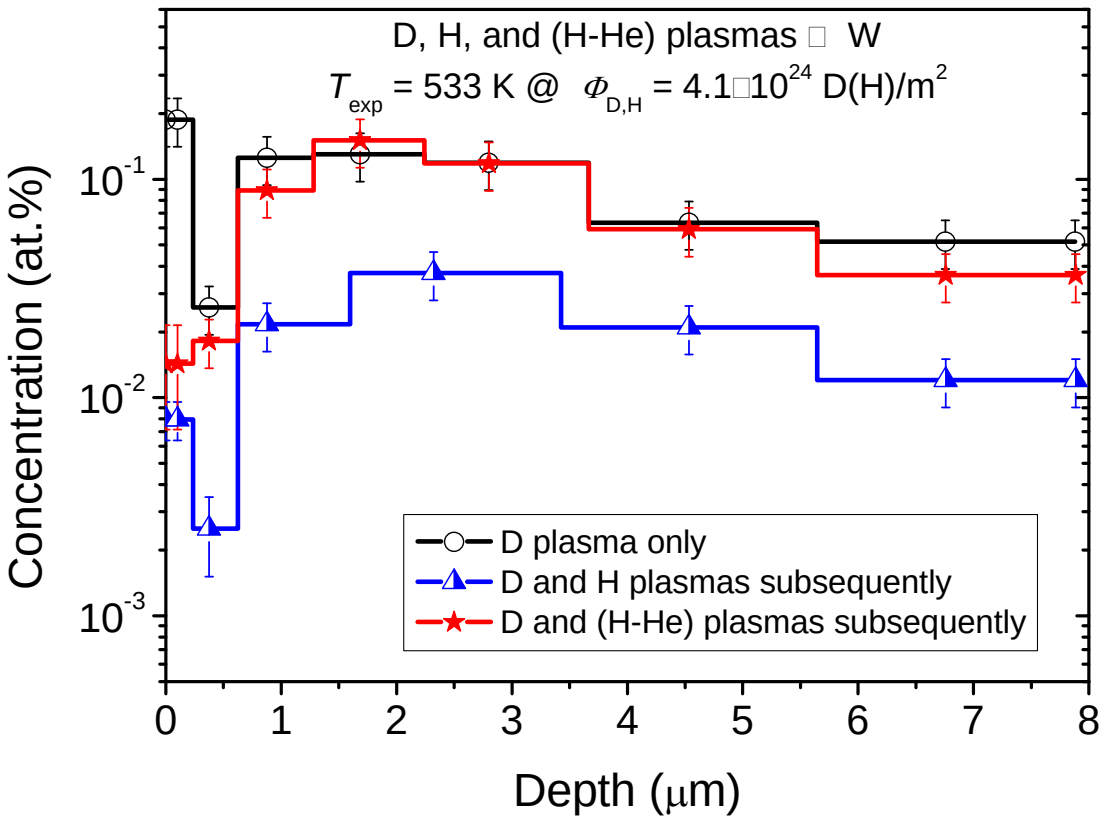

Figure 2. Depth profiles of deuterium retained in recrystallized W exposed at 553 K to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with the same fluence of 4.1×10^{24} D(H)/m^2.

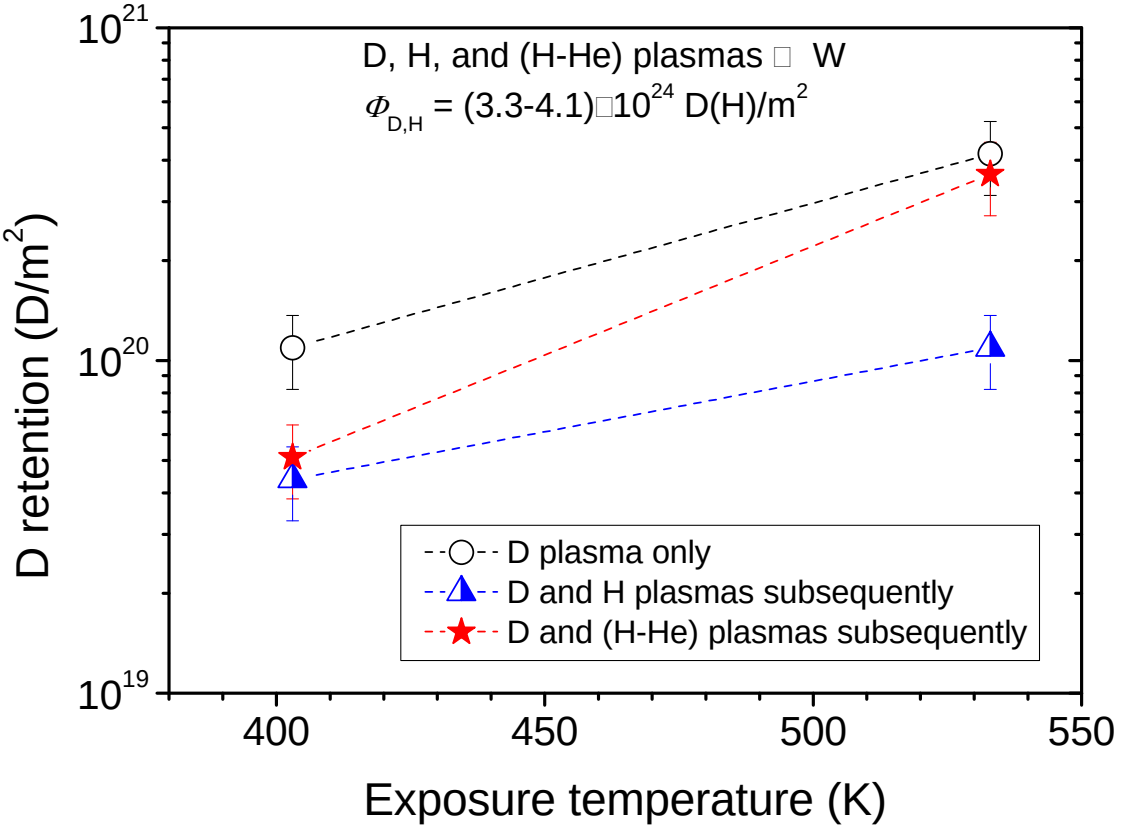

Figure 3. Amount of deuterium retained at depths of up to 8 μm in recrystallized W exposed at the same temperature to D plasma only, to D and H plasmas sequentially, and to D and (H-He) plasma sequentially with fluences of 3.3×10^{24} D(H)/m^2 (T_{exp} = 403 K) and 4.1×10^{24} D(H)/m^2 (T_{exp} = 533 K), as a function of the exposure temperature.