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Retention in tungsten resulting from extremely high fluence plasma exposure

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ABSTRACT

PISCES-B was used for a series of high-fluence plasma exposures to investigate the deuterium fuel retention properties of tungsten, when exposed to continuous plasma irradiation. The goal was to determine whether the fuel retention in the tungsten saturates at sufficiently high fluence, or continues to increase as a function of the plasma fluence. During pure deuterium plasma exposure, up to a maximum deuterium fluence of 2×10^{28} m^{-2}, retention results indicate that saturation is not reached and that retention scales as the square root of time, indicative of diffusion dominating the fuel uptake of the tungsten. However, measurements performed while sculpting the PISCES plasma to replicate a burning plasma, by adding a small amount (5%) of helium ions to the incident deuterium plasma, indicate the deuterium uptake in the target is severely inhibited.

Introduction

Retention of hydrogen isotopes in tungsten plasma-facing components is a concern for future burning plasma devices using tritium as a part of its fuel cycle. The concern is two-fold; first, the amount of trapped tritium affects the required tritium breeding ratio necessary to operate the reactor, and secondly, from the viewpoint of safety during an unexpected accident scenario. While much data is available in the literature on retention of hydrogen isotopes in plasma exposed tungsten, most of this data was obtained at fluences below 10^{26} m^{-2}. In a power-producing reactor, where one expects to operate in steady state, one would exceed a fluence of 10^{31} m^{-2} during a year of operation.

The question of the extrapolation of retention to higher values of fluence was one of the topics of a meeting held at the Plasma Science and Fusion Center at MIT in 2010. At the time, the data presented at the meeting (and included in the meeting report [1]) suggested the possibility of saturation in retention with increasing fluence, but the data was by no means conclusive. Fig. 1 reproduces the data set of retention vs. fluence contained in the MIT report along with upper and lower bounds of the retention that were arrived at during the meeting. Since that time, additional data has become available, but the fluence remained below 10^{27} m^{-2} and did not settle the issue of saturation with increasing fluence. To address this issue the PISCES-B device [2] performed a fluence scan, using identical plasma conditions and only altering the time of the plasma exposure to obtain the change in fluence.

Description of experiment

The first issue to address in designing the experimental exposure condition was to specify the sample temperature during the exposure. The data collected in [1] was obtained at 500 K, but this temperature is close to the region where blisters have been observed in tungsten exposed to deuterium plasma [3,4] and it was believed that a slightly higher temperature would avoid the issue of blisters and provide a more definitive result. The competing effect was the decrease in retention with increasing temperature [3,4], so it was decided to increase temperature only slightly to avoid blisters, yet still maximize desorption signals. As the exposure temperature increases, the diffusion distance also increases and the concern arises as to the thickness of the sample needed to prevent the diffusion front from reaching the back surface of the sample during the extremely long exposure times planned. It was also necessary not to exceed the maximum sample thickness compatible with the existing sample manipulator in PISCES-B, which is 1 cm.

The operational temperature was therefore selected by extrapolating Frauenfelder's diffusivity [5] to lower temperature to

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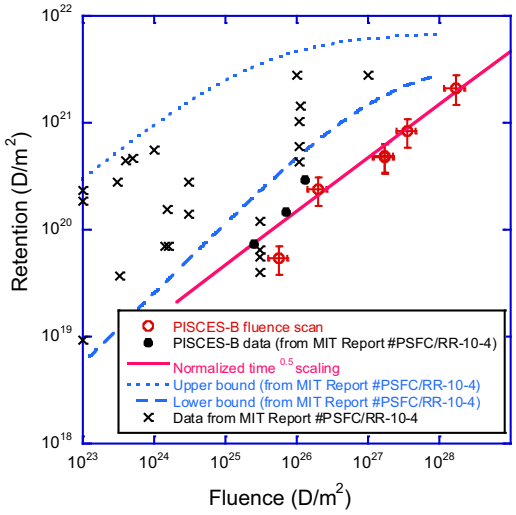

Fig. 1. Retention in W during varying fluence deuterium exposures showing data contained in [1, reproduced with permission] and the present high-fluence scan. Also shown are bounding limits derived in [1] and the square root of time scaling of the present data.

ensure that the diffusion distance during a 40 h long exposure remained less than 1 cm. Frauenfelder's diffusivity extrapolates to approximately 10^{-10} m^2/s at 640 K, which would lead to a diffusion length of 7.4 mm after 40 h. To ensure good thermal connection on the threaded sample clamping mechanism in PISCES-B it was decided to use 8 mm thick samples for the longest exposures in the fluence scan and to use standard 1 mm thick samples for the shorter exposures in the scan. Both thickness samples were used at a mid-range point in the fluence scan (i.e. 2×10^{27} m^{-2}) to compare the results from both sample sizes. All samples were ITER grade tungsten made by Midwest Tungsten Service, Inc. via powder metallurgy by press-sintering 99.95% pure tungsten powder. The tungsten grains are elongated in the direction normal to the surface and all samples were polished using a 3 micron diamond suspension fluid. The samples were cleaned and outgassed after polishing to 1273 K for one hour before the plasma exposure.

A Langmuir probe was used to measure deuterium plasma parameters during the exposures and to ensure similarity between the exposures. The probe measured $T_e \sim 3$ eV and $n_e \sim 1.4\times10^{19}$ m^{-3}, resulting in an ion flux of 1×10^{23} m^{-2}s^{-1}. The ion energy was set to 50 eV by biasing the sample negatively. Since the plasma in the PISCES devices is comprised of a mixture of D^+, D_2^+ and D_3^+, a model [6] was used to estimate the molecular ion mix in the plasma giving 0.55, 0.3, 0.15, respectively. This species mix means that the energetic deuterium particle flux to the target was 1.6×10^{23} m^{-2}s^{-1}. The exposure time was then adjusted to scan the fluence to the samples from 3×10^{25} m^{-2} up to 2×10^{28} m^{-2}. In order to reach the highest fluence data point, the PISCES-B device operated continuously for over 30 h.

Retention in the samples was measured by thermal desorption spectroscopy (TDS). The sample temperature was increased at 0.3 K/s by radiative heating in a quartz bell jar up to 1340 K. Mass to charge ratios 3 (HD) and 4 (D_2) were used in the analysis of the retained deuterium. Deuterium released through the signals related to mass to charge ratios 19 (HDO) and 20 (D_2O) have been ignored since the intensity of those peaks are typically only a percent or less of the D_2 peak. A high resolution quadrupole mass spectrometer capable of discriminating between He and D_2 signals was also used. The signals were calibrated using an absolutely calibrated D_2 leak. HD was assumed to have the same calibration as D_2. SEM images of the surfaces after exposure were also collected.

Results and discussion

The data for the fluence dependence of deuterium in W from PISCES-B is also plotted in Fig. 1. The results clearly show an increase with increasing fluence. The slope of the increase is consistent with the square root of time, indicating that no saturation of the samples has occurred. A few other less obvious conclusions from the data in Fig. 1 can also be observed. The first is the retention data at a fluence of 2×10^{27} m^{-2} showed no difference between the thin and thick samples. This implies that the diffusion front did not reach the back surface of the 1 mm thick sample during the 12,000 s plasma exposure needed to reach that fluence. Using Fick's Law, $\lambda^2 = 4Dt$, allows us to calculate a maximum diffusion coefficient of 2×10^{-11} m^2/s, which is much smaller than the extrapolated value from Frauenfelder's [5] measurements that was mentioned previously. This reduction in diffusion occurs because simply extrapolating Frauenfelder's data, which was collected at high temperature to avoid the influence of trapping, to low temperature overestimates diffusion. The overestimation comes from the influence of de-trapping and trapping which can slow diffusion substantially.

The second observation is the apparent reduction in retention as the flux during the plasma exposure increases. The PISCES-B device operates at a flux above 10^{23} m^{-2}s^{-1}, whereas much of the data presented in the MIT report was collect at fluxes of a few x 10^{22} m^{-2}s^{-1}. A similar dependence of retention on flux has also recently been observed and reported [7].

SEM images of the sample surfaces following the deuterium plasma exposure are shown in Fig. 2 as a function of fluence to the samples; a) 6.8×10^{25} m^{-2}, b) 2.4×10^{26} m^{-2}, c) 2×10^{27} m^{-2} and d) 2×10^{28} m^{-2}. It is possible that the higher fluence surfaces are exhibiting small blisters on the surface, although to be certain higher resolution SEM images of the surfaces would be required. Unfortunately, due to potential beryllium contamination from PISCES-B such examination is not possible. However, recent work has shown an increase in the temperature at which blisters form on tungsten surfaces as the deuterium plasma flux is increased [8]. It is possible that during these high flux deuterium plasma exposures, the selected operating temperature of 640 K was not sufficiently high to avoid blister formation.

Representative D_2 desorption curves are plotted in Fig. 3, from the three thick samples. As can be seen in the figure, a single desorption peak dominates the release. As the fluence (i.e. time) of each exposure increases, the maximum in the release peak shifts to higher temperature. This is indicative of deeper diffusion into the sample during each exposure and hence slower release from each sample.

A final set of measurements were performed in PISCES-B to make the data set more relevant to a burning plasma device, and specifically to ITER. A similar deuterium plasma was seeded to contain 5% He ions in the background deuterium plasma to simulate the effects of He ash being entrained in the plasma flux encountering a tungsten surface. In PISCES-B the helium ion concentration in the plasma is continuously monitored spectroscopically, more details of the technique can be found in [9]. The retention results from these plasma exposures revealed no measurable deuterium retention when the plasma contained helium ions, consistent with previous measurements in PISCES [4]. These results are plotted along with the pure deuterium plasma exposure data in Fig. 4. Also included in this figure is the upper axis which has been replotted in terms of cumulative ITER plasma time.

In pure helium plasma, as well as in mixed D/He plasma, a dense array of small (1–2 nm diameter) nano-bubbles form within 20–30 nm of the plasma exposed surface [10–12]. These nano-bubbles are thought to inhibit deuterium migration into the underlying tungsten by one of two methods. The first mechanism

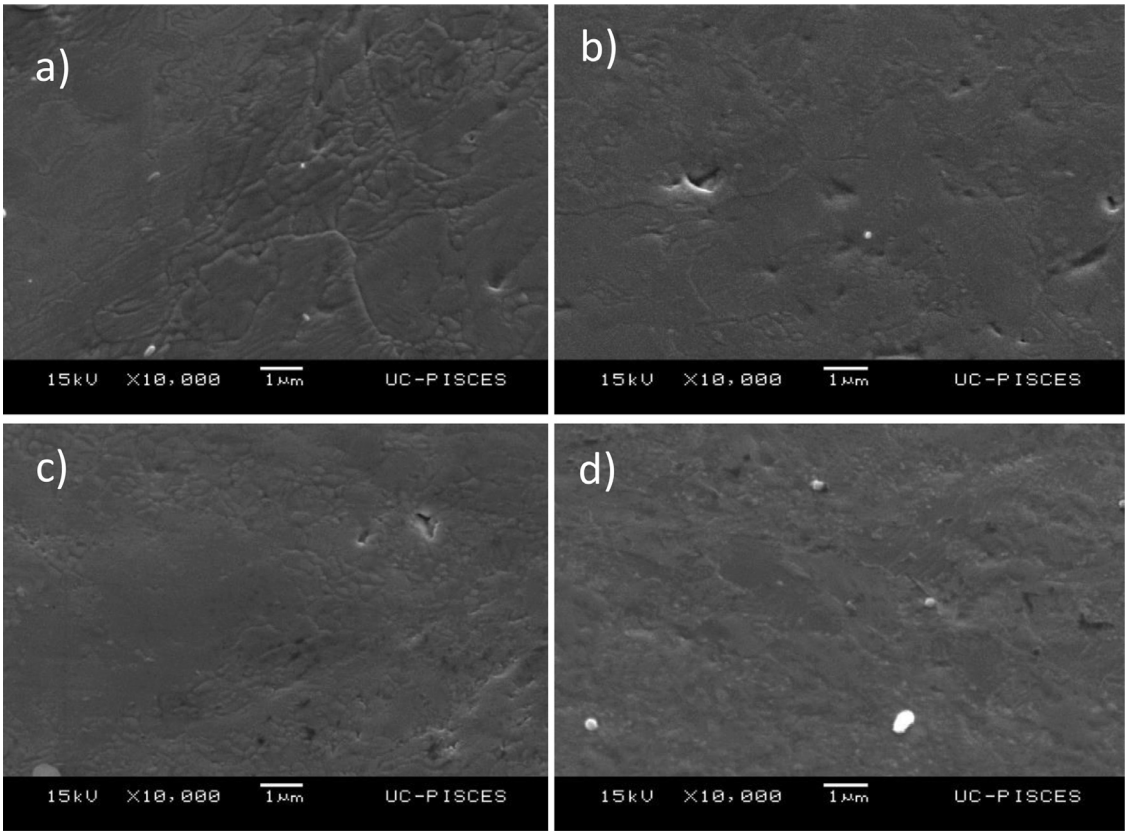

Fig. 2. SEM images of sample surfaces after deuterium plasma exposure; (a) 6.8×10^{25} m^{-2}, (b) 2.4×10^{26} m^{-2}, (c) 2×10^{27} m^{-2} and (d) 2×10^{28} m^{-2}.

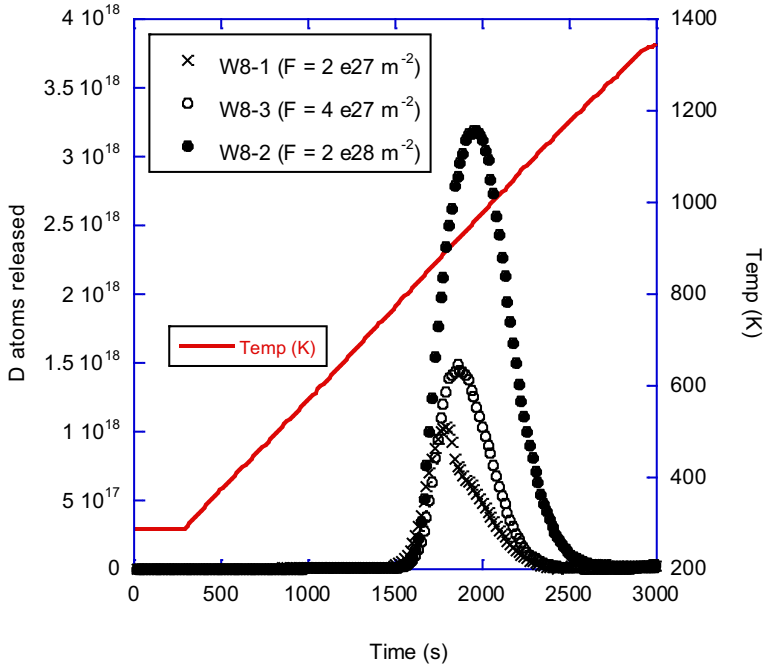

Fig. 3. Thermal D_2 release behavior and temperature ramp of samples exposed to deuterium plasma for varying fluences; 2×10^{27}, 4×10^{27} and 2×10^{28} m^{-2}.

that could be responsible for the reduction in retention is the interconnection of these bubbles which could provide a pathway back to the surface for deuterium atoms that recombine of the surfaces of the bubbles. The second idea is that the pressurized bubbles themselves create a stress field within the tungsten lattice that acts to trap any diffusing deuterium in the region of tungsten near the surface of the bubbles [13]. These measurements confirm the reduction of deuterium retention in tungsten at high fluence due to the presence of helium. However, separate experiments will need to be performed to examine which of the possible mechanisms described above are responsible for the reduction.

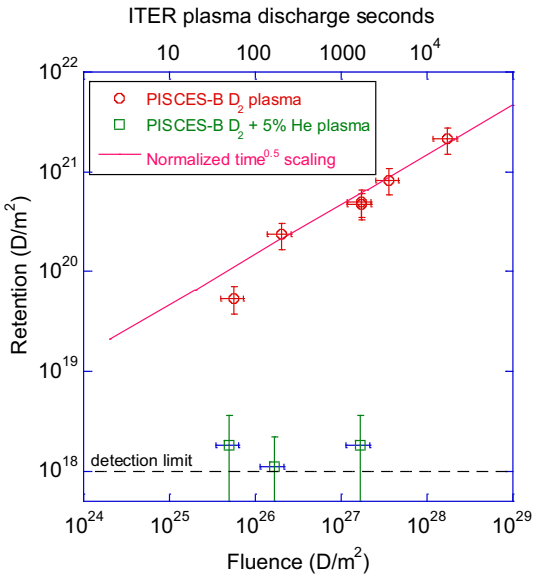

Fig. 4. Comparison of retention from samples exposed in PISCES-B to pure deuterium plasma and mixed deuterium + 5% helium plasma.

Summary

ITER grade tungsten samples were subjected to varying fluence deuterium and deuterium + 5% helium plasma in the PISCES-B device. The fluence variation was achieved by keeping nearly constant plasma conditions and varying the exposure time. A maximum fluence of 2×10^{28} m^{-2} was obtained by operating the device for over 30 h without interruption. For the deuterium plasma exposure, the retention scaled with a square root of time dependence, with no evidence of saturation. For the mixed deuterium/helium plasma exposures, the deuterium desorption from the samples was not above the limit of detection of the desorption system.

Acknowledgments

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References

[1] B. Lipshultz, J. Roth, J. Davis, R.P. Doerner, A.A. Haasz, A. Kalenbach, et al., An assessment of the current data affecting tritium retention and its use to project towards T retention in ITER, MIT report (#PSFC/RR-10-4).
[2] R.P. Doerner, M.J. Baldwin, K. Schmid, The influence of a beryllium containing plasma on the evolution of a mixed-material surface, Phys. Scr. T111 (2004) 75.
[3] V.Kh. Alimov, W.M. Shu, J. Roth, K. Sugiyama, S. Lindig, M. Balden, et al., Surface morphology and deuterium retention in tungsten exposed to low-energy, high flux pure and helium-seeded deuterium plasmas, Phys. Scr. T138 (2009) 014048.
[4] M.J. Baldwin, R.P. Doerner, W.R. Wampler, D. Nishijima, T. Lynch, M. Miyamoto, Effect of He on D retention in W exposed to low-energy, high fluence (D, He, Ar) mixture plasmas, Nucl. Fusion 51 (2011) 103021.
[5] R. Frauenfelder, Solution and diffusion of hydrogen in tungsten, J. Vac. Sci. Technol. 6 (1969) 388.
[6] E.M. Hollmann, A.Yu. Pigarov, Measurement and modeling of molecular ion concentrations in a hydrogen reflex-arc discharge, Phys. Plasmas 9 (2002) 4330.
[7] L. Buzi, G. De Temmerman, B. Unterberg, M. Reinhart, A. Litnovsky, V. Philipps, et al., Influence of particle flux density and temperature on surface modifications of tungsten and deuterium retention, J. Nucl. Mater. 455 (2014) 316.
[8] L. Buzi, G. De Temmerman, B. Unterberg, M. Reinhart, T. Dittmar, D. Matveev, et al., Influence of tungsten microstructure and ion flux on deuterium plasma-induced surface modifications and deuterium retention, J. Nucl. Mater. 463 (2015) 320.
[9] D. Nishijima, R.P. Doerner, M.J. Baldwin, E.M. Hollmann, R.P. Seraydarian, Y. Ueda, Spectroscopic determination of the singly ionized helium density in low temperature plasmas mixed with helium in a linear divertor plasma simulator, Phys. Plasmas 14 (2007) 103509.
[10] M. Miyamoto, D. Nishijima, M.J. Baldwin, R.P. Doerner, Y. Ueda, K. Yasunaga, et al., Microscopic damage of tungsten exposed to deuterium-helium mixture plasma in PISCES and its impact on retention property, J. Nucl. Mater. 415(2011)S657.
[11] M. Thompson, P. Kluth, R.P. Doerner, N. Kirby, C. Corr, Probing helium nano-bubble formation in tungsten with grazing incidence small angle x-ray scattering, Nucl. Fusion 55 (2015) 042001.
[12] R.P. Doerner, D.L. Rudakov, C.P. Chrobak, A.R. Briesemeister, C. Corr, G. De, et al., Investigation of He-W interactions using DiMES in DIII-D, Phys. Scr. T167 (2016) 014054.
[13] N. Juslin and B.D. Wirth, J. Nucl. Mater. 438(2013)S1221.