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Retention behavior in tungsten and molybdenum exposed to high fluences of deuterium ions in TPE

J.P. Sharpe [a,*], R.D. Kolasinski [b], M. Shimada [a], P. Calderoni [a], R.A. Causey [b]

[a] Fusion Safety Program, Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415, USA
[b] Hydrogen and Metallurgical Sciences Group, Sandia National Laboratories, Livermore, CA 94551, USA

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ABSTRACT

The Tritium Plasma Experiment (TPE) has been used to investigate deuterium fuel retention behavior in tungsten and molybdenum-materials utilized for plasma-facing surfaces in some existing tokamak plasma devices and under consideration for future devices. Although several studies have been performed over the past several years on these metals, many issues remain unresolved, including for example blister formation mechanisms and correlation to surface conditions. In this study, we expose several metal samples to deuterium ion fluences up to 10^{26} ions/m^2 and measure retention behavior with thermal desorption spectroscopy. Fractional retention of up to 2.0×10^{-5} is found for W at 600 K, and Mo similarly retains deuterium at a fraction of 1.5×10^{-5} at 600 K. Blistering was found for W samples exposed at temperatures above 453 K, whereas blistering was not observed for Mo samples at any experiment temperature.

1. Introduction

Safe and effective operation of future fusion reactors relies upon materials to contain the plasma and receive the thermal heat load with minimal impact on the fusion fuel by way of retention. As a refractory metal, tungsten is a prime candidate because of highly suitable thermal–mechanical properties and low sputtering yield. Molybdenum is of interest as a comparative material due to the metal's similar properties to tungsten and its use in some fusion plasma research devices (e.g. Alcator C-MOD).

The Tritium Plasma Experiment (TPE) is a linear column plasma device suitable for materials exposures in plasma conditions approaching those of a fusion reactor first wall. TPE is capable of producing steady-state plasmas composed of various hydrogen isotope mixtures – including tritium – and testing at high fluences allows characterization of a wide range of materials for fusion fuel retention and permeation behavior. This paper summarizes recent results of deuterium retention experiments in TPE with W and Mo.

2. TPE experimental conditions

Despite the large body of work that exists for H-isotope retention in fusion materials, there is generally a wide disparity between results obtained in different laboratory settings with materials prepared in different ways. This suggests the need for adequate characterization and comparison of the experimental conditions. TPE has been refurbished and incorporates additional diagnostics for plasma-surface exposure conditions, including a plunging Langmuir probe for target current density and a spectrometer for indication of potential impurities. An ex situ vacuum thermal desorption spectroscopy system was configured to measure H-isotope retention in exposed samples; this separate system is located outside of the TPE glovebox and requires sample transfer in air from the TPE chamber to the desorption system chamber. Sample preparation and experimental arrangements for W and Mo exposures are outlined below.

2.1. Sample preparation

Both the Mo and W targets used in this study were 2.54 cm diameter discs cut from thin sheets via electrical discharge machining. The 1 mm thick W raw material (obtained from PLANSEE) was of ultra high purity grade (99.9999% W by weight) and was hot-rolled into its final form. The Mo material was obtained from Eagle Alloys Corp. in 2 mm thick sheets, with a purity level of 99.95% (in accordance with ASTM B 386 standards).

The samples were polished on one side to a surface finish with an RMS roughness of 0.02 μm. To recrystallize the surface and relieve stress after polishing, the samples were annealed under UHV conditions, as discussed later in this section. SEM imagery prior to plasma bombardment indicated an average grain diameter ranging between 2 and 5 μm for W. Energy dispersive X-ray (EDX) analysis results were inconclusive regarding the presence of trace amounts

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[*] Corresponding author.
E-mail address: Phil.Sharpe@inl.gov (J.P. Sharpe).

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of O impurities on the Mo and W samples, and future work will involve using an Auger Electron Spectroscopy (AES) system to characterize such impurities.

2.2. Plasma discharge parameters

Characterization of the TPE plasma conditions has been published elsewhere [1]; we therefore focus on specific observations relevant to this study. The main plasma diagnostic was a pneumatically actuated Langmuir probe, allowing measurement of the radial the electron temperature (T_e) and density (n_e) profiles. The probe tip plunges perpendicularly through the plasma (transverse to the collimating magnetic field) at 150 cm/s, and the data sampling rate yields a spatial resolution of 0.75 cm in swept-bias mode or 0.01 cm in fixed-bias mode. Reported plasma temperatures, densities, and target flux are calculated from average values over the projected sample surface area.

One of the main goals of this work was to observe the effect of varying sample temperature on retention, while holding the incident fluence and flux constant. The target holder heat sink configuration was modified to achieve this effect. For the temperature dependent retention tests, we exposed the samples to an incident flux of 1.1×10^{22} D/m^2 s for ~2 h. This resulted in a total fluence on the order of 10^{26} D/m^2. Incident ion energy of 70 eV was achieved by adjusting the target bias voltage and correcting for the floating potential. For typical TPE operating conditions, plasma parameters ranged as $7 \leqslant T_e \leqslant 15$ eV, and $10^{16} \leqslant n_e \leqslant 10^{18}$ m^{-3}. The incident beam was assumed to be primarily D$^+$, based on prior work with similar instruments such as PISCES-A [2].

2.3. Thermal desorption spectroscopy

After plasma bombardment, the samples were removed from TPE, and the amount of retained D was determined via a thermal desorption spectroscopy (TDS) system. While some exposure to air was unavoidable during this process, a continuous Ar purge served to mitigate any sample contamination during placement in the TDS system. The vacuum facility consisted of an analysis chamber equipped with a Dycor 50 amu residual gas analyzer (RGA). The main chamber was attached to a Pfieffer 70 l/s turbo molecular pump, capable of maintaining a base pressure of 4×10^{-6} Pa (3×10^{-8} torr) after bake-out. The analysis chamber was attached to a single-ended quartz tube enclosed in a cylindrical furnace for sample heating. During this process, a tantalum tray held the sample in place. Two K-type thermocouples monitored the process temperature. Three standard leaks provided an absolute D_2 sensitivity calibration for the RGA.

The annealing process prior to plasma exposure involved heating the samples at rate of 33.3 K/min to a final temperature of 1273 K. This temperature was maintained for 1 h. We used a slower ramp rate of 10 K/min for thermal desorption, and the final temperature of 1273 K was maintained for only 30 min in this case. A background desorption spectrum was established by heating a virgin sample. Only the peaks associated with D_2^+ and HD$^+$ peaks were considered in this analysis, and the same RGA sensitivity was assumed for both [3].

2.4. Error analysis

It is worth considering the uncertainty of the results reported here to allow for an appropriate comparison with other published studies. A major source of error in this study was considered to be the magnitude of the incident ion flux. The uncertainty in this measurement was based on the least squares fit for n_e and T_e. Once the plasma was initiated, the beam was relatively stable over the course of a single test, with typical variations less than 5%. This effect is included in the integration of the total fluence. Another smaller source of error is the accuracy of the TDS system. The calibrated leaks used to determine the desorbed flux during the desorption process had a rated accuracy of ±10%. This is consistent with our observation that over the course of several weeks, the RGA sensitivity did not drift by more than 3%. These error sources are combined using standard error propagation techniques and are represented by the vertical error bars.

The variation in sample temperature over the course of the test represents an error source of at least equal importance to the effects described above. Once the plasma was initiated, ~15–20 min were required for the temperature to steadily approach equilibrium. After this, the temperature increased very gradually over the remainder of the 1 h 40 min portion of the test. The horizontal error bars shown in Figs. 1 and 2 indicate the extent of thermal drift over this time period, and the maximal extent of drift was ~40 K.

3. Retention results and surface morphology effects

The test matrix for this study included 12 exposures with Mo targets and 8 with W targets. Both sets of tests were aimed at characterizing the retention in these materials as a function of sample temperature, T_s. Retention curves are presented in Fig. 1 for Mo and in Fig. 2 for W for $380 \leqslant T_s \leqslant 1000$ K. For these temperatures, the total retention in both metals varied over the same range between 1×10^{18} and 2×10^{21} D/m^2. The molybdenum samples exhibited high retention for $T_s \leqslant 410$ K. A sharp drop in retained fluence occurs shortly thereafter, reaching a local minimum at $T_s = 450$ K. After this point, the curves gradually increases until a peak value is achieved at $T_s = 650$ K. The retention behavior for W was somewhat simpler than that of Mo, as shown in Fig. 2. In this case, retention increases with temperature gradually until a peak value is achieved between $600 \leqslant T_s \leqslant 650$ K. The fraction retained generally varied from 10^{-8} to 5×10^{-4} for both metals.

Overlays of selected TDS spectra are presented in Fig. 3 for Mo and in Fig. 4 for W. The spectra correspond to specific sample exposure temperatures comprising data in Figs. 1 and 2. For both metals, at least three distinct desorption peaks were observed. For Mo, these occurred at $T_d = 600$, 750, and 1100 K. Likewise for W,

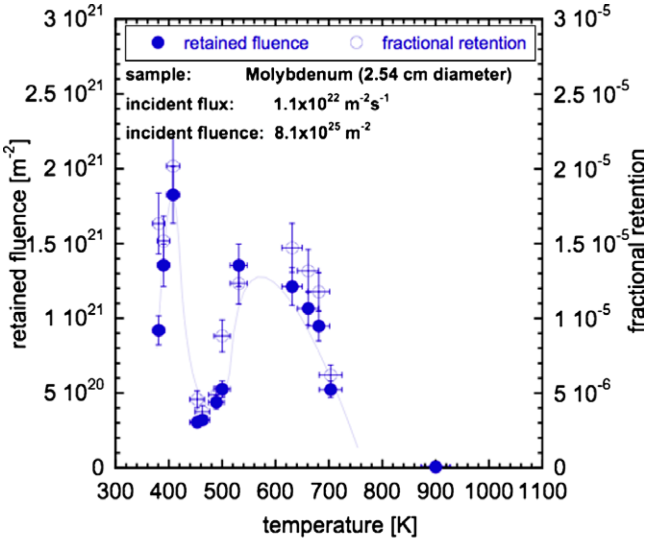

Fig. 1. Retained fluence and fractional retention as a function of temperature for Mo samples bombarded with 70 eV D$^+$ ions. For each exposure, the ion flux was 1.1×10^{22} D/m^2 s, and the total fluence was 8.1×10^{25} D/m^2. TDS spectra for the highlighted cases are shown in Fig. 3.

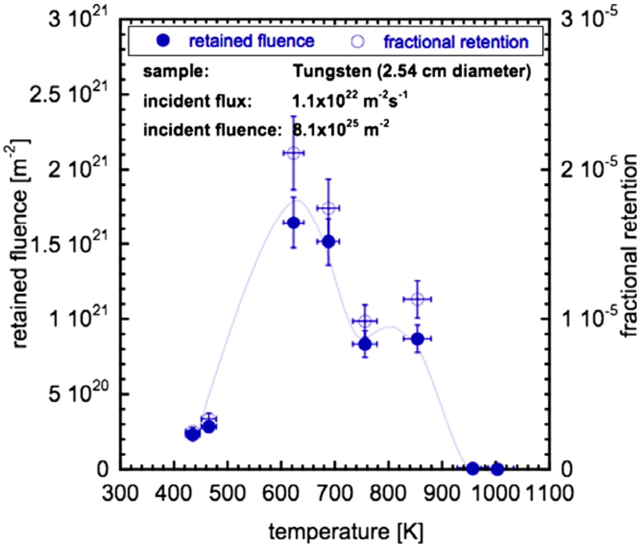

Fig. 2. Retention behavior of W for varying sample temperature (plasma conditions for these tests were identical to those for Mo).

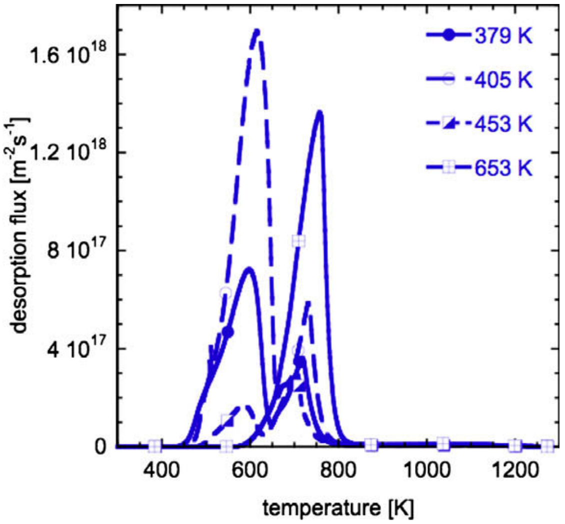

Fig. 3. Evolution of TDS spectra as a function of sample temperature for Mo samples.

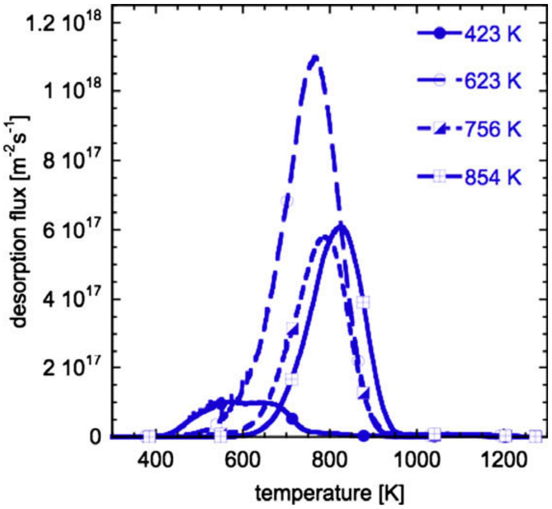

Fig. 4. Evolution of TDS spectra as a function of sample temperature for W samples.

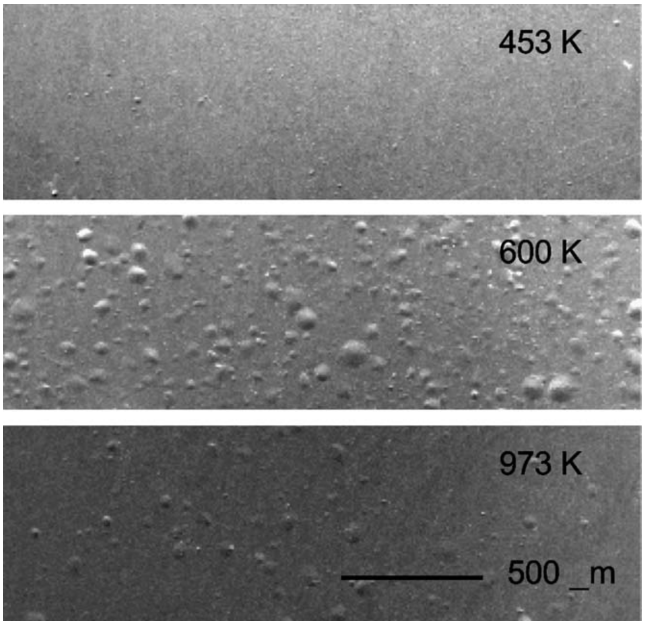

Fig. 5. SEM images showing blistering of the W surface after plasma exposure, for several different temperatures.

desorption peaks emerged at T_d = 650, 800, and 1100 K. The peaks at 1100 K are hidden from view in Figs. 3 and 4 because the resulting desorption flux is plotted on a linear scale to better demonstrate the spectral structure of the lower-temperature peaks. For each set of tests, a gradual variation was observed in the structure of the TDS spectra, where the three dominant peaks shift to slightly higher values for samples of increasing exposure temperature. For example, the actual values of the peak characterized as T_{d} = 800 K for W vary from 770 K for the sample exposed at 623 K to 820 K for the sample exposed at 854 K. A similar behavior was observed for the Mo samples. The source of these shifts remain unclear, although one may envision several possible mechanisms: different surface oxidation conditions among samples, detrapping from varying depths within the bulk, or even multiple traps leading to peak pileup. Unfortunately, the existing data inadequately elucidate the contributions of such mechanisms because temperature drift during sample exposure introduces uncertainty in the thermal desorption spectral response. Efforts to improve sample thermal control in TPE are underway, and future experiments will attempt to reproduce and discern the underlying mechanisms responsible for this behavior. Additionally, during the W desorptions, we observed small spikes in the measured D_2^+ partial pressure. These occurred only for W samples exposed at intermediate temperatures $(450 \leqslant T_s \leqslant 700\ \mathrm{K})$.

We used images from both an SEM and an optical microscope to analyze changes in the surface structure as a function of temperature. The Mo samples showed no indication of blistering, with the only visible surface modification being the presence of arc tracks. This was in striking contrast to the W samples which showed significant blistering, with the size and quantity of blisters varying with temperature. A collection of SEM images showing the change in W surface morphology are shown in Fig. 5.

4. Discussion

There is a considerable published database of experimental results for D retention in W. For the temperature dependence, we consider recent studies led by Causey et al. [4], Venhaus et al. [5], and Alimov et al. [6]. The results reported in Refs. [4,5] were performed in TPE during its tenure at LANL, also with PLANSEE samples annealed at 1273 K. In these studies, the targets were exposed to an ion flux of 8.7×10^{21} $(D + T)/m^2$ s for a duration of 1 h, resulting in a total fluence that was ~40% of that achieved in this work. In Ref. [5], the peak in retained fluence for these conditions is estimated to be 5.0×10^{20} $(D + T)/m^2$, occurring at $T_s \approx 623$ K. Adjusting for the 40% difference in total fluence, the retention values discussed in Refs. [4,5] are in reasonable agreement with the results reported in our study. The data of Alimov also show a similar variation in retention with temperature, with the peak in retention varying between $500 \leqslant T_s \leqslant 700$ K depending on the C content of the incident plasma. The TDS peaks for W shown in Fig. 4 match the general structure of those observed in Ref. [4]. Differences in TDS peak locations and intensities are likely due to differences in thermal ramp rate and sample preparation.

The retention behavior in Mo has in general received less attention than W, although recent results from Alcator C-MOD have heightened interest in this area. Laboratory studies conducted in PISCES-A by Goebel et al. in Ref. [7] and in a Penning discharge by Causey et al. in Ref. [8], report a retention fraction which varies between 10^{-4} and 10^{-6} depending on sample temperature. Our data appear to be in satisfactory agreement with these studies in this respect. In Ref. [9], Wright discusses in situ measurements of ‘dynamic retention’ in Mo using DIONISOS. Measurements of D inventory indicate a fractional retention of $\sim 10^{-4}$, at the higher range of most other laboratory experiments. None of the laboratory studies are yet able to account for the large retention fraction observed in the Alcator C-MOD reactor (on the order of 10^{-3}, based on wall pumping tests [9]). Additionally, the temperature-dependence found for Mo exposures in TPE, as shown in Fig. 1, is not commonly observed in other studies. Ongoing investigation with TPE may elucidate the underlying cause; we will start by modifying the characteristics of sample preparation and annealing, in a similar approach presented by Tanabe et al. in Ref. [10] for Mo retention studies with 25 keV deuterium ions.

Exposing W samples to the TPE plasma resulted in blister formation. At sample temperatures ranging between 600 and 850 K, the blister size and coverage fraction was found to strongly depend on sample temperature, as demonstrated in the series of images in Fig. 5. At peak conditions, blisters as large as 100 μm in diameter were observed. Lower temperature exposures were characterized by blister diameters of less than 10 μm.

5. Concluding remarks

Deuterium retention measurements performed with TPE in W and Mo indicate consistent behavior compared to previous studies, however, details of material fabrication and preparation likely result in important differences. Fraction of retained flux for Mo shows an intriguing behavior at lower temperatures and is perhaps related to the dynamic retention observed in Alcator C-MOD. Retention behavior of W concurs with previous work, although the onset of blistering occurs with samples prepared in a similar fashion to samples of previous experiments in which blistering did not occur. Further experiments are underway to elucidate the details of W blistering during high flux plasma exposure.

Acknowledgments

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