Nuclear Instruments and Methods in Physics Research B xxx (2014) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Deuterium retention in tungsten after heavy ion damage and hydrogen isotope exchange in PISCES

J.L. Barton [a,*], Y.Q. Wang [b], T. Dittmar [a], R.P. Doerner [a], G.R. Tynan [a]

[a] University of California, San Diego, 9500 Gilman Drive #0417, La Jolla, CA 92093, USA
[b] Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545, USA

ARTICLE INFO

Article history:
Available online xxxx

Keywords:
Deuterium inventory
Tritium inventory
Tungsten
Ion irradiation
Nuclear reaction analysis

ABSTRACT

The effect of H isotope exchange and radiation damage on the retention of D in W was examined in the PISCES linear plasma device. W samples were treated with D plasma at low sample temperatures (473 K), with a fluence of 10^{26} ions/m^2 and ion energies of 150 eV. Each sample was then exposed to varying doses of H plasma with similar sample temperature and plasma conditions to fluences ranging from 0 to 10^{26} ions/m^2, to examine the effectiveness of isotope exchange as a means of tritium removal. The D(^{3}He, p)^{4}He nuclear reaction was used to measure D concentration profiles up to a depth of 7.7 μm. Thermal desorption spectroscopy (TDS) was used to determine the D retained throughout the bulk of the sample. Isotope exchange allows for a unique study of atomic migration by separately examining the diffusion of implanted atoms from those bombarding the surface. D atoms are exchanged out of traps as a result of H plasma bombardment and diffuse until either falling into another trap or reaching the surface to recombine and escape. Radiation damage at levels of 0.01, 0.1, and 1 displacements per atom (dpa) was carried out before plasma exposure on some samples with 2 MeV Cu ions as a surrogate for damage caused by fusion neutrons. The Cu ion damage was compared to damage induced by 6 MeV W ions to see if there is an effect of Cu contamination on retention. We saw little difference in Cu versus W ion damage at low dpa, but at 1 dpa, where Cu content reached 65 appm, contamination seems to be significant. Retention measurements showed that ion damage has little effectiveness on isotope removal at these sample temperatures; however, there is evidence to suggest that the trapping mechanisms in W change as damage is increased.

Published by Elsevier B.V.

1. Introduction

Tritium retained in the plasma facing components (PFCs) of large magnetically confined nuclear fusion test reactors, such as ITER, pose concerns regarding loss of fusion fuel and radiation safety [1–3]. Methods to reduce trapped H isotopes have been tested [4–6] by exposing D doped W materials to H ions in order to see the effect of D displacement. The goal is to preserve the tokamak walls and divertor plates from reaching the tritium contamination limit [7]. However, isotope exchange experiments also provide interesting new insight into the atomic migration paradigm that trapping and de-trapping of implanted atoms are not just a thermally activated process but also can be released by a mechanical exchange from a diffusing atom.

DT fusion reactions produce 14.1 MeV neutrons, which can impact the PFCs of a working fusion reactor. These high energy collisions cause damage cascades in the lattice that can change the retention properties of the material [8,9]. Since present facilities available cannot replicate neutron bombardment in a fusion reactor and because handling neutron irradiated samples is difficult, heavy ion beams have been used to mimic neutron damage [8–11]. However, using ions as surrogates for neutrons do not fully replicate the uniform damage profiles that neutrons can provide, but within the penetration depth of these heavy ions, they model the retention characteristics well. The other issue with using heavy ions to create damage is impurity build-up in the material. Some experiments have used self-damage, i.e. W ions on W, to avoid contamination [8,11], but W ions have a very shallow stopping distance in W (about an average of 500 nm with a 6 MeV beam [12]) as well as creating a W ion beam has complications of its own. Cu ions have been used to create damage in W [6,9], and they can produce similar damage profiles as W ions. Therefore we want

* Corresponding author. Tel.: +1 8588223672.
E-mail address: jbarton@ucsd.edu (J.L. Barton).

http://dx.doi.org/10.1016/j.nimb.2014.02.077
0168-583X/Published by Elsevier B.V.

to investigate when contamination from impurity Cu ions start to play a role in changing retention characteristics in W.

This paper examines H isotope exchange in W by first treating W samples with D plasma to high fluence and subsequently exposing them to H plasma to measure the remaining D retention. In order to simulate realistic material properties after neutron irradiation in a fusion reactor, some W samples were irradiated with heavy ions to varying levels of damage before plasma treatment, where the use of Cu ions versus W ions was compared.

2. Experimental

2.1. Materials

Plansee [13] polycrystalline W samples 6 mm in diameter and 2 mm thick – machined from W rods – were used in these experiments. The surfaces were mechanically polished to a mirror finish, and were cleaned in an acetone ultrasonic bath. To decrease the amount of intrinsic defects and remove impurities, the samples were then annealed at 1273 K for 1 h.

2.2. Damage production in W with Cu and W ions

The tandem ion accelerator at the Ion Beam Materials Laboratory (IBML) in Los Alamos National Laboratory (LANL) was used to produce 2 MeV Cu^+ and a 6 MeV W^{2+} ion beams to create displacement damage in our W samples. Damage profiles were simulated with the SRIM-2012 simulation code [12], with the "detailed calculation with full damage cascades" option turned on and a displacement damage threshold of 90 eV for W as recommended in [14]. These damage profiles and ion stopping ranges are shown in Fig. 1. The left-hand vertical axis shows the distribution of displacements per atom (dpa) from Cu and W ions to produce 1.0 dpa at its peak, and the right-hand vertical axis shows the stopping range of these ions. Peak dpa were calculated as

$$\mathrm{dpa}_{\mathrm{peak}} = \frac{N_{\mathrm{displacement}}\,\phi}{N_{\mathrm{W}}}, \tag{1}$$

where $N_{\mathrm{displacement}}\left(\frac{1}{\mathrm{ions\cdot m}}\right)$ is the number of displacements per ion per unit length given by SRIM output, $\phi\left(\frac{\mathrm{ions}}{\mathrm{m^2}}\right)$ is the ion fluence, and $N_{\mathrm{W}}\left(\frac{\mathrm{at}}{\mathrm{m^3}}\right)$ is the atomic density of W. Since Cu ions have significantly lower mass than W ions and therefore have a lower number of displacements per incoming ion, Cu ion fluences had to be increased to 3.5 times the W ion fluence to represent a similar damage profile in order to compare retention characteristics. The ion stopping ranges are quite similar, but it is important to note that Cu contamination occurs only in the first 1 μm. In order to produce 0.01, 0.1, and 1.0 dpa in our W samples, we applied Cu ion fluences of 4.2×10^{16}, 4.2×10^{17}, and 4.2×10^{18} ions/m^2, respectively, where the W ion doses were 3.5 times lower.

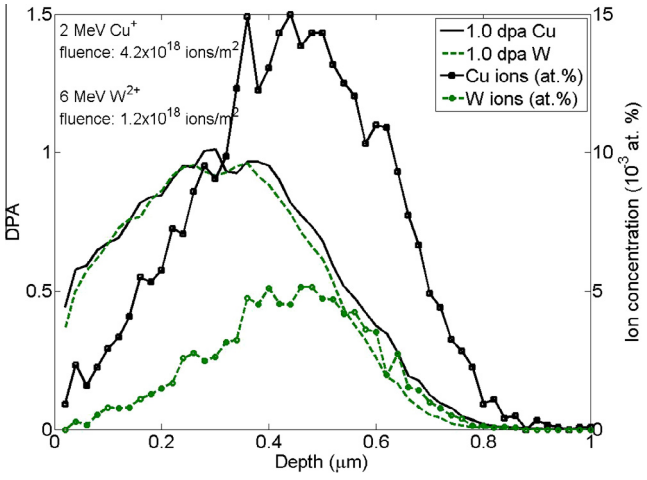

Fig. 1. SRIM simulated damage profiles and ion concentration in W (90 eV displacement threshold) for 2 MeV Cu and 6 MeV W ions.

2.3. Plasma treatment

Undamaged as well as Cu irradiated and W irradiated samples were exposed to plasma in the PISCES linear plasma device at UCSD. Fluxes in the PISCES-A device for both D and H plasmas were $1.5(\pm 0.3) \times 10^{22}$ ions/m^2/s. We expect radial uniformity in the plasma wetted area of our 6 mm diameter samples because our beam diameter is greater than 4 cm. The ion energies were 150 eV per D_2^+ or H_2^+, calculated by subtracting the bias voltage of the sample holder (−160 V) from the plasma potential measured from a swept Langmuir probe. In our experiments, neutral gas pressures entering the vacuum chamber below 0.67 Pa produce molecular ions as the dominant plasma species [15]. We kept the W sample temperature at a constant 473 K, measured by a thermal couple pressed against the back side, isolated from the plasma. All samples were treated with D plasma to 10^{26} ions/m^2, which is a fluence well into the saturation regime [16]. Undamaged samples used for isotope exchange were subsequently exposed to H plasma fluences of 2.6×10^{24}, 5×10^{25}, or 10^{26} ions/m^2; and isotope exchange experiments with Cu irradiated samples were also exposed to H plasma to fluences of 5×10^{25} or 10^{26} ions/m^2.

2.4. Nuclear reaction analysis and thermal desorption spectroscopy

Profiles of D concentration as a function of depth were obtained by nuclear reaction analysis (NRA) up to 7.7 μm by the D(^{3}He,p)^{4}He nuclear reaction and measurement methods described in [17]. The measured energy of protons that escape through the surface determines the depth into W from which the reaction took place [18]. A 2 mm thick Si surface barrier detector, with a 300 mm^2 aperture and 45 mm from the sample at a 135 degree angle relative to the beam, was covered by 24 μm thick Al foil to stop elastically scattered ^{3}He particles as well as a 3 mm curved slit aperture, as in [17], to isolate the proton energy measurement. To increase the precision of our measurements, we used ^{3}He beam energies of 0.8, 2, 2.75, 3.5, and 4.5 MeV, where greater energies provide confidence in resolution at greater depths as the penetration distance of ^{3}He increases before it reacts with D. The analysis software SIMNRA [19] was employed to fit the yield versus energy spectra to extract concentration versus depth profiles. The simulation inputs assume constant concentration of D per W layer, where each layer thickness can be varied.

The total amount of D retained in W was measured with thermal desorption spectroscopy (TDS). Samples were heated from room temperature to 1273 K at a rate of 0.5 K/s with infrared heating lamps and a background pressure of 1.1×10^{-5} Pa. A quadrupole mass spectrometer (QMS) measured the D_2 and HD pressure signals as the temperature increased and released these molecules from the sample. After each analysis, the QMS was calibrated with a standard D_2 leak to convert our pressure data to molecular flux. Integrating the signals over the temperature ramp gives the fluence, which is total amount retained.

3. Results and discussion

Exposures to 150 eV/H_2^+ plasma at various fluences and a 473 K sample temperature decreased the amount of D atoms saturated in the W samples by exchanging with the implanted D. The

Please cite this article in press as: J.L. Barton et al., Deuterium retention in tungsten after heavy ion damage and hydrogen isotope exchange in PISCES, Nucl. Instr. Meth. B (2014), http://dx.doi.org/10.1016/j.nimb.2014.02.077