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Effects of background gas impurities during D^+ irradiation on D trapping in single crystal tungsten

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Abstract

The level of impurities in the background gas during 500 eV D^+ irradiation was found to influence D trapping in single crystal tungsten. Reduced D retention levels were observed for lower partial pressures of impurity gases during irradiation. D^+ irradiations created near-surface peaks in the C and O depth distributions; the peak magnitudes depended on the background gas impurity levels. It is proposed that the recoil implantation of carbon and oxygen impurities arriving at the surface created vacancies in tungsten, thereby increasing D trapping.

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1. Introduction

In previous studies, it was found that the vacuum conditions during D^+ irradiation could influence D retention in tungsten [1]. The objective of the present study was to quantify the background impurity gas levels during D^+ irradiation and determine a link between the background gas levels and D trapping. Single crystal tungsten (SCW) was used to eliminate some of the variability due to grain boundaries, dislocations and other defects in polycrystalline tungsten materials [2–6].

2. Experiment

Four specimens (5×5 mm^2 and 0.5 mm thick) were cut from a 25 mm diameter single crystal tungsten disc (99.98 wt%), produced by the Rare Metals Institute, Moscow. The surface normals were within 15° of the [0 0 1] direction, as measured by orientation imaging microscopy. The specimens were mechanically polished, electropolished, and annealed for 20 min at 1775 K prior to each D^+ irradiation.

Specimens were irradiated with 1.5 keV D_3^+ (500 eV/D^+) with a flux density of 6×10^{19} D^+/m^2 s. A W foil of 25 μm thickness with a 2 mm aperture, clamped directly in front of the specimen, defined the irradiated area. A ceramic heater (15 mm dia) was used to heat the specimens, and the temperature during irradiation was measured with a chromel–alumel thermocouple. Irradiations were performed under two different vacuum conditions: (i) without baking the vacuum system – referred to as *regular* irradiations; and (ii) under vacuum

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conditions achieved by an 18 h bakeout at 373 K and the use of a liquid nitrogen cooled collimation aperture – referred to as *clean* irradiations. A Hiden quadrupole mass spectrometer (QMS) was used in conjunction with a calibrated D_2 leak bottle to quantify the constituents of the background gas during irradiation. Irradiations were performed at 300 K and 500 K for 3×10^{23} D^+/m^2 and 10^{24} D^+/m^2 fluences, with a different specimen used for each irradiation temperature/fluence combination. However, for each temperature/fluence combination, the same specimen was used for both *regular* and *clean* irradiations.

The amount of D retained was measured by thermal desorption spectroscopy (TDS) in a separate vacuum system, requiring transport of the specimen through air. The TDS system was baked at 410 K for 2 h to reduce the background signal. The typical background pressure was 2×10^{-6} Pa. The SCW specimen was placed on a W foil heating cradle and heated linearly at ~5 K/s up to 1775 K. The specimen temperature was measured using a W–5%Re/W–26%Re thermocouple. The desorbed D_2 was measured using the Hiden QMS, which was calibrated with a D_2 leak bottle.

Secondary ion mass spectroscopy (SIMS) depth profiling was performed after polishing/annealing, irradiation, and TDS. The analysis beam was 25 keV Ga^+, with 2.4 pA over (50 μm)^2 area. The sputter beam (depth profiling) was 1 keV Cs^+, with 8.4 nA over (200 μm)^2 area. SIMS was used to obtain the depth distributions of selected impurities just below the surface, at depths of 2–60 nm; it was not intended to measure the adsorbed surface impurities due to atmospheric exposure.

3. Results

The background gas levels measured during irradiations are summarized in Table 1. The main components were D_2 (~85%) and H_2 (~8%; not listed). The *clean* irradiations exhibited significantly lower background levels of CH_4, H_2O, CO, O_2, and CO_2, with the largest reductions observed for H_2O.

The TDS profiles and the differences in D retention (Δ_D) for the *regular* and *clean* cases are given in Fig. 1. For the 3×10^{23} D^+/m^2 irradiations at 500 K, the desorbed D_2 signals were indistinguishable from the background D_2 levels and are not shown. It is evident from

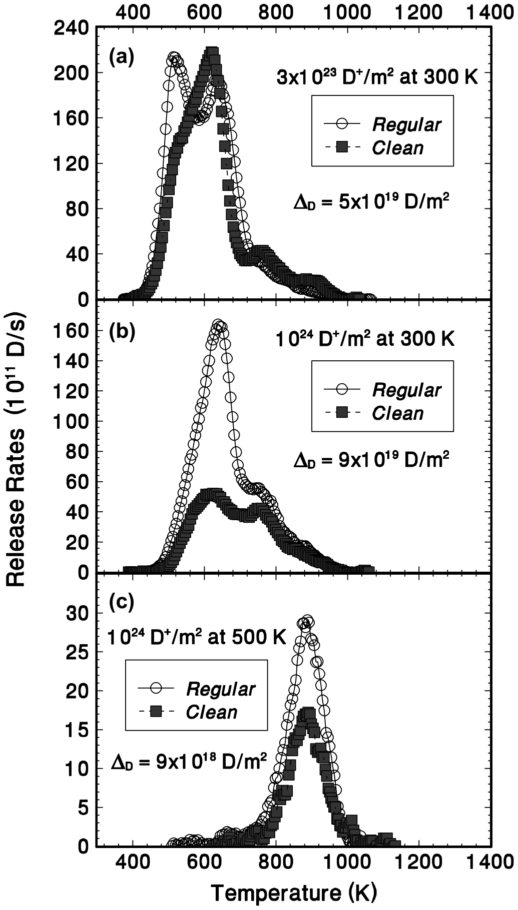

Fig. 1. Effects of *regular* and *clean* irradiations on the TDS profiles. (a) 3×10^{23} D^+/m^2 at 300 K, (b) 10^{24} D^+/m^2 at 300 K, and (c) 10^{24} D^+/m^2 at 500 K. Δ_D Denotes D retention for *regular*, minus D retention for *clean* irradiations.

Table 1
Background gas partial pressures during D^+ irradiation at 300 K and 500 K

($\times 10^{-5}$ Pa)	3×10^{23} D^+/m^2 (300 K)		10^{24} D^+/m^2 (300 K)		3×10^{23} D^+/m^2 (500 K)		10^{24} D^+/m^2 (500 K)	
	Regular	Clean	Regular	Clean	Regular	Clean	Regular	Clean
D_2	4.2	4.2	3.7	3.8	4.8	5.2	4.6	4.3
CH_4	0.014	0.0065	0.01	0.0064	0.012	0.0072	0.011	0.0055
H_2O	0.26	0.01	0.17	0.011	0.32	0.015	0.26	0.0086
CO	0.11	0.068	0.071	0.071	0.1	0.09	0.082	0.066
O_2	0.0018	0.00025	0.0011	0.00041	0.0017	0.00022	0.0017	0.00013
CO_2	0.021	0.0075	0.015	0.0073	0.024	0.0088	0.018	0.0076

the profiles that D retention was lower for the *clean* irradiations. In all cases, the lower background gas levels during irradiation resulted in the decrease of one particular desorption peak, without changing the temperature or widths of the desorption peaks. However, in each case, a different temperature desorption peak was affected by the *clean* irradiation background.

SIMS depth profiles for 'before' and 'after' irradiations are shown in Figs. 2 and 3. SIMS analysis of the surfaces outside of the irradiated area always produced depth profiles similar to the unirradiated cases in (a) and (d). By comparing Fig. 2(b) with (e), and (c) with (f), a reduction is seen in the near-surface O and C peaks as a result of the *clean* background during irradiations at 300 K. A decrease in the D profile is also observed. Similar behaviour is seen for irradiations at 500 K; Fig. 3(b) and (e). Unfortunately, there was difficulty in obtaining the SIMS depth profiles after the clean irradiation at 10^{24} D^+/m^2 at 500 K.

4. Discussion

The reduction of impurities in the background gas during D^+ irradiation has led to changes in the near-surface O and C concentrations, the D desorption profiles, and the retained amount of D. The correlation between the reduction of background gas impurities and the decrease in the near-surface oxygen and carbon peaks (Figs. 2 and 3) suggests that oxygen and carbon from the background gas entered the tungsten to create these near-surface peaks. The thermal energies of the impurity molecules and atoms is insufficient to enter the tungsten, as evidenced by the off-irradiation-spot SIMS analysis that showed no near-surface impurity peaks. However, through elastic collisions, the impacting 500 eV D^+ can transfer up to 200 eV and 250 eV to O and C atoms, respectively [7]. At these energies, O and C can enter the tungsten, but only to depths of a few nanometers [8]. Collisions with successive D^+ particles and

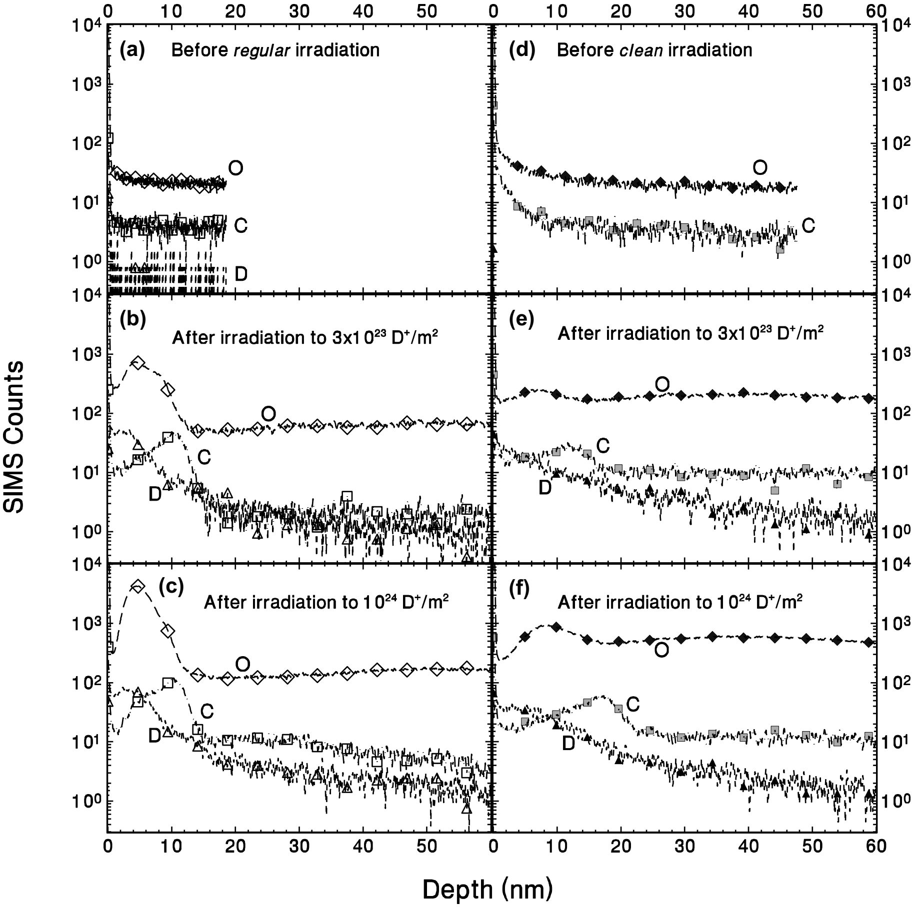

Fig. 2. Ex situ SIMS analysis of SCW: (a) before irradiation, (b, c) after *regular*, and (e, f) after *clean* irradiations at 300 K (only 1 of every 100 data points are plotted as symbols).

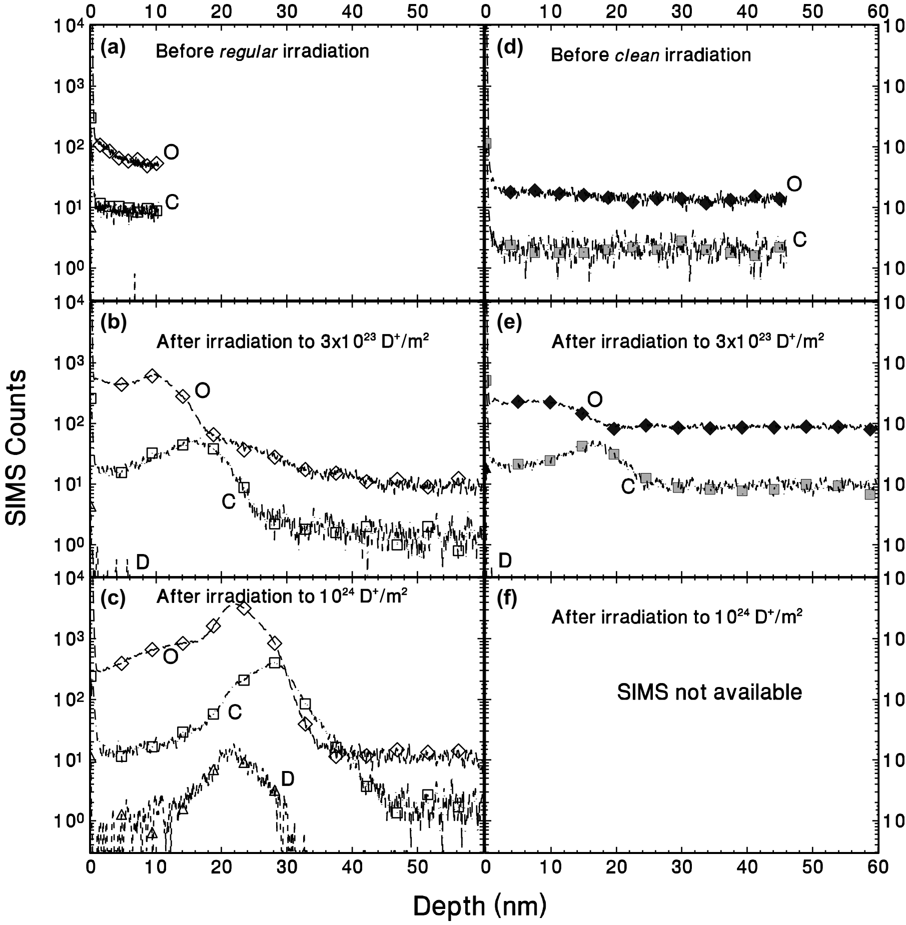

Fig. 3. Ex situ SIMS analysis of SCW: (a) before irradiation, (b,c) after *regular*, and (e) after *clean* irradiations at 500 K (only 1 of every 100 data points are plotted as symbols).

radiation-enhanced diffusion may allow the impurity atoms to reach depths equivalent to the 500 eV D$^+$ ion range (~20 nm [8]).

In these experiments, a thin adsorbed impurity layer was always present on the specimens at the start of the irradiation because of atmospheric exposure. These surface impurities (typically a few monolayers thick) would serve as the initial source of carbon and oxygen atoms for implantation by recoil collisions. As the D$^+$ irradiation continues, this initial source of surface impurities will be sputtered off, removed by ion-impact-enhanced desorption or recoil-implanted. The arrival of impurities at the surface during D$^+$ irradiation will be governed by the partial pressures of the background impurity gases. (With a flux of 6×10^{19} D$^+$/m^2 s, and a conservative sputter/ion-impact-desorption yield of 10^{-2}, it would take 60 s to remove three monolayers of adsorbed impurities, compared to implant times of 5000–16000 s.) With background gas impurity levels of the order of 10^{-6} Pa (less for *clean* conditions), impurity arrival rates would be approximately one monolayer in 100 s or more, with only a small fraction of these impurities likely to be recoil implanted. For relatively short D$^+$ irradiation times, one might not expect a large effect from the improved background gas conditions (*clean* vs. *regular*) because the impurity levels in the near surface of the specimen would be dominated by recoils from the initial adsorbed layer. However, for relatively long irradiations, the concentration of background gas impurities during D$^+$ irradiations would have a bigger impact. This is consistent with the observed larger differences between the *regular* and *clean* irradiations in the TDS (Δ_{D} in Fig. 1) and SIMS spectra (Figs. 2 and 3) for the higher fluence cases.

The maximum energy transfer from 500 eV D$^+$ to a W atom is 21 eV – insufficient to create a displacement [9]. However, the energy transfer from oxygen or carbon is more efficient, such that vacancies can be created by relatively low energy O or C. TRIM calculations [8] using 200 eV O$^+$ and 250 eV C$^+$ on a pure W target show vacancy production rates of 0.4 vacancies/O$^+$ and 0.3 vacancies/C$^+$. The O and C energies used in this calculation are the calculated maximum energy transfer from 500 eV D^+, assuming two-body collisions [7]. Thus, it is possible for 500 eV D^+ to create vacancies in tungsten, through intermediate recoil collisions with O and C impurities on the surface. Deuterium trapping at vacancy-type defects with different trap energies is known to be a significant D retention mechanism in tungsten [6,10,11]. It then follows that by reducing the concentration of surface impurities during D^+ irradiation, the number of vacancies created will be reduced, and the amount of D trapped will decrease.

5. Conclusions

Impurities present in the background gas during D^+ irradiation affect D trapping in tungsten. Higher levels of background impurities lead to higher D retention. It is suggested that impurities from the gas arrive at the tungsten surface and are implanted by recoil collisions with the incident D^+ ions. In some cases, the energy transferred to the impurity atoms during these collisions is sufficient for the impurity atoms to create vacancy defects, which become a new source of traps for deuterium.

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