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Detection of deuterium trapping sites in tungsten by thermal desorption spectroscopy and positron annihilation spectroscopy

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ABSTRACT

Thermal desorption spectroscopy (TDS) of tungsten implanted with D_2^+ ions was performed after irradiation with 8 MeV-electrons, 5.0 keV-D_2^+, and 6.0 MeV-Fe^{3+}. The release peak temperatures of the TDS spectra are discussed. Positron annihilation lifetime (PAL) measurements of electron-irradiated tungsten were also performed, showing that single vacancies migrate a sufficient distance to arrive at a sink or meet interstitial-type defects during annealing at 673 K. A decrease in the PAL was detected for single vacancies that contain deuterium atoms. The peak temperature of deuterium release from dislocations was lower than that from single vacancies. In samples irradiated with 6.0 MeV-Fe^{3+}, the effect of Fe contamination on deuterium trapping and the deuterium release from voids were detected. These tendencies correspond to previous works.

1. Introduction

In a fusion reactor, plasma-facing materials (PFMs) must withstand the damage produced by plasma particles at energies ranging from several electron volt to several kiloelectron volt [1], heat loads from the plasma, and neutrons with high energy, high flux, and high fluence. Therefore, important criteria for the selection of PFMs are high melting point, high thermal conductivity, and low sputtering erosion. High-Z materials such as tungsten, and molybdenum, together with low-Z materials such as carbon, carbon fibre composites and beryllium have been selected as PFMs due to outstanding thermal properties and erosion resistance. The plasma-induced erosion in high-Z materials, especially tungsten, is considerably lower than that in low-Z materials. However, high-Z materials have a far lower tolerance to impurity concentrations within the plasma [2], as the radiative power loss of the plasma is proportional to the square of the charge number of impurity ions. Another disadvantage is bubble formation, where hydrogen isotopes are retained in vacancy-type defects in PFMs [3,4], which is a critical problem for fusion reactors. Thus, it is very important to determine the interactions between vacancy-type defects and hydrogen isotopes.

Positron annihilation spectroscopy (PAS) is a very powerful tool for detecting vacancy type defects. Many research groups adopted PAS to investigate the irradiation effects in tungsten [5–10]; however, they mainly performed PAS after irradiation. Van Veen et al. and de Vries et al. reported the change in the positron annihilation lifetime (PAL) of hydrogen decorated vacancies and voids by annealing in tungsten [11,12]. In this study, the size of vacancy-type defects in tungsten was evaluated from PAL measurements, which were performed before and after deuterium charging to identify the type of pre-existing defects (deuterium trapping sites) and obtain the change in the PAL of vacancies by deuterium trapping. Thermal desorption spectra of deuterium were also obtained after deuterium charging. The trapping sites and peak temperatures of deuterium release from tungsten were identified from these results.

2. Experimental procedure

High purity tungsten (99.95%, A.L.M.T. Corp.) samples were employed in this study. Samples with diameters of 5 mm were punched from 0.2 mm thick tungsten sheets and annealed at 1773 K for 1 h in a vacuum ($<10^{-4}$ Pa) to facilitate recrystallization. All samples were electropolished before and after annealing to remove any oxidation. As mentioned in the next paragraph, electron irradiation was performed with water cooling. An oxidation layer was formed on the surface during electron irradiation and was re-

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Table 1
Sample list and experimental conditions.

Sample	Annealing condition	Irradiation condition	D_2^+ ion implantation
As-received	–	–	1 keV 1.0×10^{22} /m^2
Well-annealed	1773 K for 1 h	–	1.0 – 1.5 keV 5.0×10^{21} /m^2
Dislocation	1273 K for 5 h	–	1 keV 1.0×10^{22} /m^2
Electron-irr.	1773 K for 1 h	8 MeV electrons 9.4×10^{21} /m^2 (333 K) 5 keV D_2^+ ions	1 keV 1.0×10^{22} /m^2
D_2^+-irr. (R.T.)	1773 K for 1 h	1.0×10^{22} /m^2 (room temperature.)	–
D_2^+-irr. (H.T.)	1773 K for 1 h	5 keV D_2^+ ions 1.0×10^{22} /m^2 (673 K)	1 keV 1.0×10^{22} /m^2
Fe^{3+}-irr.	1773 K for 1 h	6 MeV Fe^{3+} ions 2.3×10^{23} /m^2 (573 K)	1 keV 1.0×10^{22} /m^2

moved by electropolishing. All samples were electropolished after annealing so thermal desorption spectroscopy (TDS) could be performed under the same surface conditions. Defects were introduced by irradiation with electrons, D_2^+ ions and Fe^{3+} ions.

The electron irradiation was performed at 8 MeV at doses to 2.1×10^{21} and 9.4×10^{21} /m^2 (7.1×10^{-6} and 3.2×10^{-5} dpa) at 333 K using the Electron Linear Accelerator of the Research Reactor Institute, Kyoto University. Water cooling was adopted to remove beam heating, and the sample temperature was measured with a thermocouple. For dose calculations, we applied linear interpolation to the values listed in reference [13] to obtain the atomic displacement cross section of 70.4 barns. Ogorodnikova et al. reported the cross section of 34 barns in the case of 3.5 MeV electron irradiation [14]. Their value is almost equal to the atomic displacement cross section with the displacement threshold energy of 84 eV for 3.5 MeV electron irradiation [13]. Therefore, the displacement threshold energy of 84 eV was used for the dose calculation of electron irradiation. Deuterium ion irradiation was performed with 5 keV D_2^+ ions at up to 1.0×10^{22} /m^2, providing 2.8 dpa at the defect peak, 5 nm from the surface, at room temperature and 673 K using the low-energy ion irradiation system of the Research Reactor Institute, Kyoto University. Heavy ion irradiation was performed with 6 MeV Fe^{3+} ions at up to 2.3×10^{19} /m^2, providing 1.9 dpa at the defect peak, 11 μm from the surface, at 573 K at Quantum Science and Engineering Center, Kyoto University. The irradiation doses and defect peaks of the ion irradiation were obtained by analysing the results of an SRIM simulation [15] with the Quick Kinchin-Pease option [16].

Samples containing only dislocations were prepared by annealing the as-received samples at 1273 K for 5 h. Table 1 lists the details for each sample and the experimental conditions employed. The untreated sample and the samples annealed at 1723 K for 1 h and at 1273 K for 5 h are referred to as the "as-received", "well-annealed", and "dislocation" tungsten samples, respectively. The irradiated samples are referred to as the "Electron-irr.", "D_2^+-irr. (R.T.)", "D_2^+-irr. (H.T.)" and "Fe^{3+}-irr." in accordance with the irradiation conditions shown in Table 1.

D_2^+ ions were implanted (1 keV, 1×10^{22} /m^2) at room temperature to the as-received, dislocation, well-annealed, electron-irr., D_2^+-irr. (H.T.), and Fe^{3+}-irr. samples. Since we could not obtain any peaks in the TDS spectrum of the D_2^+-irr. (H.T.) sample after 5 keV D_2^+ irradiation, subsequent 1 keV D_2^+ irradiation was needed. On the other hand, because the D_2^+-irr. (R.T.) sample sufficiently retained D atoms after irradiation, we did not conduct 1 keV D_2^+ irradiation. Thermal desorption spectroscopy (TDS) was conducted from room temperature to 1523 K at a heating rate of 1 K/s to study the behaviour of deuterium release from tungsten defects. The samples were transferred from the D_2^+ ion implantation apparatus to the TDS instrument in a vacuum chamber. However, the samples were exposed to air for approximately 10 min while fixed to the sample holder of the TDS instrument. The PAL measurements were performed at room temperature using a fast-fast coincidence system with a time resolution of 190 ps (full width at half maximum; FWHM) [17]. We used Na-22 as the positron source. The energy spectrum of positrons emitted from Na-22 is broad up to an energy of approximately 540 keV [18]. The maximum penetration depth in tungsten was about 100 μm using the so-called Makhov distribution [19]. The PAL spectra were collected with a total count of $1.0 - 1.5 \times 10^6$ and were analysed using the PALSfit program [20]. The positron annihilation lifetime spectra of well-annealed and electron-irr. W are shown in Fig. 1. The errors given in Table 2 denote the standard deviations obtained in the decomposition of the PAL spectra, which are not dependent on the time resolution of the experimental system. The total amount of residual defects can be derived from the mean positron lifetime (τ_m). The amount of positron annihilation that occurs in the matrix can be determined from the short lifetime component (τ_1), the size of vacancy-type defects can be derived from the long lifetime component (τ_2), and the density can be obtained from the relative intensity of the long lifetime component (I_2). When vacancy clusters grow, the positron lifetime increases. Table 2 gives the positron annihilation lifetimes for the as-received, dislocation, well-annealed, and electron-irr. tungsten. The long lifetime component, τ_2, of 149 ps for the dislocation sample was shorter than that calculated for single vacancies, 161 – 200 ps [21, 22], indicating that the sample contains no vacancy clusters or single vacancies. The short lifetimes of 19 ps (dislocation) and 85 ps (electron-irr.) denote the lifetime of positron disappearance not only at the matrix but also from it (migration to the defect site). In the two-state trapping model [23–25], the short lifetime τ_1 is expressed as $\tau_1 = 1 / (\lambda_f + \kappa)$ (κ: the trapping rate to defects). As $\tau_f = 1 / \lambda_f$

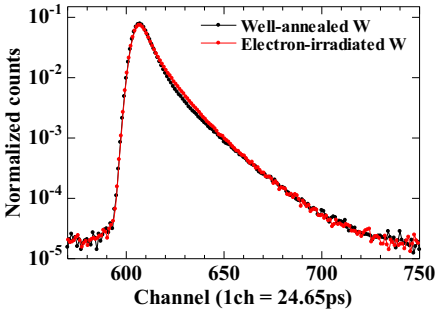

Fig. 1. Positron annihilation lifetime spectra of well-annealed and electron-irr. tungsten. The dose of electron irradiation was 3.2×10^{-5} dpa, and the spectrum after electron irradiation changed at around 630 ch.

Table 2
Positron annihilation lifetime for as-received, dislocation, well-annealed and electron-irr. tungsten. τ_m, τ_1, τ_2, and I_2 denote mean, short, and long lifetimes, and long lifetime intensity, respectively.

Sample	τ_m (ps)	τ_1 (ps)	τ_2 (ps)	I_2 (%)
As-received	175 ± 1	163 ± 1	400 ± 1	6 ± 1
Dislocation	148 ± 1	19 ± 8	149 ± 1	94 ± 1
Well-annealed	109 ± 1	–	–	–
Electron-irr.	127 ± 1	85 ± 2	173 ± 4	42 ± 3

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