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MODELING OF HYDROGEN ISOTOPE TRAPPING IN SELF-DAMAGED TUNGSTEN

As a material for plasma-facing components in future fusion reactors, tungsten (W) will be subjected to intensive fluxes of energetic deuterium (D) and tritium (T) as well as 14 MeV neutrons (n). One of the ways to investigate the influence of n-induced defects on the hydrogen isotope inventory is to simulate displacement damage in tungsten using irradiation with energetic ions.

In the last decade, a number of studies have been carried out [1-5] in which the concentration of deuterium [1-4] and tritium [5] in the damage zone of tungsten samples, previously damaged by 20 MeV W ions to damage levels of 0.23 and 0.41 displacements per atom (dpa), was determined.

At relatively low temperatures of hydrogen loading, traps are fully occupied by H. However, if the temperature is sufficiently high to attain trapping-detrapping equilibrium, the probability of trap occupancy by hydrogen sensitively depends on the concentration of H atoms in a solid solution state. Note that speaking of hydrogen (H) we also mean hydrogen isotopes such as deuterium and tritium. The fraction of traps, θ_t, occupied with hydrogen at temperature T is expressed as

$$\theta_t/(1 - \theta_t) = \theta_L \exp(E_{bin}/k_{Bol}T), \quad (1)$$

where θ_L is the fraction of interstitial sites occupied with hydrogen (solid solution state), E_{bin} is the binding energy (the enthalpy difference between H atom in a solid solution state and in a trapped state), and k_{Bol} is the Boltzmann constant [6]. This means θ_t varies with the H concentration in the solid solution state, C_{SS}, being proportional to θ_L. Note that

$$C_{SS} = C_L \theta_L, \quad (2)$$

where C_L is the concentration of interstitial sites.

Under exposure to H_2 gas, C_{SS} is determined by Sieverts' law:

$$C_{SS} = S_0 \exp(-E_S/k_{Bol}T) P^{1/2}, \quad (3)$$

where S_0 is the solubility constant, E_S is the heat of solution, and P is the H_2 pressure.

Under exposure to H plasma, C_{SS} is determined by the balance between penetrating flux Γ_{in} and recycling flux Γ_{rec} of hydrogen. In the first approximation, $\Gamma_{in} = \Gamma_{rec} = k_r C_{SS\text{-}S}^2$ in a steady state where $C_{SS\text{-}S}$ is the concentration of solute H beneath the surface and k_r is the recombination rate coefficient. Hence,

$$C_{SS\text{-}S} = (\Gamma_{in}/k_r)^{0.5}. \quad (4)$$

Normalizing the concentration of hydrogen and the concentration of traps with the atomic density of tungsten, equation (1) can be modified as

$$\theta_t = \frac{C_{SS}\exp(\frac{E_{bin}}{k_{Bol}T})}{1+C_{SS}\exp(\frac{E_{bin}}{k_{Bol}T})} . \quad (5)$$

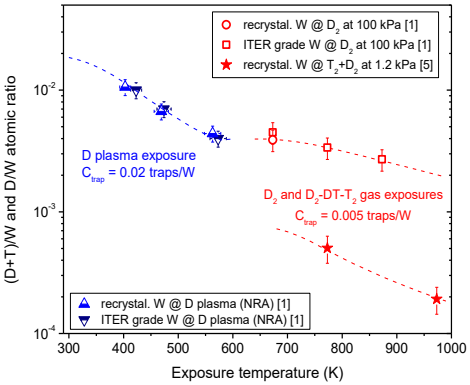

Figure 1. The hydrogen isotope concentration in the damage zone of self-damaged W samples exposed (i) to D_2 gas at a pressure of 100 kPa [1], (ii) to $D_2 - DT - T_2$ gas mixture with a tritium content of 6% at a total gas pressure of 1.2 kPa [5], and (iii) to low-energy (≤ 380 eV), low-flux D plasma [1], as a function of the gas/plasma exposure temperature.

Under the assumption that one trap captures one hydrogen atom, the concentration of trapped hydrogen, C_H (in units of the H/W atomic ratio) can be expressed as $C_H = \theta_t C_{trap}$, where C_{trap} is the concentration of traps.

Measured values of the D concentration in the damaged W samples exposed to D_2 gas at 100 kPa and temperatures ≥ 673 K are well described by equation (5) using the binding energy of deuterium in the ion-induced traps E_{bin} = 1.40 eV and the concentration of traps C_{trap} = 0.005 traps/W (Fig. 1).

In case of the low-flux D plasma exposure [1], an implantation flux of D particles (the penetrating flux Γ_{in}) was estimated to be 2×10^{18} D/m^{2}s [5].

According to Pick and Sonnenberg [7], the recombination rate coefficient $k_r = \chi\,\mu/S^2$, where the solubility $S = S_0 \exp(-E_S/k_{Bol}T)$, $\mu = 1/\sqrt{2\pi m k_{Bol} T}$ (m is the mass of hydrogen isotope molecule, for deuterium $m = 6.689\times10^{-27}$ kg, and $k_{Bol} = 1.3806\times10^{-23}$ J K^{-1}), and the sticking probability $\chi = \chi_0 \exp(-2E_{dis}/k_{Bol}T)$ (E_{dis} is the activation energy for hydrogen dissociation).

To fit the calculated values to the experimental data obtained after irradiation with low-energy, low-flux D plasma [1] (Fig. 1), the following parameters were used: in the temperature range from 300 to 600 K the binding energy E_{bin} varied from 1.278 to 1.303 eV (Fig. 2), the concentration of traps C_{trap} = 0.02 traps/W, the pre-exponential sticking coefficient $\chi_0 = 5\times10^{-9}$, and the activation energy for hydrogen dissociation E_{dis} = 2.5 eV.

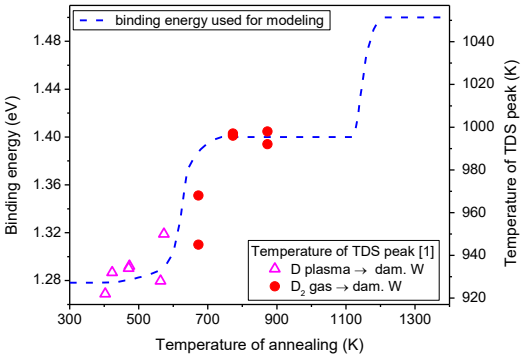

Figure 2. Dependence of the binding energy of deuterium atoms with trapping sites, E_{bin}, for the self-damaged W samplesm as a function of annealing temperature. A sharp increase in the binding energy in the temperature range from 1123 to 1200 K is explained by the formation of cavities at stage V of defect recovery and an increase in the binding energy for D atom trapped at the inner wall of a void.

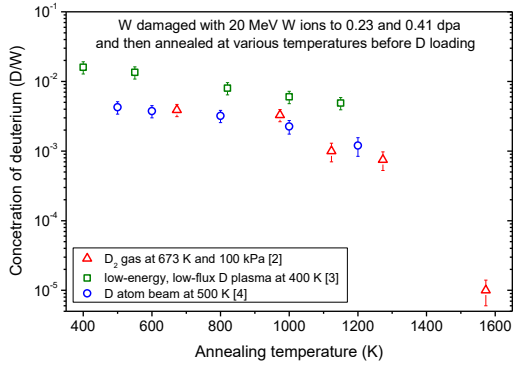

Figure 3. Concentration of deuterium (in units of the D/W atomic ratio) trapped in the damage zone of self-damaged W annealed in vacuum at various temperatures for 1 h [3, 4] and 6 h [2] followed by exposure (i) to D_2 gas at a pressure of 100 kPa and a temperature of 673 K for 10 h [2], (ii) to low-energy (15 eV) and low-flux (5×10^{19} D/m^{2}s) D plasma at 400 K to a fluence of 1×10^{25} D/m^2 [3], and (iii) to low energy (0.2 eV) neutral D atoms at flux of 2.6×10^{19} D/m^{2}s for 144 h at 500 K [4].

After exposure of the W-ion-damaged W samples to both the low-energy, low-flux D plasma at the sample temperatures ranging from 403 to 573 K and D_2 gas at 673 and 773 K, single thermal desorption peaks were observed in a rather narrow temperature range from 922 to 998 K (Fig. 2). This fact with some caution suggests that the binding energy of deuterium in the ion-induced traps, which were subjected to temperature effects when exposed to low-energy, low-flux D plasma and D_2 gas, increases from 1.278 to 1.40 eV with increasing the sample temperature from 300 to 973 K. A sharp increase in the binding energy in the temperature range from 1123 to 1200 K is explained by the formation of voids due to the thermal coarsening of vacancy clusters.

In the case of annealing of the displacement-damaged W samples with subsequent exposure to D_2 at 673 K and 100 kPa [2], the concentration of traps, C_{trap}, was determined using equation (5), the solubility parameters, and the binding energies E_{bin} shown in Fig. 3. The concentration of traps in annealed self-damaged W samples, estimated by D_2 gas exposure, is shown in Fig. 4, as a function of annealing temperature.

Using the concentration of traps C_{trap} = 0.005 of traps/W at the annealing temperature of 800 K as the reference one and using equation (5), we determined the concentration of deuterium in the solid state, C_{SS}, for the case of exposure to D atoms at 500 K [4], at which the concentration of traps in the case of D atom exposure would be equal to the reference value. After that, we use this concentration C_{SS} to calculate the concentration of traps for other annealing temperatures selected in [4] (Fig. 4). Thus, it was estimated that in the case of exposure of the displacement-damaged W samples to low energy (0.2 eV) neutral D atoms at flux of 2.6×10^{19} D/m^{2}s at 500 K the concentration of D atoms in the solid solution state $C_{SS} = 3.7\times10^{18}$ D/m^3.

A similar processing of experimental data was carried out for the case of exposure of displacement-damaged and annealed W samples to the D plasma at 400 K with a flux of 5×10^{19} D/m^{2}s [3]. In this case, the concentration of deuterium in the solid solution state was chosen equal to $C_{SS} = 1\times10^{21}$ D/m^3.

The data on the concentration of traps obtained from the above-mentioned processing of experimental data [2-4] are presented in Fig. 4.

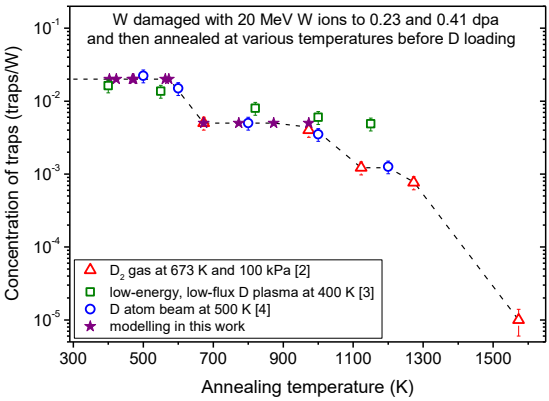

Figure 4. Concentration of traps in self-damaged W irradiated at room temperature with 20 MeV W ions to 0.23 dpa [2, 4] and 0.41 dpa [3] at the damage peak and then annealed in vacuum at various temperatures for 1 h [3, 4] and 6 h [2], as calculated using equation (5) and the D concentration data shown in Fig. 3. The concentration of traps used in modeling the accumulation of deuterium in this work is also shown in the Figure. The concentration of traps was determined under the assumption that one trap can capture one atom of the hydrogen isotope.

References

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