Calculation of deuterium retention in, re-emission and reflection from a tungsten material under D^+ ions irradiation with ACAT-DIFFUSE*

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Abstract

We calculated, with a dynamic Monte Carlo code ACAT-DIFFUSE, fluxes of thermal D_2 re-emission, reflection and self-sputtering from a wrought tungsten material during a time sequence of l00 eV D^+ implantation, post-implanted isothermal out-gassing and thermal desorption spectroscopy. The obtained result agreed well with an existing experiment, where diffusion was considered in the calculations from the beginning of implantation. The three fluxes in the implantation period were shown to be almost comparable. The integrated deuterium flux released in the same period was estimated. The depth profiles of deuterium retained at 300 K in that period indicate that, while their maximum locations did not move, the profiles were broadened out because of fast diffusion. The amount of deuterium retained at 300 K was one order of magnitude higher than that at 473 K.

Keywords: retention, re-emission, deuterium, diffusion, modeling, Monte Carlo, tungsten

*A manuscript of a paper presented at 18th International Conference on Plasma Surface Interactions in Controlled Fusion Devices during May 26-30, 2008 in Toledo in Spain.

Introduction

A tungsten material has been selected as a candidate material of the ITER divertor plate because of its high melting temperature, good thermal conductivity and high resistive nature of erosion due to high threshold energy for sputtering [1].

The ion temperature of the boundary plasma in the ITER is estimated to be around 30 eV [2]. Many experiments [3-9] have so far studied retention, re-emission and thermal desorption spectra using ion beams with energy of hundreds of eV to a few keV, except for a few experiments which employed such low-energy ion beams [10-13].

Above issues obtained experimentally for such low-energy ions have been simulated by computer codes [10, 11, 13]. In the work in [13, 14], a source term for the mobile atom concentration, which is given by the range distribution of implanted ions, was prescribed with a reference to the results obtained with the TRIM.SP [15]. However, as will be indicated later, the concentration profiles of implanted ions were broadened out because of diffusion within the irradiation time concerned here.

In this paper, we have calculated, with a dynamic Monte Carlo code ACAT-DIFFUSE [16], fluxes of thermal D_2 re-emission, reflection and self-sputtering from a wrought tungsten material during a time sequence of l00 eV D^+ implantation, post-implanted isothermal out-gassing and thermal desorption spectroscopy (TDS), and compared them with above experiment [13]. We have also derived the retention and the depth distributions of deuterium retained in the material at temperatures of 300 K and 473 K. In these calculations, the radiation-enhanced diffusion of deuterium and the damage effect of trap energies were taken into account in the damage range.

2. Simulation method

The ACAT-DIFFUSE consists of ACAT [17] part and DIFFUSE part which is based on DIFFUSE [18] code. Since the code has been introduced in detail elsewhere [16], its main features necessary to help understand the following discussions are outlined here.

A total fluence Φ is divided into smaller fluence $\Delta\Phi$ during which incident ions do not change the target composition appreciably, and ions corresponding to $\Delta\Phi$ are exposed to the target together and slowed down instantaneously. First, their slowing down and the associated vacancy and range distributions are calculated through the ACAT routine for a number of incident ions to a binary collision approximation by a Monte Carlo method. Then, thermal processes such as diffusion and recombination of implanted ions are treated during the time interval of $\Delta\Phi/J$ (J being the ion flux), by solving a diffusion equation numerically through the DIFFUSE routine. The two routines were repeated iteratively n times, where $n=\Phi/\Delta\Phi$.

Radiation-enhanced diffusion and a damage effect on the trap energy were so considered as to be proportional to the accumulated radiation damage. Since temperature concerned here was relatively low, we assumed that deuterium atoms instantly recombine with each other and

leave the surface as D_2 when reaching the surface. We used a recombination rate coefficient $K_r = K_r^0 / \sqrt{T} \exp(-E_r / kT)$ [13] with K_r^0 having a value 1.2×10^{-25} cm$^4 \cdot \sqrt{\text{K}}$/s, E_r activation energy for the surface recombination taken to be equal to -0.59 eV [19], k Boltzmann constant, and T temperature. From above assumption, released flux J_r at the surface is given by

$$J_r = K_r c_s^2 , \tag{1}$$

where c_s is concentration at the surface. The diffusion coefficient for deuterium atoms was employed as $D(T) = 1.0 \times 10^{-8} \exp(-0.39 \text{ eV}/kT)$ cm^2/s, where the activation energy was taken from the work in [20]. The pre-exponential factor was chosen to fit the experiment. Eq. (1) offers the boundary condition at the surface for the diffusion equation.

3. **Numerical results and discussions**

We calculated fluxes of thermal re-emission of D_2, reflection and self-sputtering of D from the wrought tungsten in the period of implantation with 100 eV D^+ flux 2.0x10^{15} D^+/cm^{2}s, and flux of thermal re-emission in the out-gassing and the degassing (TDS) periods, as shown in Fig.1, together with the experimental data [13].

Erosion of the material does not occur because the incident energy is lower than the threshold energy for sputtering.

The spectrum of the experimental data had two peaks at 475 K and 850 K in the TDS period. We assumed two trap sites (traps #1 and traps #2) with de-trap energies of 0.85 eV and 2.2 eV and the density fractions of 0.05 D/W and 0.01 D/W, respectively. It is clear that the calculated result fits with the experimental data on the whole. The peak temperatures in the TDS period obtained were 450 K and 860 K, and close to those of the experiment cited above. The result suggests that these peaks observed by the experiment can be ascribed to the existence of the two different trap sites in the material. Since vacancy is hardly produced by 100 eV D^+ ions in tungsten, these trap sites may have previously been created before the experiment.

The fluxes of re-emission, reflection and self-sputtering show almost constant in the implantation period except for approximately early 100 s. The self-sputtering indicates a large amount of deuterium had already been accumulated near the surface. The ratios of these fluxes were approximately 0.45: 1: 0.15 relative to the reflection flux. So, it is noteworthy to point out that self-sputtering of hydrogen isotopes should also be considered in assessing recycling of the first wall and the divertor plate.

Figure 2 indicates the calculated column density of mobile and trapped deuterium atoms versus time. The column density of mobile atoms is an order of magnitude smaller than that of trapped atoms during the irradiation period. The column density of atoms held in traps #1

reaches a certain level at about 50 s after the irradiation started, and gradually decreases with time in the out-gassing period. This reduction contributes to increase the amount of mobile atoms, as depicted in the figure. Without that additional supply of mobile atoms, the flux of re-emission in the out-gassing period would have been decreased much faster than is shown in Fig. 1. On the other hand, the amount of atoms held in traps #2 keeps constant during the first two periods and almost the TDS periods. The sharp drop of the amount of atoms trapped in traps #1 appearing at the beginning of the TDS period results in producing the first peak of the amount of mobile atoms. The second peak is due to the decrease in the amount of atoms kept in traps #2. However, a very small portion of the de-trapped atoms falls into traps #1 again, as shown in Fig. 2. These peaks of mobiles atoms, in turn, give rise to produce the two peaks of the TDS spectrum illustrated in Fig. 1.

Shown in Fig.3 are fluxes of thermal re-emission, reflection and self-sputtering in the first two periods calculated for the material at 473 K which is close to the temperature (475 K) corresponding to the first peak of the TDS spectrum as cited above and in the TDS period. The flux of thermal re-emission becomes constant a little faster than that for 300K in the implantation period and drops drastically compared to that for 300 K and stays below 10^{10} D/cm^2/s in the out-gassing period. The flux of self-sputtering is more than ten times smaller than that for 300 K, which corresponds with a fact that the column density of deuterium in tungsten at 300 K shown in Fig. 2 is several times larger than that for 473 K shown later in Fig. 4. Only one peak is seen in the TDS spectrum.

Fig. 4 illustrates calculated amount of mobile and trapped atoms corresponding to the released flux illustrated in Fig. 3. It is clear that almost all atoms are trapped only in traps #2 throughout the whole periods. The peak of the TDS spectrum shown in Fig. 3 can also be understood with the same explanation as above for the second peak of the TDS spectrum depicted in Fig.1.

The amounts of deuterium atoms retained at 300K and 473 K are shown in Fig. 5. The integrated fluxes released at these temperatures in the implanted period were estimated to be $7.49\text{x}10^{17}$ D/cm^2 and $7.78\text{x}10^{17}$ D/cm^2, resulting in 95.5% and 99.2% of the fluence $7.84\text{x}10^{17}$ D/cm^2. The amount of deuterium retention at 300 K is nearly an order of magnitude larger than that for 473 K. Thus, the amount is expected to be reduced largely at temperature nearly equal to or higher than that corresponding to the trap energy of defects. This difference is understood by estimating roughly the column density of mobile and trapped atoms at the temperatures illustrated in Fig.s 2 and 4. Nearly the same ratio of retained amounts in polycrystalline tungsten at almost the same temperatures as cited above was also shown by an experiment and calculations for 200 eV D^+ ion fluence [8].

The depth profiles of deuterium atoms retained at 300 K are shown in Fig. 6 for fluences of $4\text{x}10^{15}$ D/cm^2, $7\text{x}10^{17}$ D/cm^2 and $1.3\text{x}10^{18}$ D/cm^2 corresponding to irradiation periods of 2 s, 350 s, and 650 s, respectively. It is clear that, while the locations of their maxima did not

move with time, the profiles were broadened out because of fast diffusion.

4. Conclusion

With ACAT-DIFFUSE, we have calculated fluxes of thermal D_2 re-emission, reflection and self-sputtering of D from a wrought tungsten material during a time sequence of l00 eV D^+ implantation, post-implanted isothermal out-gassing and thermal desorption spectroscopy. Diffusion was considered in the implantation period, and radiation-enhanced effect was also considered in the damage range. The obtained result agreed well with the experiment by employing a diffusion coefficient of $1.0\text{x}10^{-8}$ exp(-0.39 eV/kT) $\text{cm}^2\,\text{s}^{-1}$, a recombination rate coefficient of $1.2\text{x}10^{-25}$ exp(-0.59 eV/kT)$\cdot T^{-1/2}$ $\text{cm}^4\cdot\text{s}^{-1}$ and two trap sites with de-trap energies of 0.85 eV and 2.2 eV and density fractions of 0.05 D/W and 0.01D/W. Since vacancy is hardly created by 100 eV D^+ ions in the tungsten material, these trap sites may have previously been created before the experiment.

Only the TDS peak corresponding to temperature of 860 K was seen in the calculated result for the material kept at 473 K in the first two periods. So, the defects with trap energy 0.85 eV were not capable of holding deuterium atoms at the temperature, indicating almost all atoms in the first two periods were trapped in traps with trap energy 2.2 eV. The amount of deuterium atoms retained at 473 K was an order of magnitude smaller than that at 300 K. The amount of retained atoms is expected to be reduced largely for temperature nearly equal to the value corresponding to trap energy of defects. The integrated fluxes released in the implanted period were estimated to be $7.49\text{x}10^{17}$ D/cm^2 and $7.78\text{x}10^{17}$ D/cm^2, resulting in 95.5% and 99.2% of the fluence $7.84\text{x}10^{17}$ D/cm^2.

The depth profiles of deuterium atoms retained in the implanted period showed that, while the location of their maxima for 300 K did not move with time, they were broadened out because of fast diffusion.

Acknowledgements

This work has been carried out under the collaborating research program of National Institute for Fusion Science.

The authors would like to thank Prof.s T. Tanabe, K. Ohya and Y. Ueda for their cooperation and discussions. This work was partly supported by KAKENHI (19055005).

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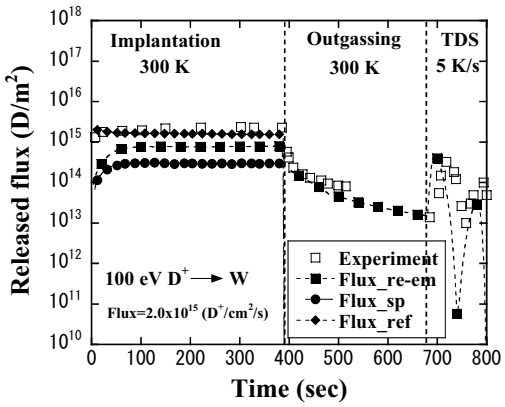

Fig.1

Fig. 1: Fluxes of thermal D_2 re-emission, reflection and self-sputtering of D from a wrought tungsten material during a time sequence of l00 eV D^+ implantation, post-implanted isothermal out-gassing and thermal desorption spectroscopy calculated with ACAT-DIFFUSE, together with the experimental result [17]. Temperature of the material was kept at 300 K in the first two periods and was raised with a ramp speed of 5 K/s in the TDS period.

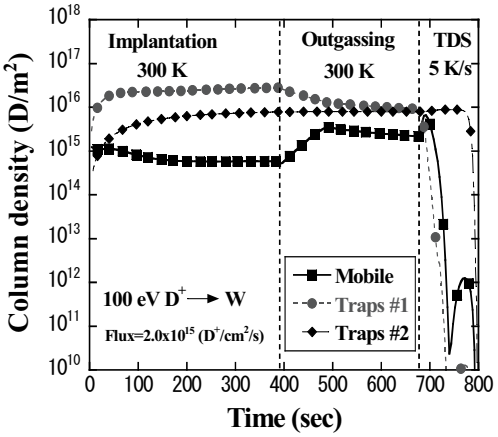

Fig. 2

Fig. 2: Colum density of mobile deuterium atoms and trapped ones in trap #1 and trap #2 in the case of Fig. 1.

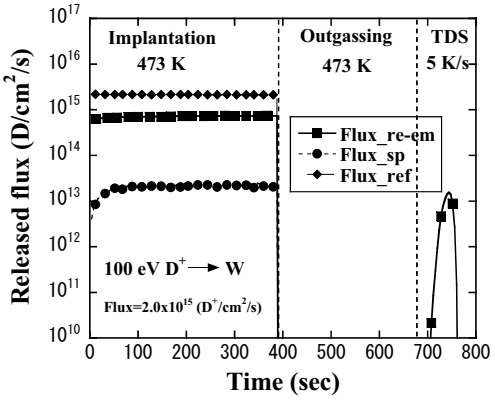

Fig. 3

Fig. 3: The same fluxes as shown in Fig. 1 except the temperature 473 K in the first two periods.

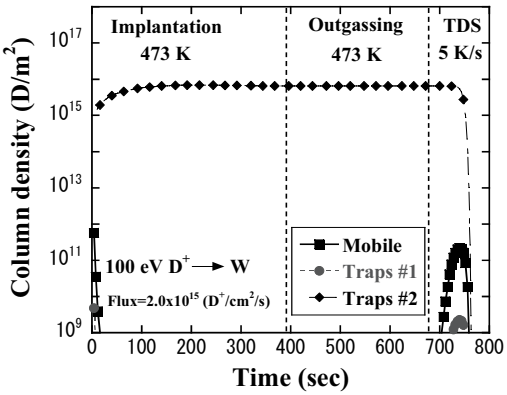

Fig. 4

Fig. 4: Colum density of mobile deuterium atoms and trapped ones in trap #1 and trap #2 in the case of Fig. 3.

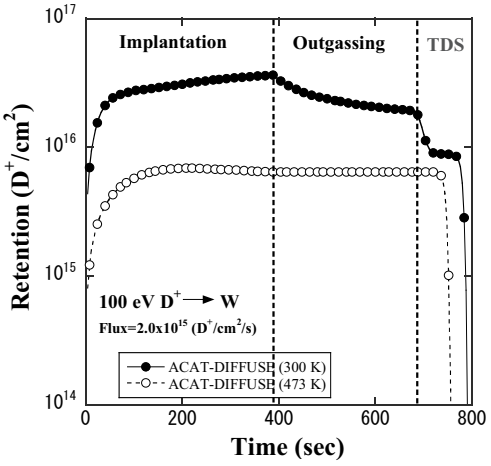

Fig. 5

Fig.5: Amounts of deuterium atoms for the tungsten material kept at 300 K and 473 K in the first two periods.

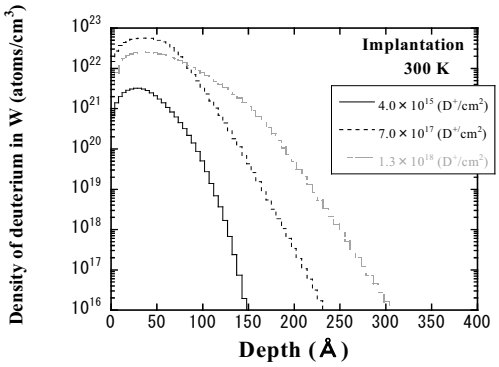

Fig. 6

Fig. 6: Depth profiles of retained deuterium atoms for fluences of $4x10^{15}$ D/cm^2, $7x10^{17}$ D/cm^2 and $1.3x10^{18}$ D/cm^2 corresponding to irradiation periods of 2 s, 350 s and 650 s, respectively.