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Interaction of tritium plasma and defects in tungsten irradiated with neutrons

Q. Xu *, K. Sato, T. Yoshiie

Research Reactor Institute, Kyoto University, Osaka 590-0494, Japan

A B S T R A C T

Multiplier effects of tritium plasma and neutron irradiation on microstructural evolution in tungsten were investigated using computer simulations based on a rate theory. The effects of irradiation temperature and dissociation energy of tritium-vacancy clusters on the interaction between tritium and defects were also investigated. At the lower temperature (473 K), vacancies trapped tritium to form tritium-vacancy clusters, which grew rapidly. When the irradiation temperature increased to 873 K, however, it was difficult for a tritium-vacancy cluster to absorb a new tritium, and thus, the concentration of tritium-vacancy clusters was about eight orders of magnitude lower than that at 473 K.

1. Introduction

Reducing the tritium inventor, which is associated with damage in the surface region of plasma-facing materials (PFMs), such as blistering and erosion, is a key issue in fusion reactors. Important criteria for choosing PFMs are a high melting point, high thermal conductivity and low sputtering erosion. Tungsten, molybdenum (high-Z materials), carbon, carbon fiber composites and beryllium (low-Z materials) have been selected as PFMs because of their outstanding thermal properties. However, recent studies have revealed that carbon-based materials retain high levels of tritium by co-deposition with eroded carbon, which could severely limit plasma operation [1–3]. Metallic materials, such as tungsten and molybdenum, prevent tritium retention, but maintaining plasma purity with high-Z materials remains a concern. PFMs must typically withstand damage produced by particles with up to 10 keV of energy and heat loads from the plasma, in addition to neutrons with high energy, high flux and high fluence, similarly to structural components. Neutron irradiation produces radiation-induced defects and changes in microstructure, and hence, physical and mechanical properties. Thus far, the tritium retention and irradiation damage produced by neutrons in PFMs have usually been investigated separately. In order to gain a better understanding of tritium retention in PFMs, such as tungsten, the present study focused on the interaction of tritium and defects produced by neutron irradiation and tritium plasma.

2. Outline of the model

To investigate the multiplier effects of tritium plasma and neutron irradiation from the standpoint of basic research, tritium with an energy of 1 keV and flux of $10^{18}/m^2$ s, and neutron irradiation at 10^{-6} dpa/s were taken as typical cases. The damage and tritium distribution in tungsten under 1-keV tritium irradiation were calculated using the TRIM code [4], where the threshold displacement energy in tungsten was assumed to be 50 eV, which depends strongly on orientation [5]. The damage is accumulated in a 3-nm-wide zone in the incident surface, and tritium is distributed from around 5–15 nm in the incident surface. Thus, in the present model, the depth distributions of damage and tritium are simply assumed to have a rectangular shape. Tritium, interstitials, vacancies and their clusters with tritium could migrate freely in the matrix, and their concentrations at both surfaces are zero at all times. The time evolution of the concentration of point defects, defect clusters and tritium were calculated using dynamic rate theory [6–8] with the following assumptions:

(1) Only tritium, interstitials, vacancies and tritium-vacancy clusters are mobile.
(2) Maximal number of tritium atoms absorbed by a vacancy is six.
(3) Thermal dissociation is considered in tritium-interstitial and tritium-vacancy clusters.

In addition, the formation of vacancy clusters and tritium-vacancy clusters (nT–mV, $m > 1$) is neglected in the present study to simplify the modeling, since the mobility of vacancies under the present conditions is relatively low.

Throughout this paper, concentrations are given in fractional units. The rate of change of concentration of interstitials, vacancies, tritium and tritium-vacancy clusters can be expressed as

* Corresponding author.
E-mail address: xu@rri.kyoto-u.ac.jp (Q. Xu).

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$$\frac{dC_i}{dt} = P + P_{D-T}(x) + D_i\frac{\partial^2 C_i}{\partial x^2} - (M_i + M_T)C_iC_T - Z_{iv}M_iC_iC_v - 2Z_{ii}M_iC_i^2 - Z_{Li}M_i(C_{Li}C_L)^{1/2}C_i - Z_{Si}M_iC_{Si}C_i + EMIT_{Ti}C_{Ti} - Z_{Ti}M_iC_i(C_{Tv} + C_{2Tv} + C_{3Tv} + C_{4Tv} + C_{5Tv} + C_{6Tv}), \quad (1)$$

$$\frac{dC_v}{dt} = P + P_{D-T}(x) + D_v\frac{\partial^2 C_v}{\partial x^2} - (M_v + M_T)C_vC_T - Z_{iv}M_iC_iC_v - Z_{vT}M_vC_vC_T - Z_{Lv}M_v(C_{Li}C_L)^{1/2}C_v - Z_{Sv}M_vC_{Sv}C_v + EMIT_{Tv}(C_{Tv} + C_{2Tv} + C_{3Tv} + C_{4Tv} + C_{5Tv} + C_{6Tv}), \quad (2)$$

$$\frac{dC_T}{dt} = P_T(x) + D_T\frac{\partial^2 C_T}{\partial x^2} - (M_i + M_T)C_iC_T - (M_v + M_T)C_vC_T - Z_{Tv}M_TC_T(C_{Tv} + C_2Tv + C_{3Tv} + C_{4Tv} + C_{5Tv}) - Z_{LT}M_T(C_{Li}C_L)^{1/2}C_T - Z_{ST}M_TC_{ST}C_T + Z_{Ti}M_iC_i(C_{Tv} + 2C_{2Tv} + 3C_{3Tv} + 4C_{4Tv} + 5C_{5Tv} + 6C_{6Tv}) + Z_{vTi}M_vC_{Ti}C_v + EMIT_{Ti}C_{Ti} + EMIT_{Tv}(C_{Tv} + 2C_{2Tv} + 3C_{3Tv} + 4C_{4Tv} + 5C_{5Tv} + 6C_{6Tv}), \quad (3)$$

$$\frac{dC_{2Tv}}{dt} = (M_T + M_{Tv})C_TC_{Tv} + D_{2Tv}\frac{\partial^2 C_{2Tv}}{\partial x^2} - M_iC_iC_{2Tv} - (M_T + M_{2Tv})C_TC_{2Tv} - Z_{L2Tv}M_{2Tv}(C_{Li}C_L)^{1/2}C_{2Tv} - Z_{S2Tv}M_{2Tv}C_{S2Tv}C_v - EMIT_{Tv}C_{2Tv}, \quad (4)$$

$$\frac{dC_{6Tv}}{dt} = M_TC_TC_{5Tv} + D_{6Tv}\frac{\partial^2 C_{6Tv}}{\partial x^2} - M_iC_iC_{6Tv} - Z_{L6Tv}M_{6Tv}(C_{Li}C_L)^{1/2}C_{6Tv} - Z_{S6Tv}M_{6Tv}C_{S6Tv}C_v - EMIT_{Tv}C_{6Tv}, \quad (5)$$

where P and P_{D-T} are the production rate of Frenkel pairs due to neutron and tritium irradiation, respectively. D is the diffusion coefficient of point defects or tritium. M_I, M_V, M_T and M_{nTv} are the mobility of interstitials, vacancies, tritium and tritium-vacancy clusters, respectively, which are related to the migration activation energy Em by $v\exp(-E_m/kT)$, where v is the jump frequency. The values of D equal a^2M, where a is one atomic distance. Details of the equation were described in Ref. [7].

P_{D-T} and P_T in Eqs. (1)–(3) are, respectively, the defect production rate and injection rate of tritium. The defects induced by tritium are only produced close to the incident surface. P_{D-T} and P_T can be expressed as

$$\begin{aligned} &P_{D-T}(x) = P_{D-T}, \quad 0 \leqq x \leqq 3 \text{ nm} \\ &P_{D-T}(x) = 0 \quad x > 3 \text{ nm} \\ &P_T(x) = P_T, \quad 5 \text{ nm} \leqq x \leqq 15 \text{ nm} \\ &P_T(x) = 0, \quad x < 5 \text{ nm}, \quad x > 15 \text{ nm}. \end{aligned} \quad (6)$$

Table 1
Parameters used in simulations.

P (s^{-1})	10^{-6} dpa/s
p_{D-T} (s^{-1})	8×10^{-5} dpa/s
p_T (s^{-1})	5×10^{-4}
E_m^I (eV)	0.15 eV [7]
E_m^V (eV)	1.4 eV [7]
E_m^T (eV)	0.39 eV [9]
$E_{Emit\ (T\text{-}I)}$ (eV)	0.5 eV [10]
$E_{Emit\ (T\text{-}Loop)}$ (eV)	1.05 eV [10]
$E_{Emit\ (T\text{-}V)}$ (eV)	1.4 eV [11]
C_S	0
Z	1
v (s^{-1})	10^{13}

The rate of change of concentration of interstitial loops, C_L, and interstitials aggregated in the loops, C_{Li}, in Eq. (1) can be expressed as

$$\frac{dC_L}{dt} = Z_{ii}M_iC_i^2, \quad (7)$$

$$\frac{dC_L}{dt} = Z_{Li}M_i(C_{Li}C_L)^{1/2}C_i - Z_{Lv}M_v(C_{Li}C_L)^{1/2}C_v. \quad (8)$$

Eqs. (4) and (5) express the time dependence of tritium-vacancy clusters. The parameters used in the present simulations are listed in Table 1, where the migration energy of tritium and dissociation energy of a tritium defect are assumed to be equal to those of hydrogen. There is little data on the migration energy of a tritium-vacancy cluster. Thus, it was assumed to be equal to the migration energy of a single vacancy. The dissociation energies of tritium-interstitial pairs, tritium-dislocation and tritium-vacancy clusters were assumed to be 0.7, 1.05 and 1.4 eV on the basis of existing experimental data [10,11].

3. Results and discussion

3.1. Tritium retention at low temperature

First, the diffusion of tritium at low temperature, where vacancies are almost immobile, is discussed. Fig. 1(a) shows the time dependence of tritium concentration in a 4.3-mm-thick tungsten sample at 473 K, without any damage produced by tritium or neutron irradiation. The concentration of tritium increased with time (except near the incident surface) and decreased markedly with increasing depth. The tritium concentration saturated to 10^{-10} after 10^4 s. Fig. 1(b) shows the time and depth dependence of tritium concentration, taking into account the damage production of tritium and neutron irradiation. In contrast to tritium diffusion in the tungsten without damage produced by neutrons and tritium plasma, tritium was concentrated near the incident surface in the presence of damage produced by neutrons and tritium, though the tritium concentration peak was 10^{-10}. For elapsed irradiation time up to 0.1 s, the tritium concentration in the bulk first increased, and then decreased with irradiation time. The highest concentration of tritium near the incident surface was the same as that shown in Fig. 1(a). The decrease in tritium concentration with increasing irradiation time after 0.1 s suggests that most of the injected tritium was trapped by radiation-induced defects, and thus diffusion of tritium into the bulk of the specimen decreased.

In order to elucidate the interaction of tritium and defects, especially vacancies, the formation of tritium-vacancy clusters was investigated. Fig. 2(a) shows the depth distribution of tritium-vacancy clusters, i.e., 1T-V clusters, for several irradiation times. The 1T-V clusters accumulated was concentrated at 3 nm, which was the damage peak region produced by 1-keV tritium. The concentration of 1T-V clusters decreased dramatically in the area free of damage by tritium ions. With elapsed irradiation time, the concentration of 1T-V clusters increased, exceeding 10^{-9} after 10^4 s even at a depth of 0.02 mm, which is about three orders of magnitude deeper than the ion range produced by 1-keV tritium. This suggests that tritium is also trapped by vacancies produced by neutron irradiation to form tritium-vacancy clusters.

Time and depth dependence of formation of tritium-vacancy clusters 2T-V, 3T-V, 4T-V and 5T-V are similar to those of 1T-V, though the concentration of tritium-vacancy clusters decreases slightly with increasing number of tritium atoms per cluster. The concentration of tritium-vacancy cluster 6T-V (see Fig. 2(b)), however, was 40 times higher than that of 1T-V. This indicates that there are enough tritium atoms in the matrix to be absorbed by the tritium-vacancy clusters. The tritium-vacancy clusters with a

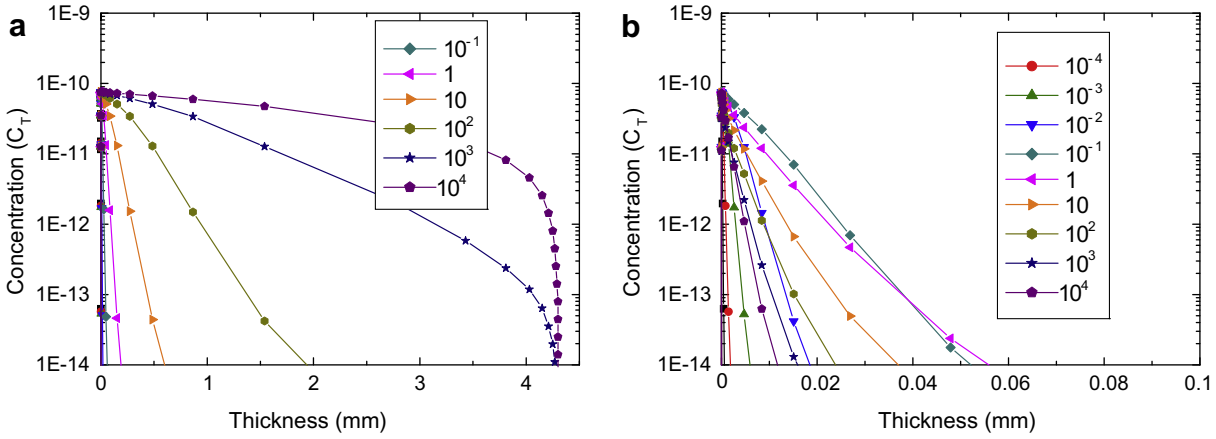

Fig. 1. Time and depth dependence of mobile tritium concentration in tungsten at 473 K with an energy of 1 keV and flux of $10^{18}/m^2$ s in the absence of damage produced by tritium and neutron irradiation (a), and in the presence of damage by tritium and neutron irradiation (b). The irradiation time (in seconds) for each curve is shown in the insert legend.

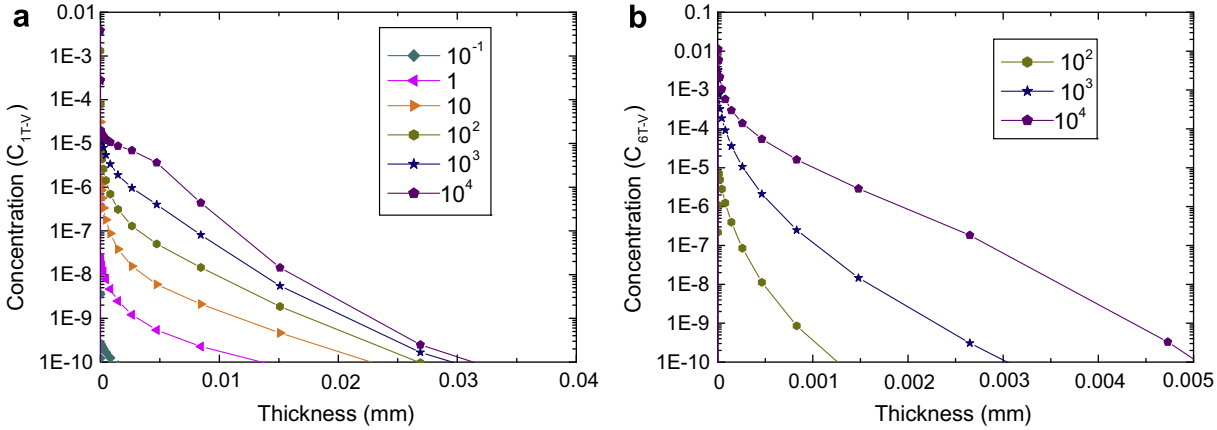

Fig. 2. Time and depth dependence of formation of tritium-vacancy clusters 1T-V (a) and 6T-V (b) under tritium irradiation with an energy of 1 keV and flux of $10^{18}/m^2$ s and neutron irradiation at 473 K. The irradiation time [s] for each curve is shown in the insert legend.

small number of tritium atoms absorb the tritium immediately in order to grow.

3.2. Tritium retention at high temperature

We now turn to the diffusion of tritium at high temperature, where the vacancies are mobile. Compared with the tritium distribution in tungsten at low temperature (473 K), shown in Fig. 1(a), the distribution of tritium in this case rapidly reached steady state (100 s). The saturation concentration of tritium near the incident surface at 873 K was two orders of magnitude lower than that at 473 K. The tritium distribution and the change of tritium concentration in the matrix with damage at 873 K were the same as those at 473 K, as shown in Fig. 1(b), but the tritium concentration near

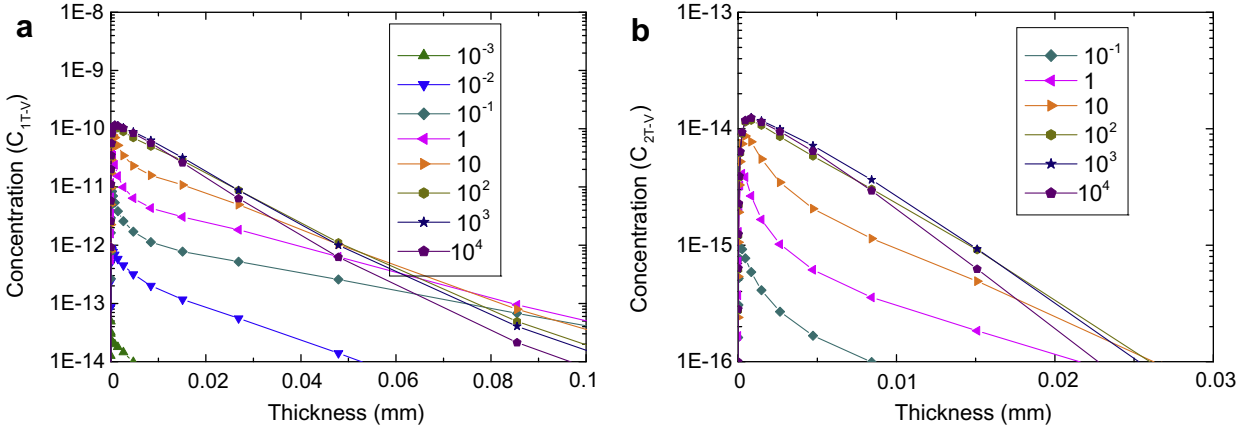

Fig. 3. Time and depth dependence of formation of tritium-vacancy clusters 1T-V (a) and 2T-V (b) under tritium irradiation with an energy of 1 keV and flux of $10^{18}/m^2$ s and neutron irradiation at 873 K. Irradiation times [s] for all curves are shown in the insert legend.

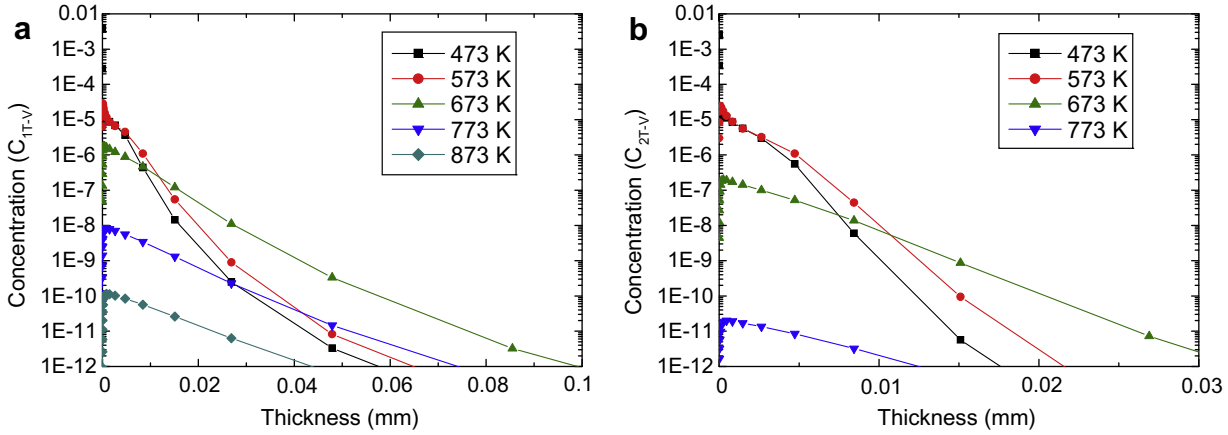

Fig. 4. Temperature dependence of tritium-vacancy clusters 1T-V (a) and 2T-V (b) under tritium irradiation with an energy of 1 keV and flux of $10^{18}/m^2$ s and neutron irradiation. Irradiation times [s] for all curves are shown in the insert legend.

the incident surface was two orders of magnitude lower than that at 473 K, and the tritium distribution was deeper than that at 473 K.

Fig. 3 shows the time dependence of 1T-V clusters and 2T-V clusters at 873 K. With increasing irradiation temperature, the behavior of tritium-vacancy cluster formation differed from that at 473 K. First, the tritium-vacancy clusters with high concentration near the incident surface disappeared. Second, the distribution of tritium-vacancy clusters in the matrix reached saturation after 100 s, and the saturation concentration of 1T-V clusters near the incident surface was about seven orders of magnitude lower than that at 473 K. Finally, the concentration of tritium-vacancy clusters decreased with increasing number of tritium atoms per cluster. The vacancies are sufficiently mobile and the tritium-vacancy clusters were unstable at 873 K, thus, vacancies near incident surfaces were mobile enough to sink, and the peak of tritium-vacancy clusters formation disappeared near the incident surface. In addition, the instability of 1T-V clusters makes it difficult to grow larger clusters absorbing the tritium, and the concentration of tritium-vacancy clusters decreased with increasing number of tritium atoms per cluster.

The present study also investigated the temperature dependence of tritium-vacancy cluster formation. Fig. 4 shows the temperature dependence of 1T-V clusters (a) and 2T-V clusters (b) after 10^4 s. The tritium-vacancy clusters with high concentration near the incident surface disappeared from 673 K, and the concentration of tritium-vacancy clusters decreased with increasing number of tritium atoms per cluster. This indicates that tritium retention decreases as temperature increases. The experimental data obtained by Igarashi et al. agreed with our simulation results [12].

3.3. Effects of dissociation energy of tritium-vacancy clusters on the formation of defect clusters

The dissociation energy of tritium-vacancy clusters is assumed to be 1.4 eV in the present study; however, some reports indicate that deuterium-vacancy clusters are unstable even at 473 K [13]. Thus, here, we investigate the effects of the dissociation energy of tritium-vacancy clusters on the formation of defect clusters. Compared with the results shown in Fig. 4, where the dissociation energy of tritium-vacancy clusters is 1.4 eV, 1T-V and 2T-V clusters with high concentration near the incident surface disappeared at the same temperature (673 K), while the concentrations of 1T-V and 2T-V clusters decreased by two and five orders of magnitude, respectively. These results indicate that the disappearance of tritium-vacancy clusters with high concentration, induced by tritium ions with 1 keV, is governed by the temperature of mobile vacancies. The vacancies produced by 1-keV tritium ions move to the surface at 673 K, and the concentration of tritium-vacancy clusters decreases. In addition, tritium-vacancy clusters become increasingly unstable with decreasing dissociation energy. It is difficult for tritium-vacancy clusters to absorb a new tritium atom to grow. Thus, the concentration of tritium-vacancy clusters decreases with decreasing dissociation energy of tritium-vacancy clusters. A future study will focus on multiplier effects of tritium plasma and neutron irradiation on microstructural evolution in tungsten under high tritium flux with a view toward improving the performance of tungsten in a divertor or a tokomak.

4. Conclusion

To estimate the effect of the tritium plasma on the microstructural evolution induced by neutron irradiation, computer simulations based on the rate theory considering both tritium and defect diffusion were performed. Tritium diffusion in 4.3-mm-thick tungsten free of damage by tritium ions and neutrons only took 10^4 s and 100 s to reach saturation at 473 K and 873 K, respectively. Irradiation-induced defects enhance the accumulation of tritium in the matrix. At lower dissociation energies of tritium-vacancy clusters, it is more difficult for tritium-vacancy clusters to grow by absorbing tritium atoms. These results indicate that tritium trapped near the incident surface is an important issue at high dissociation energies of tritium-vacancy clusters, whereas tritium diffusion into the bulk becomes important at low dissociation energies of tritium-vacancy clusters.

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