

Dynamical Simulation of Tritium Depth Profile due to 130 keV D⁺ Ion Bombardment on a TiT Target

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Abstract

The depth profiles of tritium atoms in a TiT target at liquid nitrogen temperature induced by 130 keV deuterium ion bombardment have been studied, using the hybrid Monte Carlo simulation code ACAT-DIFFUSE. The present ACAT-DIFFUSE code can estimate the slowing down process of incident ions and the thermal process of moderated atoms. It is found that the experimental depth profile is explained by the Doyle's local mixing model and the target expansion brought out by the accumulated deuterium atoms or recoil tritium atoms which cannot enter the interstitial sites. Another interesting feature is that the range of the implanted deuterium ion becomes shorter as the ion fluence increases.

Keywords:

Tritium depth profile, Monte Carlo simulation, neutron generator, replacement collision, local mixing model, deuterium ion bombardment,

1. Introduction

For generating 14 MeV neutrons from the $T(d,n)^{3}$ He reaction, a metal tritide layer is commonly used as the tritium target. Nuclear reactions producing the neutrons take place at depth within a few micrometers from the surface. In order to know the details of the energy spectrum and the neutron yield, it is necessary to get the precise information about the depth profile of the tritium atom in a metal tritide target induced by deuterium ion bombardment. The saturation and replacement of hydrogen isotopes implanted in materials have been studied experimentally [1-3] and theoretically [4-7]. A lot of works have been made on behavior of hydrogen and deuterium atoms successively implanted into metals to simulate the behavior of tritium and deuterium atoms in the first walls of thermonuclear fusion reactors. The experimental

results showed that the atoms implanted earlier are partly removed by the later implantation, making a dip on the depth profile of the former atoms around the projected range of the latter [7,8].

In 1981 Doyle and his coworkers [5] studied the hydrogen retention and replacement properties of the materials by implanting H and D at an energy of 1.5 keV/ion. They proposed successfully the so-called local mixing model (LMM) in order to explain the experimental saturation curves for various materials. Okuda, Yamamoto and Fujishiro [8,9] made the insitu measurement of the depth profiles of the tritium atom in a titanium tritide target at liquid nitrogen temperature with the method of the ion beam analysis using the T(d, α)n nuclear reaction, and they also observed a dip on the depth profile of the tritium atom around the projected range of the latter implanted 研究論文

deuterium ion. They explained their depth profiles by the "up-hill migration" of tritium atom in materials near around the projected range of the latter implanted deuterium ion [10].

The main concern of this paper is to simulate the Okuda, Yamamoto and Fujishiro's work by our Monte Carlo simulation code ACAT-DIFFUSE [11]. At present there are several Monte Carlo computer simulation programs which can estimate the ion-fluence dependence of depth profiles and compositional changes during ion bombardment, i.e., EVOLVE [12], TRI-DYN [13] and ACAT-DIFFUSE [11], etc. These three codes are based on the binary collision approximation. The EVOLVE code was developed by Roush et al., TRIDYN was by Möller and Eckstein, and ACAT-DIFFUSE was by Yamamura. The ACAT-DIFFUSE code can estimate the thermal process such as the diffusion and the segregation during ion bombardment.

2. The simulation model

The detailed description of the ACAT-DIFFUSE program, which is based on the ACAT [14] and the DIFFUSE code [15], was given elsewhere [11]. Therefore, only the main feature concerning the present problem will be addressed here. In the ACAT-DIF-FUSE code, the total dose Φ is divided into sufficient small dose $\Delta \Phi$ during which the ion bombardment does not change the target composition appreciably. In the ACAT-DIFFUSE simulation the ions corresponding $\Delta \Phi$ are assumed to be bombarded at instance and be slowed down instantaneously ($< 10^{-12}$ s), and their slowing down and the associated collision cascade are calculated by the ACAT routine of the ACAT-DIF-FUSE code. These thermalized ions and recoil atoms diffuse during the period of $\Delta \Phi/J$ which is of the order of second usually (J being the current density) which is estimated by solving the diffusion equations numerically in the DIFFUSE routine in the ACAT-DIFFUSE program. In the ACAT-DIFFUSE program these procedures are repeated n times, where $n = \Phi/P$ $\Delta \Phi$. Therefore, the logical representation of the ACAT-DIFFUSE program is as follows:

$$\begin{array}{l} ACAT-DIFFUSE \\ = [(ACAT)(DIFFUSE)]^{n} \quad . \qquad (1) \end{array}$$

The slowing down process is calculated by the

ACAT routine which was developed to simulate the atomic collision in amorphous solids based on the binary collision approximation. After each operation of the ACAT-DIFFUSE cycle, the atomic densities of component elements in the medium are calculated taking into account depth distributions of implanted atoms, interstitial atoms and vacancies. In other words, the atomic fraction of each atom including implanted atom is a function of ion fluence and depth. In the slowing down process a newly implanted deuterium ion has a chance to collide with previously implanted deuterium atoms. In the ACAT routine, the implanted particle and the recoil atom are followed throughout their slowing-down process until their energies fall below predetermined cutoff energies E_{c} . The displacement threshold energy of a target Ti atom is set to be 25 eV, and that of an interstitial hydrogen isotope is 5 eV. The Kr-C potential [16] is used as an interatomic potential for elastic scattering. The electronic energy loss in solids is estimated by using the path-dependent non-local model, where the parameters included in the electronic stopping formula are calculated from the Ziegler's table [17].

At low temperature (~110 K), it is reasonable to consider the interstitial and the vacancy as the trapping sites and to assume that there are three states for the implanted deuterium atom and recoil tritium atom, i.e., a free-state atom which can migrate quickly with the migration energy, a trapped atom by trap 1 (interstitial) and a trapped atom by trap 2 (vacancy). Then, the activation energy Q_j^{I} of the interstitial diffusion of the *j*th element can be written as $Q_j^{I} = E_{mj} + E_{Tj}^{I}$, where E_{mj} and E_{Tj}^{I} are the migration energy and the detrapping energy from the interstitial site, respectively. Similarly, the activation energy Q_j^{V} of the vacancy diffusion of the *j*th element is given as $Q_j^{I} = E_{mj} + E_{Tj}^{I}$, where E_{Tj}^{V} is the detrapping energy from the lattice site.

The DIFFUSE routine of the ACAT-DIFFUSE code solves numerically the diffusion equation of the *j*th solute concentration $c_j(x,t)$ with a source term $G_j(x,t_0)$ given by the range distribution of the implanted atom or by the relocation distribution of detrapped atoms and with the trapping term taking into account the capture into and the release from two independent trap distributions. At low temperature the basic equation is the diffusion equation for the migrating particle which is given by

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$$\frac{\partial c_{j}(x,t)}{\partial t} = \nabla \left[D_{j} \nabla c_{j}(x,t) + G_{j}(x,t_{0}) \right] \\ - \sum_{i=1}^{2} \frac{\partial c_{Tj}^{i}(x,t)}{\partial t} , \qquad (2)$$

where t_0 is the time at which the DIFFUSE routine of the ACAT-DIFFUSE code begin to start. The diffusion constant D_j of the *j*th migrating element (free state) is given as

$$D_j = D_{0j} \exp[-E_{mj}/kT]$$
, (3)

where T is the temperature and k is the Boltzmann constant.

The concentration c_{Tj}^i of the *j*th atom trapped in the *i*th trap site satisfies the following rate equation:

$$\frac{\partial c_{Tj}^{i}(x,t)}{\partial t} = D_{j}c_{j}(x,t)C_{T}^{i}(x,t)/\lambda^{2}$$
$$-c_{Tj}^{i}(x,t)\nu_{0}\exp\left[-E_{Tj}^{i}/kT\right] \quad . \tag{4}$$

The first term on the right hand side of eq. (4) means the capture into the trap site, and the second term means the thermal detrapping from the trap site. The jump distance is denoted by λ in eq. (4), ν_0 is the detrapping attempt frequency, and $C_{\rm T}^{\rm i}(x,t)$ is the degree of the vacancy of the *i*th trap given as

$$C_{\rm T}^{\rm i}(x,t) = C_{\rm T}^{\rm i}(x,t_0) - \sum_j f_j^{\rm i} c_{Tj}^{\rm i}(x,t) \quad , \qquad (5)$$

where $C_{\rm T}^i(x,t_0)$ is the degree of the vacancy of the *i*th trap at $t = t_0$ which is calculated by the ACAT routine, and f_j^i is the effective volume of the *j*th atom for the *i*th trap.

If we couple eqs. (2) and (4), we can describe both the interstitial diffusion and the vacancy diffusion. At low temperature of present interest there is neither the usual interstitial diffusion nor the usual vacancy diffusion. In the collision cascade region, however, the trapped atom is detrapped due to the elastic collision even if the atom cannot jump out from the trap site thermally. This radiation-induced detrapping promotes the diffusion effectively. This process can be regarded as a kind of the radiation-induced diffusion [18].

In the present simulation the surface segregation is not included, but the radiation-induced segregation is taken into account by introducing the atomic-volume dependent recombination radius. As the boundary condition of the diffusion equation eq. (2), we employed the zero surface concentration for hydrogen isotopes, because the transport in this problem is the diffusion-limited process since the Doyle's transport parameter W [6] is much larger than unity for the present case.

3. Simulated results and discussion

The present interest is to simulate the tritium depth profiling in a TiT target under the bombardment of 130 keV deuterium ions, where we assume the tritium atoms are uniformly absorbed in the interstitial positions of a Ti target before deuterium bombardment. Since the tritium atom is located in the interstitial site in a Ti target, the density of TiT_s is calculated by the following formula [19]:

$$\rho_{\mathrm{TiT}_{\mathrm{s}}} = (1 - \Delta) \frac{M_{\mathrm{Ti}} + sM_{\mathrm{T}}}{M_{\mathrm{Ti}}} \rho_{\mathrm{Ti}}$$
(6)

where M_{Ti} and M_{T} are the atomic masses of Ti and tritium atoms, respectively, and ρ_{Ti} is the density of a Ti target, and Δ is the the expansion factor which the titanium target undergoes during the TiT production.

One of the most important parameters for the simulation is the saturated concentration of the tritium atom in a Ti target at liquid nitrogen temperature. In the present simulation we assume the saturated concentration of the absorbed tritium atom in the Ti target is equal to that of the target Ti atom at liquid nitrogen temperature, *i.e.*, s=1. For TiT we used $\Delta=0.15$, and so $\rho_{\rm TIT} = 4.07$ g/cm³. Since s=1 means the saturated target, $C_{\rm T}^{\rm i}(x,0)=0$ before deuterium bombardment. Therefore, $C_{T}^{i}(x,t)$ is equal to the radiation-induced detrapping distribution. Figure 1 shows the spatial distribution of the implanted deuterium atom, the relocation profile of the recoil tritium atom and the depth profile of the $C_{T}^{I}(x,t_{0})$ produced by 130 keV deuterium ion bombardment on the TiT target. As a matter of course, the detrapping distribution is nearly equal to the relocation profile of the recoil tritium atom, because the spatial mesh interval for the depth profile of Fig. 1 is large ($\Delta x = 0.05 \ \mu m$).

An important point of Fig. 1 is that the density of the vacant interstitial site is by two order of magnitude larger than that of the implanted deuterium atom near $0.9 \ \mu m$. Just after the slowing down process has finished, near the range of the implanted deuterium, there are a lot of vacant interstitial sites which are 研究論文



Fig. 1 Depth profiles of vacant interstitial sites, relocated recoil tritium atoms and implanted deuterium atoms which are induced by 130 keV deuterium ion bombardment on the TiT target, where each profile is scaled by one incident ion.



Fig. 2 Experimental tritium depth profile [8,9] induced by 130 keV deuterium ion bombardment on the TiT target at 111 K. Solid symbols and solid lines: observed; open symbols and dashed lines: corrected for deuterium electronic stopping power.

produced by the elastic collisions between implanted deuterium atom and the interstitial tritium atom or between the energetic recoil tritium atom and the interstitial tritium atom. In thermal process these vacant interstitial sites are filled by thermalized neighboring tritium atoms or deuterium atoms. Since the atomic volume of tritium atom is about 1.5 times larger than that of deuterium atom, these vacant interstitial sites are preferentially filled by tritium atoms. If the distance between the vacant interstitial site and the thermalized deuterium atom is sufficiently small, there is a chance that the vacant interstitial site is filled by the deuterium atom. Broadly speaking, this process can be regarded as the replacement collision.

Figure 2 shows the ion-fluence dependence of the experimental tritium depth profiles in the TiT target induced by 130 keV deuteron bombardment at 110 K [9,10], and the ratio $c_{\rm T}/c_{\rm Ti}$ (bulk) is plotted against depth (mg/cm²), where c_{Ti} (bulk) means the Ti atomic density in the bulk. Figure 3 shows the ACAT-DIF-FUSE tritium depth profiles induced by 130 keV deuteron bombardment on the TiT target at 110 K, where the ratio $c_{\rm T}/c_{\rm Ti}$ (bulk) is also plotted against depth. In the simulation, we used $E_{\rm m}=0.3$ eV, $E_{\rm T}^{\rm I}=0.4$ eV, and $E_{\rm T}^{\rm V}=1.0$ eV for all hydrogen isotopes, and D_{0i} is inversely proportional to a square root of the atomic mass. Another important parameter is how far the deuterium atoms or recoil tritium atoms in the free state contribute to the target expansion. We assumed the 15% of their atomic volumes contribute to the target expansion. Other physical quantities used in the simulation are the same as the experimental values, *i.e.*, $J=1.56\times10^{13}$ D atoms/cm² and T=110 **K**.

Though the experimental tritium depth profile is not flat, especially near the surface, we can observe two interesting features from the experimental depth profiles. The one is that the minimum position of the depth profile is gradually shifted to the surface as the ion fluence increases. The other is that a FWHM of the dip profile become the broader as the ion fluence increases. These two interesting features are clearly reproduced in the tritium depth profile (see Fig. 3) simulated by the ACAT-DIFFUSE code.

Figure 4 shows the tritium depth profiles simulated by the ACAT-DIFFUSE code under the condition that there is no target expansion, where other parameters used in the simulation are the same as those of プラズマ・核融合学会誌 第71巻第12号

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Fig. 3. There is a big difference between Fig. 3 and Fig. 4. The tritium depth profiles of Fig. 4 have the strong asymmetric distribution, and the over-saturated density is observed in the deeper region due to recoil implantation. The profile in the deeper region (>0.4 mg/cm²) does not change appreciably, but in the shallow region the depth profile is influenced appreciably by deuterium bombardment. The minimum point of the dip profile is shifted more rapidly toward the surface than that of Fig. 3. This means that the projected range become shorter as the accumulation of implanted





deuterium atoms proceeds. In Fig. 5 we plot the projected range and straggling of implanted deuterium atoms as a function of ion fluence. The ion-fluence dependence of the range and straggling with no expansion (corresponding to Fig. 4) is much stronger than the case that the target expansion effect is included (corresponding to Fig. 3).

Yamamoto *et al.* [10] explained their experimental tritium depth profiles in terms of the tritium "uphill" migration due to the change of the chemical potential near the range of the implanted deuterium 研究論文



lon fluence (1018 D atoms/cm2)

Fig. 5 Plot of the projected range and the straggling of the implanted deuterium ion as a function of ion fluence, where the incident energy of a deuterium ion is 130 keV. The solid symbols correspond to Fig. 4 and the open symbols to Fig. 3.

atom. When one watch only the behavior of the tritium atoms, one should consider the tritium "up-hill" migration to explain the dip profile near the range. The chemical potential is composed of the density-independent component and the density-dependent component [20,21]. The difference of the former component between two neighboring layers is appreciable only near the surface, which is known as the Gibbsian segregation, and the difference is small in the bulk. Therefore, in the present problem, we have only to consider the density-dependent component. It is known that the density-dependent component of the chemical potential does not bring out the "up-hill" migration, but the diffusion-like migration [21].

From the ACAT-DIFFUSE simulation, it is found that the tritium depth profile in a TiT target induced by 130 keV deuterium bombardment is completely explained by the LMM [3] and the target expansion due to accumulated deuterium atoms and recoil tritium atoms which cannot enter the interstitial positions in the Ti target. As shown in Fig. 1, in the mixing area there are a lot of thermalized recoil tritium and vacant interstitial sites and implanted deuterium atoms the amount of which depends on the ion fluence. Almost all recoil tritium atoms recombine with the vacant interstitial sites instantaneously. Even if the ion fluence increases, the magnitude of the detrapping distribution or $C_{\rm T}^{\rm I}(x,t_0)$ induced by the implanted deuterium ion does not increase appreciably. The density of implanted deuterium atoms increases linearly as the ion fluence increases. At high fluence, the density of the implanted deuterium atom is larger than that of recoil tritium atoms. As a result, an appreciable amount of implanted deuterium atoms combine with the vacant interstitial sites. The tritium atoms and deuterium atoms which cannot enter the interstitial sites are in a free-state and expand the target. This results in the difference between Fig. 3 and Fig. 4.

4. Conclusions

The ACAT-DIFFUSE code is applied to the investigation of the tritium depth profile in a TiT target induced by 130 keV deuterium ion bombardment at liquid nitrogen temperature. The experimental depth profile is found to be explained by the Doyle's local mixing model and the target expansion which is brought out by the accumulated deuterium atoms or recoil tritium atoms which cannot enter the interstitial sites. Another interesting feature is that the range of the implanted deuterium ion becomes shorter as the ion fluence increases.

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