ELSEVIER

Journal of Nuclear Materials 248 (1997) 439–442

journal of nuclear materials

Measurement of a knock-on process induced by an ion beam

Makoto Teshigawara *, Kenji Konashi, Hideo Kayano

The Oarai Branch, Institute for Materials Research, Tohoku University, Oarai, Ibaraki, 311-13, Japan

Abstract

Understanding of collision processes, which are induced by irradiation of neutrons and ions, is important for predicting the material performance in operational fission and proposed fusion reactors. Specially, an energetic primary knock-on atom (PKA) plays a crucial role in the production of collision cascade damage. The target of single phase $TiD_{1.61}$ was irradiated by ion beams of ^4He and ^3He with energies of 50, 100, 150 and 200 keV. In the experiments with a ^3He ion beam, the deuterium density in the target was monitored by the detection of a nuclear reaction of $D(^3He, p)\alpha$. A high energy primary knock-on process has been successfully observed by monitoring the nuclear reaction of D(d, p)t. Experimental results were compared with the results calculated by the Monte Carlo code TRIM. It was confirmed that the Monte Carlo code can successfully predict the production of energetic primary knock-on atoms under irradiation of the ion beam. © 1997 Elsevier Science B.V.

1. Introduction

Understanding of collision processes, which are induced by irradiation of neutrons and ions, is important for predicting the material performance in operational fission and proposed fusion reactors. Specially, an energetic primary knock-on atom (PKA) plays a crucial role in the production of collision cascade damage [1]. Radiation damages of nuclear materials are mainly evaluated by post-irradiation examination, that is, microscopic observation (e.g., TEM, SEM) and mechanical testing (e.g., tension test and Vickers hardness) [2]. Microstructural evolution of materials is observed by an experiment using an ion beam from an accelerator. There are, however, very few data on the direct observation of the radiation induced collision process, which are important in understanding the basic process involved in the production of radiation damage.

In this study, we used a target material of titanium deuteride, TiD_x, to monitor the knock-on process in a solid target by nuclear reaction. It is necessary to disperse the nuclear reaction particle, a D atom, homogeneously in the target for comparison of the experimental results with numerical prediction. The crystal structure of titanium deuteride is well known and it is easy to make a single phase of its deuteride composition. TiD_x targets were bombarded by an ion beam of ^4He. When the ion from the accelerator traverses the lattice of a crystalline solid of the target, it may lose energy by means of direct nuclear collisions (i.e., elastic collisions) and inelastic collisions with the electrons of atoms. If in an elastic collision the lattice atom receives an energy in excess of the binding energy of the atom in its lattice site, it is released from its normal lattice site. If the atom receives a high energy in an elastic collision, it can produce knock-on atoms of higher orders, that is, collision cascade. The nuclear reaction of D(d, p)t between a knock-on D atom and a lattice D atom were observed in this study by detection of protons from the nuclear reaction.

The deuterium density in the target is important to estimate the nuclear reaction yield. The $D(^3He, p)\alpha$ nuclear reaction has been used for the measurement of the deuterium density in materials [3]. Experiments with an ion beam of ^3He have been also done to monitor the D density in the target by the nuclear reaction. In the experiment with ^3He, both nuclear reactions of $D(^3He, p)\alpha$ and D(d, p)t have been simultaneously observed. This is useful to reduce the experimental uncertainty about the D density for the evaluation of the nuclear reaction yield. Experimen-

* Corresponding author. Tel.: +81-29 267 3181; fax: +81-29 267 4947; e-mail:teshi@ob.imr.tohoku.ac.jp.

PII S0022-3115(97)00165-7

tal results of the nuclear reaction yields are compared with the calculation results by the Monte Carlo computer code.

2. Experimental procedure

The experimental setup is schematically shown in Fig. 1. Ions of $^3He^+$ and $^4He^+$ were produced in a RF plasma ion source and were then accelerated by a Cockcroft–Walton type accelerator. The 45° focusing magnet was used for selection of ions. The length of the ion path from the ion source to the target is about 7 m. The ion beam entered into the target through a slit with a diameter of 7 mm. The beam current was measured by a retractable Faraday cup before and after irradiation. The change of beam current was monitored by the target current during irradiation.

As described above, the deuterium density in the target is important to estimate the nuclear reaction yield. The deuterium is easily released from the target surface heated by ion beam irradiation. The TiD_x target was attached on a Cu target holder, which was cooled by liquid N_2. The temperature of the Cu target holder and the ion beam current were measured during irradiation. The temperature was almost constant, about 140 K, during irradiation. It is clear that the TiD_x target was cooled enough not to release deuterium in the target. Experiments were conducted by ion beams with energies of 50, 100, 150 and 200 keV. The ion beam currents were sustained at 0.1 to 10 μA.

Protons from a nuclear reaction of D(d, p)t and α particles from D(^{3}He, p)α were detected by the silicon detector with 500 μm sensitive depth and 200 mm^2 active area. The detector was placed at an angle of $\theta_{lab} = 120°$ with respect to the beam axis and at a distance of 94 mm from the target. Aluminum foil was placed in front of the detector to stop the intense elastically scattered particles. The thickness of aluminum foil was selected to be 6 μm, considering separation of the proton and α particle.

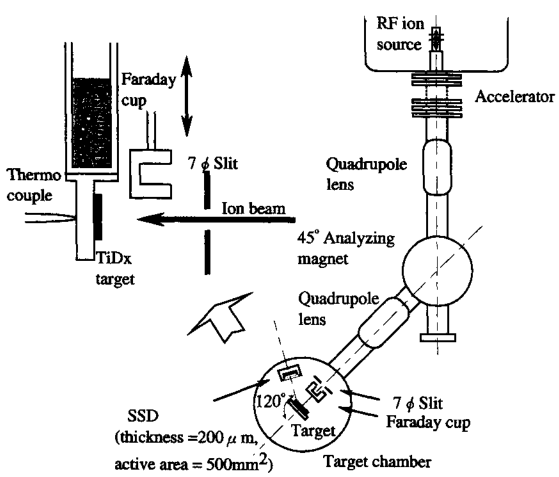

Fig. 1. Schematic diagram of the experimental setup.

Although the energy of a proton (3.02 MeV) from D(d, p)t is close to that of an α particle (3.68 MeV) form D(^{3}He, p)α, these charged particles can be separated after passing through the aluminum foil due to the different stopping powers of each particle. The energy calibration of the detector was determined with a ^{241}Am source, which was placed in the position of the target. The efficiency of the silicon detector was found to be 1.97×10^{-3} pulse/particle in 4π sr.

Target samples ($10 \times 15 \times 0.6$ mm^3) were prepared by deuterium absorption method as follows: Ti of 99.95 wt% purity was etched by an acid of H_2O:HNO_3:HF = 10:4:1 and then outguessed at 1073 K in vacuum until the residual pressure reached 10^{-5} Pa. The target samples were placed in an absorption vessel filled by deuterium gas at a pressure below 0.2 MPa. The target samples slowly absorbed deuterium gas keeping the equilibrium condition at the temperature in the absorption vessel. The deuterium density in the target was controlled by the temperature.

A homogeneous sample is important for calculation of the nuclear reaction yield in solids. After absorption, the target sample was dropped into cooling oil to keep the crystal structure. The formation of single phase (δ phase) $TiD_{1.61}$ in the target samples was confirmed by X-ray diffraction measurements. The density of deuterium in the target was calculated by the weight change of the deuterium absorbed target.

3. Monte Carlo code simulation of nuclear reaction yield

The collision processes in the irradiated TiD_x target are simulated by the Monte Carlo code TRIM [4], which calculates the 2-dimensional ion penetration in solids. The TRIM code, however, is not designed to calculate the nuclear reaction, but is mainly used to obtain information on the ion distribution, the energy loss distribution, lattice displacements, and so on. Calculation results by the TRIM code are used to estimate the yield of the nuclear reactions as follows: as shown in Fig. 2 the TRIM code simulates sequential collision events in solid. The particle moves a distance (track length) with constant energy between collision events. The probability of the nuclear reaction of D(d, p)t for the ith track length of a recoiled d-atom with an energy E_d at position $\vec{r}$, $P_i(\vec{r}, E_d)$, is calculated by

$$P_i(\vec{r}, E_d) = l_i(\vec{r}, E_d) \times \sigma_{d-d}(E_d) \times \rho_d, \quad (1)$$

where $l_i(\vec{r}, E_d)$ is the ith track length of the recoil d-atom, which is calculated by the TRIM code. ρ_d is the density of the d-atom in the target. For the analysis of experimental results with a ^{3}He beam, the probability of the nuclear reaction of D(^{3}He, p)α, $Q_i(\vec{r}, E_{He})$, is similarly calculated by

$$Q_i(\vec{r}, E_{He}) = l_i(\vec{r}, E_{He}) \times \sigma_{He-d}(E_{He}) \times \rho_d, \quad (2)$$

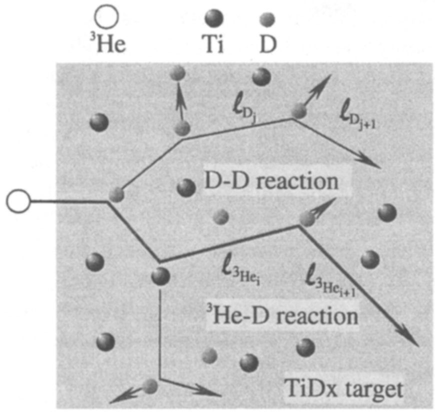

Fig. 2. Diagram of simulation of collision cascade.

where $l_i(\vec{r}, E_{He})$ is the ith track length of incident ^{3}He. ρ_d has the same meaning as in Eq. (1).

The total probabilities per incident ion at the position $\vec{r}$ are calculated as a function of reaction energy by the sum of the probabilities for each track,

$$P(\vec{r}, E_d) = \sum P_i(\vec{r}, E_d)/N_{ion}, \quad (3)$$

and

$$Q(\vec{r}, E_{He}) = \sum Q_i(\vec{r}, E_{He})/N_{ion}, \quad (4)$$

where N_{ion} is the incident ion number used in the Monte Carlo calculation.

The d–d fusion cross-section, $\sigma_{d-d}(E_d)$ (barn), is expressed as a function of the laboratory energy E_d (keV) of deuterium for the reaction of D(d, p)t [5],

$$\sigma_{d-d}(E_d) = 2.0\frac{S(E_d)}{E_d}\exp\left(-\sqrt{\frac{E_G}{E_d}}\right), \quad (5)$$

where E_G is the Gamow energy ($\sqrt{E_G} = 44.40$ keV$^{1/2}$). The S-factor, $S(E_d)$, is calculated by

$$S(E_d) = \left(S_0 + S_1 E_d + S_2 E_d^2 + S_3 E_d^3 + S_4 E_d^4\right) \exp\left(-\beta\sqrt{E_d}\right), \quad (6)$$

where $S_0 = 56.46$, $S_1 = 0.2393$, $S_2 = 5.922 \times 10^{-6}$, $S_3 = 5.786 \times 10^{-11}$, $S_4 = -1.151 \times 10^{-15}$ and $\beta = 1.587 \times 10^{-2}$. The ^{3}He – d reaction cross-section, $\sigma_{He-d}(E_{He})$ (barn), is also calculated by [5]

$$\sigma_{He-d}(E_{He}) = 3.75\frac{S(E_{He})}{E_{He}}\exp\left(-\sqrt{\frac{E_G}{E_{He}}}\right), \quad (7)$$

where E_{He} is the laboratory energy of ^{3}He in keV and E_G is the Gamow energy ($\sqrt{E_G} = 108.8$ keV$^{1/2}$). The S-factor, $S(E_{He})$ is calculated by

$$S(E_{He}) = \left(S_0 + S_1 E_{He} + S_2 E_{He}^2 + S_3 E_{He}^3 + S_4 E_{He}^4\right) \exp\left(-\beta\sqrt{0.667E_{He}}\right) + \frac{G}{(0.667E_{He} - E_r)^2 + \Gamma^2}, \quad (8)$$

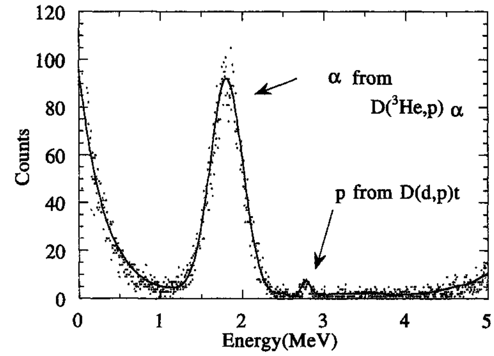

Fig. 3. An example of the energy spectrum of particles from nuclear reactions in $TiD_{1.61}$ irradiated by ^{3}He (50 keV).

where $S_0 = 645.3$, $S_1 = 6.041 \times 10^{-2}$, $S_2 = -8.671 \times 10^{-7}$, $S_3 = 4.899 \times 10^{-12}$, $S_4 = -9.124 \times 10^{-18}$, $G = 1.027 \times 10^9$, $E_r = 361.3$ keV, $\Gamma = 253.6$ keV and $\beta = 6.753 \times 10^{-3}$.

4. Comparison of experimental results with numerical results

Fig. 3 shows a typical energy spectrum of charged particles in the case of ^{3}He irradiation. The two peaks of α and p are clearly separated due to different energy losses in the aluminum foil placed in front of the detector. The experimental conditions are summarized in Table 1. The target of $TiD_{1.61}$ was irradiated by an ion beam with different energies. The nuclear reaction yields by ^{3}He ions with an energy of 100 keV were repeatedly measured in the series of the experiments. There is no significant change in the nuclear yields measured under the same conditions as above. This means that the deuterium density in the target is not charged by ion beam irradiation.

Fig. 4 shows nuclear reaction yields as a function of incident ion energy. Open circles and triangles in Fig. 4 are experimental data with an ^{4}He and ^{3}He beam respectively. Solid lines show numerical results of the Monte Carlo code simulations. Experimental results are in good agreement with the simulation results.

Table 1
Experimental conditions

Ion beam		
Species	^{3}He	^{4}He
Energy (keV)	50–200	50–200
Current (μA)	0.5–20	0.5–20
Target		
Material	$TiD_{1.61}$	$TiD_{1.61}$
Detection particles		
Detector	SSD	SSD
Detected particles	α, p	p
Reaction type	D(^{3}He, p)α, D(d, p)t	D(d, p)t

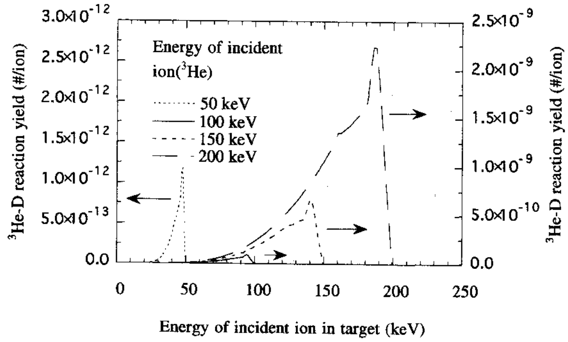

Fig. 5. ^{3}He–D reaction yield dependence upon recoiled energy of incident ion in the target.

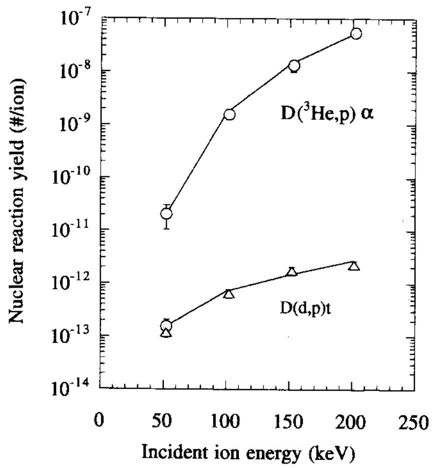

Fig. 4. Experimental data and theoretical predictions of nuclear reaction yields as a function of the energy of incident ion for target of $TiD_{1.61}$. O: Experiment with ^{4}He beam. △: Experiment with ^{3}He beam. ———: Theory.

It is important to understand in which range of energy the nuclear reactions mainly occur. Knock-on atom processes which include PKA and collision cascade atoms were analyzed in detail by Monte Carlo code Figs. 5 and 6 show energy spectra of nuclear reaction yields calculated by Eqs. (3) and (4). In the case of the direct nuclear reaction of D(^{3}He, p)α, the nuclear reactions occur at near the incident beam energy since the nuclear reaction cross-section rapidly decreases with decrease of energy. As seen in Fig. 6, the nuclear reactions of D(d, p)t mainly occur in the energy region of the recoiled D-atom from 10 keV to 100 keV. This means that the main source of the nuclear reaction yield is the high energy collision of PKA d-atoms with stationary d-atoms. Particles detected in the present experiments are mainly produced by nuclear reaction of the PKA d-atom.

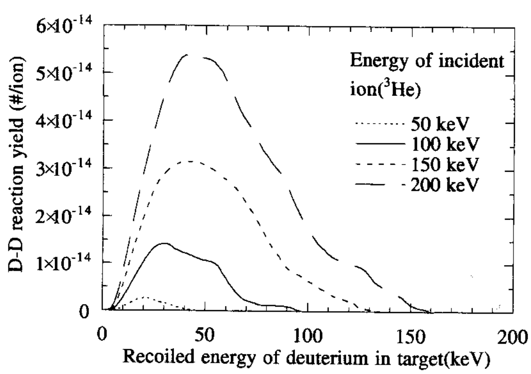

Fig. 6. D–D reaction yield as a function of energy of recoiled deuterium caused by incident ions (^{3}He).

5. Conclusions

The target of single phase $TiD_{1.61}$ was irradiated by an ion beam of ^{4}He and ^{3}He with energies of 50, 100, 150 and 200 keV. We tried to observe directly the high energy knock-on process by irradiation of the ions by means of detection of the nuclear reaction of D(d, p)t. In the experiments with a ^{3}He ion beam, the deuterium density in the target was monitored by detection of the nuclear reaction of D(^{3}He, p)α. Experimental results were compared with the results calculated by the Monte Carlo code, TRIM. The following conclusions have been obtained:

(1) The high energy primary knock-on process has been directly observed by monitoring of the nuclear reaction of D(d, p)t.

(2) It was confirmed that the Monte Carlo code can successfully predict the production of energetic primary knock-on atoms under irradiation of the ion beam.

Acknowledgements

The authors are particularly indebted to Dr T. Yamamoto and Dr T. Shibayama of Tohoku University for fruitful discussions.

References

[1] S. Ishino, N. Sekimura, K. Hirooka, T. Muroga, J. Nucl. Mater. 141 (1986) 776.
[2] J. Gittus, Irradiation Effects in Crystalline Solids (Applied Science, London, 1978).
[3] T. Laursen et al., J. Nucl. Mater. 165 (1989) 156.
[4] J.F. Ziegler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solid (Pergamon New York, 1985).
[5] G.S. Chulick, Y.E. Kim, R.A. Rice, Nucl. Phys. A551 (1993) 255.