Nuclear Instruments and Methods in Physics Research B 315 (2013) 142–145

Contents lists available at SciVerse ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Surface effect on ion track formation in amorphous Si_3N_4 films

Y. Morita [a], K. Nakajima [a], M. Suzuki [a], K. Narumi [b], Y. Saitoh [b], N. Ishikawa [c], K. Hojou [d], M. Tsujimoto [e], S. Isoda [e], K. Kimura [a,*]

[a] Department of Micro Engineering, Kyoto University, Kyoto 615-8540, Japan
[b] Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency, 1233 Watanuki-machi, Takasaki, Gumma 370-1292, Japan
[c] Nuclear Science and Engineering Directorate, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki 319-1195, Japan
[d] Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Naka, Ibaraki 319-1195, Japan
[e] Institute for Integrated Cell-Material Sciences, Kyoto University, Kyoto 615-8540, Japan

ARTICLE INFO

Article history:
Received 30 November 2012
Received in revised form 17 March 2013
Accepted 24 March 2013
Available online 6 April 2013

Keywords:
Ion track
C_{60}
Si_3N_4

ABSTRACT

Thin films of amorphous Si_3N_4 (thickness 5–30 nm) were irradiated with 360–720 keV C_{60}^{2+} ions. Ion tracks were observed using transmission electron microscopy (TEM) and high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM). The track length and the radial density profile of the track were measured for various combinations of the film thickness and the energy of C_{60}^{2+}. The length of the ion track produced in a 30-nm film was found shorter than that in a 20-nm film indicating that there is surface effect on track formation. This can be qualitatively understood in terms of the energy dissipation process. The observed radial density profile also depends on the film thickness. The result can be explained by surface cratering.

1. Introduction

When an energetic ion passes through a material an ion track may be produced along the ion path if the electronic stopping power S_e is larger than a material dependent threshold value [1,2]. In case of crystalline materials, the ion tracks can be easily observed by transmission electron microscopy (TEM). The track interior is amorphized or comprised of defect clusters depending on the material. In case of amorphous materials, direct TEM observation of ion tracks is generally difficult due to a lack of sufficient contrast. The ion tracks produced in amorphous materials were studied using mainly indirect measurement methods, such as Fourier transform infrared spectroscopy (FTIR) [3,4] and etching [4,5]. There were only a small number of studies on the ion tracks produced in amorphous materials by direct TEM observation. Dunlop and coworkers demonstrated that ion tracks produced by MeV C_{60} ions and GeV heavy ions in metallic glasses can be observed using TEM [6,7]. The ion track appears as a bright contrast in TEM images, which means reduced thickness and/or reduced density in the ion track. They employed a topographical contrast imaging technique and found that crater-like structures are formed on the irradiated surfaces. Recently, a fine structure of ion tracks in amorphous SiO_2 (a-SiO_2) was observed by small angle X-ray scattering (SAXS) [8,9]. Analyzing the observed SAXS spectra it was found that the ion track consists of a low density cylindrical core surrounded by a high density shell showing a qualitative agreement with the results of molecular dynamics (MD) simulations [9,10].

Very recently, we have demonstrated that ion tracks in amorphous Si_3N_4 (a-Si_3N_4) thin films (20 nm) produced by sub-MeV C_{60} ion impact can be clearly observed using TEM and high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) [11]. Because the contrast of the HAADF-STEM image is proportional to the integrated density along the electron beam, the density profile of the ion track can be derived from the HAADF-STEM image. We found that the core of the ion track is amorphous and the density is reduced by ~20% at the track center. The core is surrounded by a shell whose density is slightly enhanced. However, possible surface effects on the track formation, e.g. cratering, could not be excluded. In this paper, we extend our study and address the surface effect on the track formation. For this purpose, we irradiate a-Si_3N_4 films of various thicknesses ranging from 5 to 30 nm with 360–720 keV C_{60}^{2+} ions. We found that the length of the track produced in a thicker film is shorter than that in a thinner film. We also found that the density reduction in the track is pronounced for the thinner film.

2. Experimental

Irradiation of sub-MeV C_{60} ions was performed using 400-kV ion implanter at JAEA/Takasaki. Self-supporting amorphous Si_3N_4 (a-Si_3N_4) films of thickness T ranging from 5 to 30 nm were

* Corresponding author.
E-mail address: kimura@kues.kyoto-u.ac.jp (K. Kimura).

0168-583X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.03.043

irradiated with 360, 540, 720 keV C_{60}^{2+} ions to fluences 1–2 × 10^{11} ions/cm^2. After the ion irradiation, TEM and HAADF-STEM observations were performed using a JEOL JEM-2200FS equipped with a field emission gun operating at 200 kV. The samples were held at the specimen tilting holder with the tilt angle from −30 to 30°. The images were taken by GATAN Ultrascan 1000 CCD camera with a 2 × 2 k pixel. In HAADF-STEM mode, a narrow electron beam converged to 0.5 nm diameter and an annular dark detector covering over 120 mrad were used.z

3. Results and discussion

Fig. 1(a) shows an example of the observed plan-view TEM images of the a-Si_3N_4 film (20 nm) irradiated with 720 keV C_{60}^{2+} ions. There are circular structures of almost uniform diameter of ~4 nm. Each structure has a bright core which is surrounded by a dark shell. The number of these structures agrees with the fluence of the C_{60}^{2+} ions, indicating that single C_{60}^{2+} impacts create individual circular structures. In order to estimate the length L of the ion tracks, the sample was observed at a tilt angle of 25°. The observed structures are elongated along the tilt direction as is shown in Fig. 1(b). The length of each track was measured and the obtained length distribution is shown in Fig. 2 together with the results for 360 and 540 keV C_{60}^{2+} on a-Si_3N_4 films of 20 nm. The average track lengths for these ions were derived from the obtained distributions and shown in Table 1. For comparison, the projected ranges of the C_{60} ions in a-Si_3N_4 were estimated using the SRIM2011 program package [12] and shown in Table 1. In this estimation, the so-called cluster effect was neglected and the density of 3.44 g/cm^3 was employed [13].

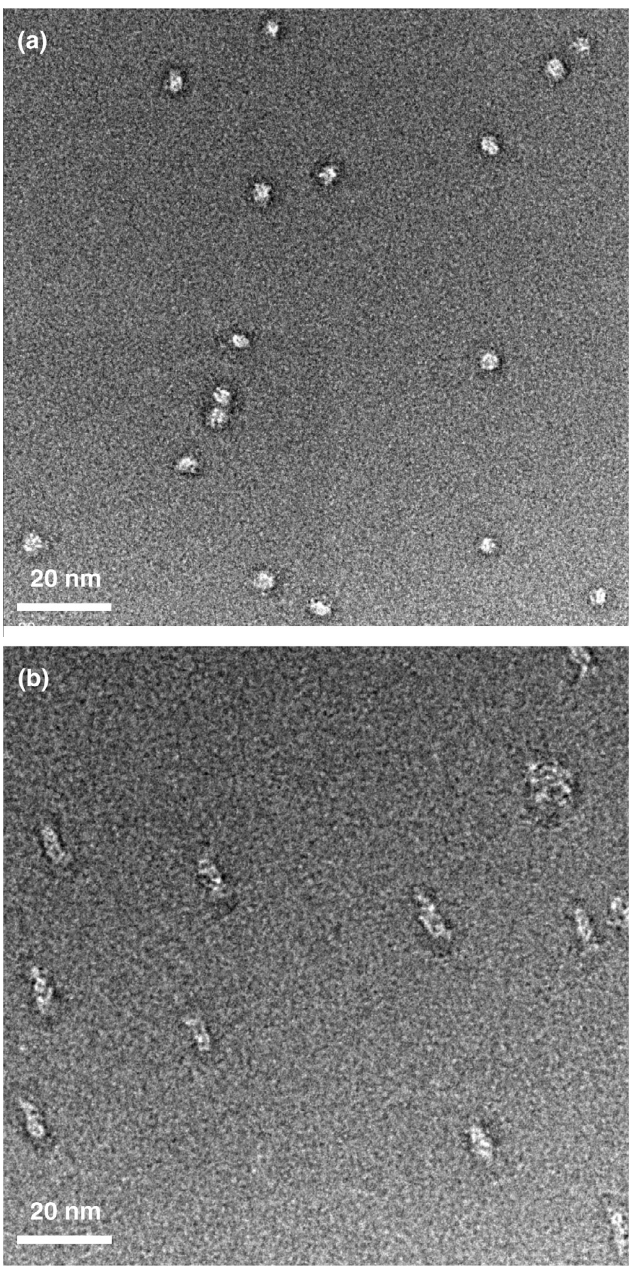

Fig. 1. TEM bright field images of a-Si_3N_4 film (20 nm) irradiated with 720 keV C_{60}^{2+} ions to a fluence of ~1 × 10^{11} ions/cm^2. The TEM images taken at a tilt angle of (a) 0° and (b) 25° are shown.

The observed track length is slightly shorter than the ion range. For example, at 360 keV the track length is shorter than the ion range by 2.4 nm. Considering that the range of the 42-keV C_{60} ion is 2.4 nm, the threshold stopping power for track formation is equal to the stopping power for 42-keV C_{60} ion. The electronic stopping power for the 42-keV C_{60} is calculated to be 2.3 keV/nm using TRIM2011, which is much smaller than the typical threshold value for track formation while the nuclear stopping power for 42-keV C_{60} ion is as large as 10.7 keV/nm. This is in accordance with the recent finding that the nuclear stopping power plays an important role in track formation [14]. The difference between the observed track length and the calculated ion range becomes larger with increasing energy (see Table 1). This is because the constituent carbon ions separate from each other along the ion path due to the mutual Coulomb repulsion (fragmentation) and multiple scattering. As a result, the volume density of the deposited energy decreases along the ion path even if the stopping power is the same. Thus the threshold stopping power is slightly smaller than the above estimated value, 2.3 + 10.7 keV/nm, if there is no fragmentation and multiple scattering.

Fig. 2 also shows the result for 720 keV C_{60}^{2+} impact on 30-nm a-Si_3N_4 film by dashed curve. Surprisingly, the mean length of the ion tracks in the 30-nm film is shorter than that in the 20-nm film. This can be understood in terms of the energy dissipation process. When the ion penetrates throughout the film the deposited energy dissipates mainly in the radial direction as is schematically shown in the inset (a) of Fig. 2. If the film thickness is larger than the projected range, there is an additional pathway for the energy dissipation especially around the end of the ion path (see the inset (b)). Consequently, the temperature of this part may not be high enough for the formation of ion track.

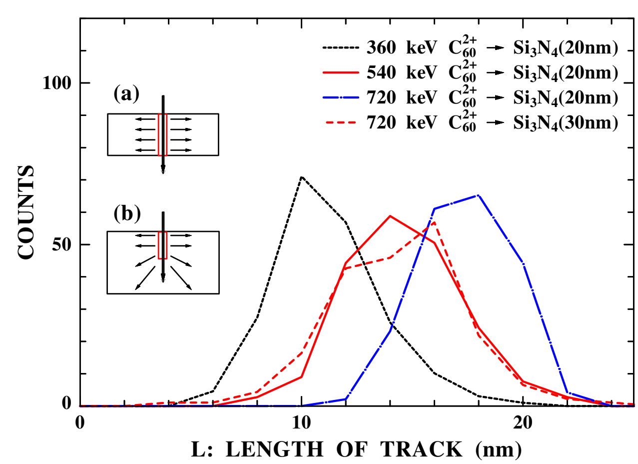

Fig. 2. The length of the ion tracks produced by 360–720 keV C_{60}^{2+} ions in 20-nm and 30-nm a-Si_3N_4 films.