Micro-track structure analysis for 100 MeV Si ions in CR-39 by using atomic force microscopy*

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To analyze the micro-track structure of heavy ions in a polymer material, parameters including bulk etch rate, track etch rate, etch rate ratio, and track core size were measured. The pieces of CR-39 were exposed to 100 MeV Si ions with normal incidence and were etched in 6.25N NaOH solution at 70 °C. Bulk etch rate was read out by a profilemeter after several hours of etching. The other parameters were obtained by using an atomic force microscope (AFM) after a short time of etching. We have measured the second etch pits and minute etch pits to obtain the track growth curve and three dimension track structures to track the core size and etch rate measurements. The local dose of the track core was calculated by the δ-ray theory. In our study, we figure out that the bulk etch rate $V_{\rm b} = (1.58 \pm 0.022)$ μm/h, the track etch rate $V_{\rm t} = (2.90 \pm 0.529)$ μm/h, the etch rate ratio $V = 1.84 \pm 0.031$, and the track core radii $r \approx 4.65$ nm. In the meantime, we find that the micro-track development violates the traditional track-growth model. For this reason, a scenario is carried out to provide an explanation.

Keywords: micro-track structure, bulk etch rate, track etch rate, track core size

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1. Introduction

CR-39 has been considered as the most sensitive solid state nuclear track detector (SSNTD) since its discovery in 1978[1] and it is widely used in charged particle detection.[2] Latent tracks can be produced by charged particles irradiating and tracks can be formed by chemical etching. Formed track properties are mainly determined by etching condition and etching time.[3] Afterwards, the tracks can be visualized by using an optical microscope (OPT), an atomic force microscope (AFM),[4] a laser scanning confocal microscope (LSCM),[5] or a scanning electron microscope (SEM).[6]

Using an AFM is a good method to get the images of tracks,[7,8] and is also used for quantitative analysis of the track etch pits, however, the results are limited, which include measurements of track core size[9] and bulk etch rate,[10] development of short distant scale track,[11] analysis of detector sensitivity,[12] and estimation of latent track.[13] Of all the work listed above, one can see that the AFM method is strongly recommended in the case of nanometer-scale track measurement, hereinafter referred to as "micro-track" in this paper.

Until now, there are still unsolved aspects in track growth mechanism, especially in micro-track growth. The traditional models of track development are based on the assumption of wave propagation being analogous, and the corresponding track size is always in the scale of millimeters, while the AFM method can get precise three dimensional micro-track images in the scale of nanometers, so we can figure out in this work if the traditional models can be adapted to micro-track structure analysis.

To solve the problem, we focus on the AFM technique to get the track structure of a 100 MeV Si ion in nanometer scale and the experiment focused on measuring parameters including bulk etch rate, track etch rate, track core size, radial dose distribution, and etch rate ratio.

2. Experiment and methodology

2.1. Materials and experiments

CR-39 nuclear track detectors from TASTRAK (Track Analysis Systems Ltd, UK) with the size of 2×3 cm^2 and thickness of 1 mm were used in our study. The irradiation experiments were performed at the Heavy-Ion Microbeam Facility[14] of the China Institute of Atomic Energy. The samples, exposed to 100 MeV Si ions with a fluence of $\sim 10^8$ cm^{-2}, were set in a vacuum chamber and the microbeam was at normal incidence to the interface as shown in Fig. 1.

Chemical etching was carried out after irradiation and the etching experiments were conducted at the Centre of Space Physics and Space Environment at the Nanjing University of Aeronautics and Astronautics. The samples were etched in NaOH solution at a concentration of 6.25N at 70 °C and stirred continuously.

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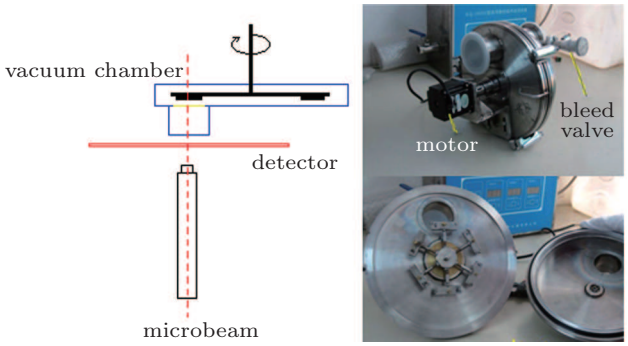


Fig. 1. (color online) Diagram of the microbeam line and vacuum chamber.

2.2. Bulk etch rate (V_b) measurement

Bulk etch rate, V_b, is the etching rate at the undamaged region. In our study, bulk etch rate measurement was achieved by using a thickness-change method in which V_b is given by: $V_b = B/t$, where B is the amount of etched thickness and t is the etching time. Some work has been reported that V_b was smaller in very surface layer (< 0.5 μm) and kept constant in deeper layer (0.5–9 μm).[15,16] Accordingly, we assume that V_b was constant in the layer with thickness larger than 1 μm. Following the previous work,[10,15] a partial masking technique was employed in this measurement. A profilometer was used instead of an AFM in this research. The main procedure used in this method can be summarized in the following steps: (i) the un-irradiated CR-39 surface was partially covered by resin; (ii) the sheets were etched under the condition described in section 1.1 for 1–8 hours; (iii) the masking was peeled off from the sample's surface; (iv) the etched thickness B was recorded by the profilemeter through line-by-line scanning.

2.3. Second etching-track core size measurement

As mentioned earlier, the AFM method has been successfully applied in track core size measurements in a situation where the track core has a nanometer scale size. Considering AFM scanning area, it is obvious that samples with high flux irradiation should be examined by this technique. According to the method described by Yamauchi *et al.*,[9] we can obtain the Si ion track core size by extrapolating the etching pit radius growth curves toward the origin of the etching time. In this irradiation experiment, the track core size of the Si ion is predicted in nanometer scale. It is better to limit the etching time to within one minute, which is why we called it second etching, so as to limit the track size within the nanometer scale.

2.4. Minute etching-track profile analysis

This part of study lies in track etch rate, V_t, derivation. V_t is the etched rate along the particle trajectory, and it can be simply derived from

$$V_t = H/t + V_b,$$

where H is the trajectory depth. Given the value of V_b, H is the only variable in this formula. V_t is the energy loss (dE/dx) dependent.[17] However, in the case of nanometer scale, dE/dx is almost invariable. As a result, V_t is considered to be unchanged in this short trajectory range. As we know, the AFM technique is applicable to short time etching and minute etching is adequate to provide a precise image of a three-dimensional track structure, from which H can be read out.

According to the sections above, the etch rate ratio, V, defined by ratio $V = V_t/V_b$, can be obtained. V is the ratio governing the track development in the traditional model.

3. Results and discussion

3.1. Bulk etch rate

Figure 2 shows the amount of bulk etching, B, as a function of time, t, obtained by a contact profilemeter. The linear fit function of data points is $B = V_b \times t$, where $V_b = (1.58 \pm 0.022)$ μm/h, at the etching condition of 6.25N NaOH solution, 70 °C. Fromm *et al.*[18] studied the relationship between V_b and the concentration and temperature of NaOH solution. They found that the bulk etch rate can be described by

$$V_b(C,T) = 1.276 \times 10^{(0.828C + 0.049T - 0.002CT - 17.624)},$$

where C is the concentration, and T is the temperature. According to this relationship, we can figure out that $V_b(6.25\text{N}, 70\ ^\circ\text{C}) = 1.520$ μm/h from this equation. Obviously, our experiment result is basically consistent with their research.

Figure 2 shows that the amount of bulk etch is more than 1 μm after 1 hour's etching and nearly 9 μm after 6 hours' etching, which means that it is beyond the reach of an AFM's cantilever tip. In this case, a profilemeter, which can measure the vertical height range from nanometers to millimeters and the scanning area can reach up to several square millimeters, is chosen to measure B. It is advantageous to use a profilemeter over an AFM for long time etching samples.

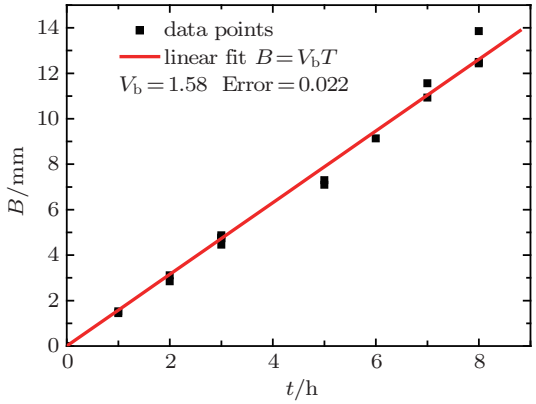


Fig. 2. (color online) Amount of bulk etch, B, as a function of etching time, t.

3.2. Track core size, energy loss, and local dose

3.2.1. Track core size and energy loss

The etch pit radius, r, for 100 MeV Si ion, as a function of time, t, have been shown in Fig. 3. As discussed by Yamauchi *et al.*,[9] the fitted line of data points will not cross the origin and the coordinates of the intersection value corresponds to the track core radii. For 100 MeV Si ion in our experiment, we obtained $r \approx 4.65$ nm from the fitted line shown in Fig. 3. According to previous studies,[9,19,20] track core radii size is energy loss ($\mathrm{d}E/\mathrm{d}x$) dependent and is proportional to $(\mathrm{d}E/\mathrm{d}x)^n$ $(0 < n < 1)$. For example, Tombrello[20] found that track diameter for heavy ions is proportional to $(\mathrm{d}E/\mathrm{d}x)^{1/3}$; Apel *et al.*[19] concluded $n \approx 0.55$ in their research and Yamauchi *et al.*[9] gave a result of $n \approx 0.38$. In our research, energy loss is calculated by using the Geant4 toolkit and it is about 1841 keV/μm for 100 MeV Si ions. The track core radius and energy loss for a 100 MeV Si ion are listed in Table 1.

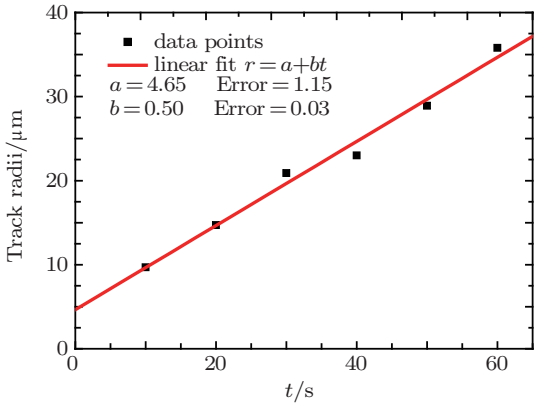


Fig. 3. (color online) The etch pit radius for a 100 MeV Si ion at different etching times.

Table 1. Track core radius, energy loss, and local dose for a 100 MeV Si ion.

Ion beam	Energy/MeV	Energy loss/keV·μm^{-1}	Track radius/nm	Local dose/kGy
Si	100	1841	4.65 ± 1.15	129 (100–180)

3.3. Local dose around the track core

When a heavy ion transports through target materials, the injection particle loses most of its energy by ejecting electrons (δ rays) outside the target atom. These electrons will not deposit the energy in the spot where they are born. As a result, the deposition energy has a spatial distribution which is called radial dose distribution. In this paper, radial dose distribution around the center of the core is computed based on the δ-ray theory of track structure,[21] which included the track model of Katz and Kobetich.[22] In this theory, the radial energy distribution can be written as

$$E(R) = \frac{(-1)}{2\pi R} \sum_i \int_{\omega_t}^{\omega_m - I_i} \mathrm{d}\omega \frac{\partial}{\partial x} W(R,\omega)\eta(R,\omega)\frac{\mathrm{d}n_i}{\mathrm{d}\omega},$$

where $E(R)$ is identified as the radial dose distribution, R is the cylindrical surface radius, $W(R,\omega)$ is the residual energy, η is electron transmission probability, $\mathrm{d}n/\mathrm{d}\omega$ is the number of free electrons by an ion per unit length of ion path with energies between ω and $\omega + \mathrm{d}\omega$, ω_t is the lower limit, I is mean excitation energy, and i is the i-th element in the compound. Radial dose distribution, calculated by equation above, is shown in Fig. 4. The figure indicates that most of the energy transferred from the injected particle is deposited around the track core and is deposited in the scale of nanometers from the track center. Accordingly, the local dose for a 100 MeV Si ion deposited in the track core area is up to about 129 kGy.

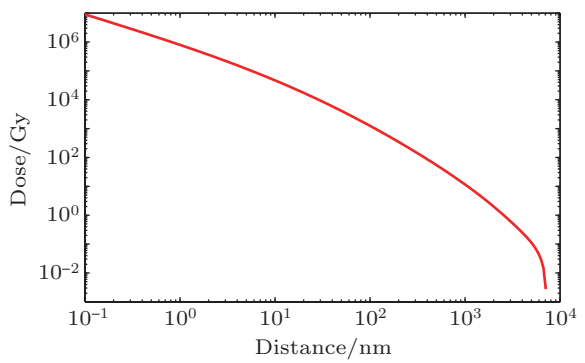


Fig. 4. (color online) Radial dose distribution in CR-39 for 100 MeV Si ion.

3.4. Track etch rate and etch rate ratio

In this section, the AFM image of an ion track for a 100 MeV Si ion after a minute's etching is studied in order to calculate the track etch rate. Figure 5 shows the AFM image of CR-39 after 5 minutes' etching by using the AFM analysis software Nanonavi. It is obvious that the etched track is a cone in three dimensions as indicated in Fig. 5(a) and the track diameter D and depth H can be read out by using a line profile toolkit, as shown in Fig. 5(b). For each ion track, at least 10 data points of the track diameter and depth are obtained which are used to determine the mean track radii and depth. All the mean values for each track measured are fitted by the least square method. As a result, we obtain $D = (307.23 \pm 6.237)$ nm, and $H = (110.36 \pm 2.575)$ nm. Accordingly, $V_t = H/t + V_b = (2.90 \pm 0.529)$ μm/h and $V = V_t/V_b = 1.84 \pm 0.031$.

The traditional track development model of normal incidence with respect to the irradiated sheets' surface and constant V_t indicate that the relationship between track diameter,

D, and bulk etched thickness, B, is given by

$$D = 2B\sqrt{\frac{V-1}{V+1}}.$$

It is apparent that $D < 2B$ and $V > 1$ in this model. In our experiment, the bulk etched thickness, $B = V_b \times t = (131.67 \pm 1.833)$ nm while the track diameter, $D = (307.23 \pm 6.237)$ nm after 5 minutes etching. So the relationship: $D > 2B$ is found in the case of a micro-track condition. This situation violates the traditional model claimed above. However, it is coincident with the results from Yasuda *et al.*,[13] which they gave explanations for and referred to the etching progress in the track core area at the samples' surface. In our view, we suggest a scenario for the micro-track development in the early stage of etching: etch rate is dose dependent; as a result, it changes with different track radii, but the etch rate at a particular track core sizes is considered to be a constant which is V_t. The etch rate between the track core and the undamaged area is not considered to be equal to V_t or V_b. But obviously, it is less than V_t and larger than V_b. At the very beginning of etching, the local dose is of special importance in micro-track development. Apparently, it violates the nature of the traditional model.

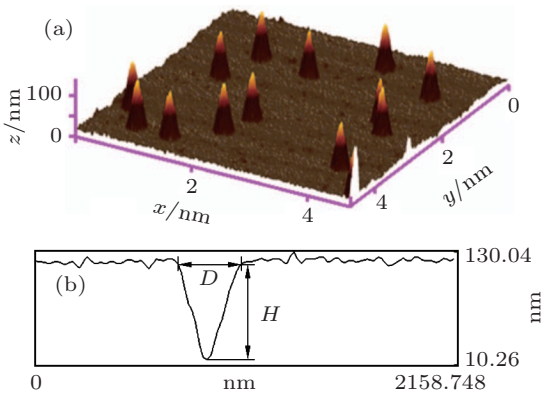


Fig. 5. (color online) The AFM image of the sample after 5 minutes' etching. (a) Three-dimensional image; (b) cross-section profile.

As described above, from the aspect of a micro-track structure, the growth of an etched track is different from the nature of the traditional normal incidence model. A new track development theory for micro-track structures needs to be worked out.

4. Conclusions

Bulk etch rate, track etch rate, etch rate ratio, and track core are considered as key factors in ion track growth. In this paper, we used alternative methods and equipment to study these parameters for a 100 MeV Si ion. The bulk etch rates were measured by using a profilemeter. Partial masking was recognized as an effective and simple method here to obtain the V_b. This method was different from AFM measurements in aspects of scanning area and etching time.

Track core size was obtained after a second etching by using an AFM. The data points of pit radius were fitted with a function of etching time and the track radii was found equal to 4.65 nm for Si particles with energy of 100 MeV. After that, the radial dose distribution was calculated according to the δ-ray theory model. And local dose deposition in the track core of a 100 MeV Si ion was about 129 kGy. In this experiment, data points are tedious to read out. An AFM was also used for tracking the etch rate and we found in this section that the track radius was larger than bulk etch thickness, which violated the geometric nature of track growth. We suggested that the track development at the beginning of etching was different from traditional track growth model.

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