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Measurement of bulk etch rate of LR115 detector with atomic force microscopy

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Abstract

Equations for calculating track parameters have been proposed, which invariably involve the track etch rate V_t and the bulk etch rate V_b. The present study measured V_b for the LR115 solid-state nuclear track detector using atomic force microscopy (AFM). The detectors were partially masked using rubber cement and then etched in 2.5 N NaOH solution at 60°C for time periods ranging from 5 to 40 min. The rubber cement was then peeled off and cross-sectional images of the LR115 detectors were obtained by AFM. V_b has been found to have different values below and beyond the etching time of about 13.5 min, with the values of 0.0555 and 0.0875 μm min^{-1}, respectively. The increase in V_b with the etching time can be explained by a diffusion-etch model, in which the additional damage of the detector material is due to those etchant ions diffused into the detector over time. Now that V_b has been determined, this can be combined with the track etch rate V_t to calculate track parameters.

Keywords: Atomic force microscopy (AFM); Solid-state nuclear track detector; LR115; Bulk etch rate

1. Introduction

The problem of track development in solid-state nuclear track detectors (SSNTDs) has attracted much attention for a long time (e.g., Henke and Benton, 1971; Paretzke et al., 1973; Somogyi and Szalay, 1973; Somogyi, 1980; Fromm et al., 1988; Hatzialekou et al., 1988; Ditlov, 1995; Meyer et al., 1995; Nikezic and Kostic, 1997). A method for the calculating track parameters based on analytical and three-dimensional consideration was presented by Nikezic (2000).

The equation of the track wall in the two-dimensional case is given by Nikezic and Kostic (1997) as

$$y = \int_z^L \frac{d\xi}{\sqrt{V^2(\xi) - 1}}, \tag{1}$$

where (z, y) are coordinates of points on the track wall, z is an axis along the particle trajectory, $V(\xi) = V_t(\xi)/V_b$ the ratio of the track etch rate to the bulk etch rate, ξ the residual range and L is the penetration depth of etching along the particle trajectory in the detector material.

The equation of track wall in the conical phase in three dimensions for normal incidence is given as

$$\sqrt{x^2 + y^2} = \int_z^L \frac{d\xi}{\sqrt{V^2(\xi) - 1}}, \tag{2}$$

where the z-axis is again along the particle trajectory, and (x, y) are coordinates of points in the track wall (Nikezic, 2000). The equation for the contour line of the track opening for an oblique angle is given as

$$\sqrt{x^2 + (y\sin\theta)^2} = \int_{y\cos\theta + h/\sin\theta}^L \frac{d\xi}{\sqrt{V^2(\xi) - 1}}, \tag{3}$$

where θ is the incident angle (with respect to the surface of a detector), (x, y) are coordinates of points on the track opening contour and h is the thickness of the removed layer. Note that the coordinates (x, y) in Eqs. (2) and (3) have different

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senses. If the function $V(\xi)$ is constant, the track opening will be an ellipse and the main track parameters, namely the major axis D and the minor axis d, can be found in explicit form. In more realistic cases, when V is not constant, the track opening is not an ellipse, but is instead egg-like or has an even more complicated shape, which depend on the function $V(\xi)$.

As can be seen above, the track development depends strongly on $V(\xi)$, or in turn on the track etch rate V_t and the bulk etch rate V_b. For accurate calculations of the track parameters, therefore, a good knowledge of V_t and V_b are needed. The present study is devoted to the experimental measurements of V_b for the LR115 SSNTD using atomic force microscopy (AFM). While AFM has been employed to measure the bulk etch rate for another commonly used SSNTD, CR39, by Yasuda et al. (1998) and Vázquez-López et al. (2001), and track developments in CR39 and LR115 by Yasuda et al. (2001) and Palmino et al. (1999), respectively, the bulk etch rate for LR115 has not been measured up to now.

2. Experimental

The LR115 films were purchased from DOSIRAD (Type 2, non-strippable, 12 μm red cellulose nitrate on a 100 μm clear polyester base, Catalog number 500 9535). The detectors used in the present experiments were cut from the films with a size of $1 \times 1\ \text{cm}^2$. Part of the LR115 detector surface was masked with rubber cement (product no.140, Union Rubber Inc., Trenton, NJ 08606-1040). The rubber cement has a very short setting time, viz. less than 10 min. After setting of the rubber cement, the partially masked detectors were etched in 2.5 N NaOH solution at 60°C using a water bath controlled with a thermostat. The etching time was chosen from 5 to 40 min, in steps of 5 min. After etching, the rubber cement was peeled off from the detectors.

The atomic force microscope used in the present study was the Autoprobe CP model from Park Scientific Instruments (1171 Borregas Avenue, Sunnyvale, CA 94089). The atomic force microscope can be used to study the surfaces of non-conducting materials non-destructively under standard conditions, such as the LR115 detectors studied in the present investigation. Microlevers were available with standard or sharpened integrated pyramidal tips with nominal radii of less than 500 and 200 Å, respectively. Each cantilever chip has five V-shaped cantilevers (coded A and C to F) and one rectangular cantilever (coded B). The microlevers were microfabricated from low stress silicon nitride and were highly resilient with a wide range of force constant, from 0.5 to 0.01 N m^{-1}. In our experiment we used the standard tip and the cantilever F which has a length of 85 μm and a typical tip length of 3 μm. Contact mode was employed with the default applied force of 0.5 N m^{-1}. The surfaces of the detectors were imaged directly in air with a resolution of 256×256 pixels. After etching and peeling off the rubber cement, cross-sectional profiles of the LR115 detectors were obtained by AFM from which the amounts of bulk etch B were determined.

3. Results and discussion

In Fig. 1, the observed values of B are shown as a function of the etching time t. For each etching time, at least 4 (and up to 12) cross-sectional profiles are used to determine a mean value of B. From Fig. 1, it seems that the bulk etch rate V_b (the amount of bulk etch B per unit time) increases from small etching time t (and thus small amounts of bulk etch). For $t \geqslant 20$ min, V_b has become a constant. We performed least-squares fits separately to the first three data (i.e., $t < 20$ min) and to the rest of the data (i.e., $t \geqslant 20$ min), and required the first best-fit line to pass through the origin. The best-fit lines were $B = (0.0555 \pm 0.0009) \times t$ and $B = (0.0875 \pm 0.0092) \times t - (0.4314 \pm 0.3060)$ μm, respectively. The intersection between the two best-fit lines corresponded therefore to an etching time of about 13.5 min and a bulk-etched layer of 0.75 μm.

Interestingly, Yamamoto et al. (1997) also noticed that the bulk etch rate for CR39 was slightly smaller in the very surface layer (< 0.5 μm) but kept constant for the deeper layer of 0.5–9 μm. Furthermore, Yasuda et al. (1998) also found the bulk etch rate of the CR39 detector to increase with etching time up to a bulk-etched layer of about 0.5 μm, and the bulk etch rate then became constant at depths over 0.5 μm. The results of Yasuda et al. (1998) also showed that the growth rate of radii of fission tracks increased to a higher value beyond an etching time of about 40 min, which corresponded to a bulk-etched layer of about 1 μm.

If we extrapolate the best-fit line $B = (0.0875 \pm 0.0092) \times t - (0.4314 \pm 0.3060)$ μm back to the x-axis, we obtain the x-intercept as 5 min which is exactly the same as the

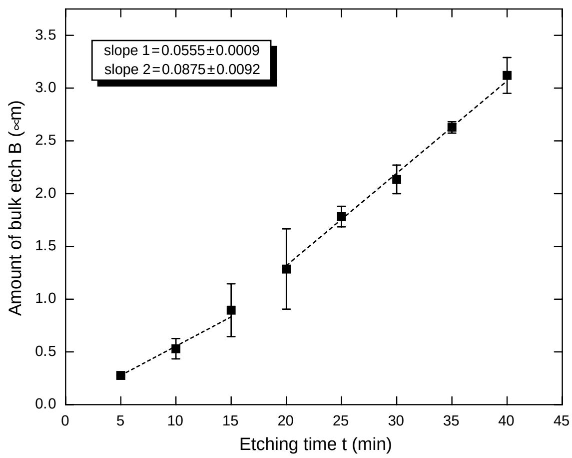

Fig. 1. Amount of bulk etch B as a function of the etching time t.

x-intercept obtained by Yamamoto et al. (1997) for the CR39 detector. Yamamoto et al. (1997) linked this to the "etch induction time" proposed by Ruddy et al. (1977). This etch induction time was in fact related to tracks, which was the time required for the particle tracks to appear in plastic detectors after the start of etching, and is not related to the bulk etch.

Yamamoto et al. (1997) proposed the appearance of the bulk etch induction time in their data was an artifact due to combination of their AFM data and data from their optical microscope. However, the results of Yasuda et al. (1998) and those of the present work are both confined to AFM only, so the changes in the bulk etch rate are likely to be real, and may indeed be a general phenomenon in all track detectors.

The increase in V_b with the etching time can be explained by a diffusion-etch model proposed by Törber et al. (1982). Törber et al. (1982) depicted a two-phase development of latent tracks in which ordinary etching of the detector is accompanied by a simultaneous diffusion of the etchant ions into the detector. Despite the focus on the tracks only, Törber et al. (1982) nevertheless commented that the damaging processes in the bulk material and in the latent track region are the same in principal, differing merely in the coefficients of equations. In this way, the increase in V_b with the etching time can be explained by the additional damage of the detector material due to those etchant ions diffused into the detector over time.

4. Conclusions

Atomic force microscopy has been employed to determine the bulk etch rate V_b for the solid-state nuclear track detector LR115. It has been found that V_b has different values below and beyond the etching time of about 13.5 min, with the values of 0.0555 and 0.0875 $\mu m\ min^{-1}$, respectively. The etching time of 13.5 min corresponded to a bulk-etched layer of 0.75 μm. Such a change in the bulk etch rate was also observed for the CR39 detector by Yamamoto et al. (1997) and Yasuda et al. (1998), also around a bulk-etched layer of 0.5–1 μm. The increase in V_b with the etching time can be explained by a diffusion-etch model, in which the additional damage of the detector material is due to those etchant ions diffused into the detector over time. Now that V_b has been determined, this can be combined with the track etch rate V_t to give $V(\xi)$, which is a required parameter in Eqs. (1) and (2) for calculating track parameters (Nikezic and Kostic, 1997; Nikezic, 2000).

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