Effects of stirring on the bulk etch rate of LR 115 detector

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Abstract

The effects of stirring on the bulk etch rate of LR 115 detector has been investigated. The surface profile measurement method using an instrument called Form Talysurf has been used to measure the thickness of the active layer of the LR 115 detectors. The etchant was 10% aqueous solution of NaOH maintained at $60°C$. The bulk etch rate under magnetic stirring has been found to be 6.65 ± 0.34 μm h^{-1} and that under no stirring to be 3.61 ± 0.14 μm h^{-1}. The initial thickness of the active layer before etching also varies.

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1. Introduction

There are various methods to measure concentrations of ^{222}Rn, among which the solid state nuclear track detector (SSNTD) is commonly used. Alpha particles emitted by ^{222}Rn and its progeny hit the detector and leave latent tracks in it. The tracks are made visible by chemical or electrochemical etching.

The problem of track development 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). In general the track development will depend on $V = V_t/V_b$ or the ratio of the track etch rate to the bulk etch rate. Therefore a precise control of the bulk etch rate, among others, is crucial for a correct measurements of the ^{222}Rn concentrations. For most studies using SSNTD, the control of etching rate is usually specified by the etching conditions, which involve only the temperature and the concentration of the etchant, and the etching duration. While this sort of surrogate is satisfactory for, e.g., the CR39 detector (Ho et al., 2003), this might not be in general adequate for all SSNTDs.

Enge et al. (1974) hypothesized that a colloid layer of partially dissolved cellulose nitrate molecules together with components of the etching solutions adhered to the surface of the plastic during the etching process. They studied the effects of stirring speed on cellulose nitrate (Daicel, Japan) for NaOH at $50°C$, and found that the bulk etch rate increased with the stirring speed for a constant normality of NaOH, which could be explained by the washing-off of the colloid layer. Gruhn et al. (1974) also studied the mechanism of etching of a cellulose nitrate track detector employing camphor as plasticizer (type RS nitro-cellulose fiber obtained from Hercules Powder Company). They proposed that a layer of camphor built up at the surface as etching proceeded, which was the principal factor governing the nature of the etching.

In the present study, the effects of stirring on the bulk etch rate of LR 115 detector will be investigated. This detector has been employed for radon measurements by many investigators.

2. Materials and methodology

Various methods have been proposed or employed for determination of the bulk etch rate of SSNTD. For example, one relied on the difference between detector mass before and after etching, and another was based on measurements of track opening diameters (Durrani and Bull, 1987). Nevertheless, these were indirect measurements. More recently,

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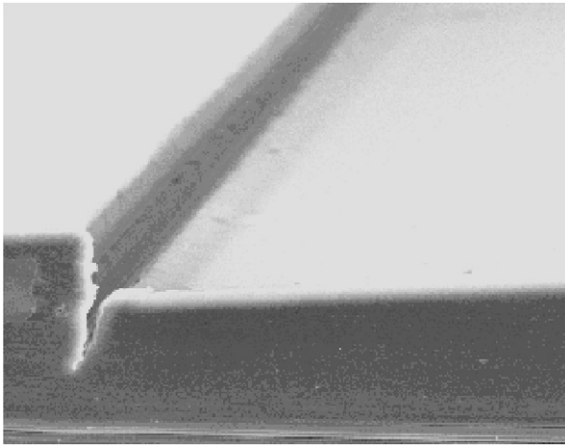
Fig. 1. The cross-sectional view of the LR 115 detector imaged using scanning electron microscope before etching showing the cliff of the active layer (on the left) over the substrate (on the right).

two experimental techniques have been developed to measure the bulk etch rate. The first technique is the atomic force microscopy (AFM). This AFM technique has been employed to determine the bulk etch rate for the CR39 SSNTD by Yasuda et al. (1998) and Vazquez-Lopez et al. (2001) and for LR 115 SSNTD by Ho et al. (2002). The resolution of the AFM is very high (in the order of Å) but the stylus for the AFM is only a few μm, so this technique is most useful for short etching-time studies. The second technique is the surface profile measurement using an instrument called "Form Talysurf" (Nikezic and Janicijevic, 2002; Ho et al., 2003; Yip et al., 2003a, b).

In this paper, the Form Talysurf (Taylor Hobson, Leicester, England) was employed to measure the bulk etch rate for the LR 115 SSNTD. The measuring system is based on a laser interferometric transducer. A computer-controlled stylus passes slowly across a surface of interest during measurements, while the data are processed by the computer to generate an output graph showing the profile of the scanned surface. The accuracy of the instrument is 0.004 μm.

The LR 115 detectors used in the present study were purchased from DOSIRAD, France (LR 115 film, Type 2, non-strippable). The detectors consist of an active layer of red cellulose nitrate on a 100 μm clear polyester base substrate. Two pieces of LR 115 detectors were employed for our experiments, each with a size of about 1.5×1.5 cm^2. Before etching, a portion of the red cellulose nitrate layer was first removed by a razor to expose the colorless polyester base (see SEM image shown in Fig. 1), and the thickness of the active layer was revealed by the profile of this cliff of active layer over the substrate. A mean value for the thickness of the active layer is obtained by taking the average of the measurements for 10 different positions of the cliff.

The detectors were etched separately in 10% aqueous solution of NaOH maintained at a 60°C by a water bath, which is the most frequently used etching condition for LR 115 detectors. The temperature was kept constant with an accuracy of ±1°C. One detector was etched under no stirring while the other was etched using a magnetic stirrer (Model No: SP72220-26, Barnstead/Thermolyne, Iowa, USA). At selected time intervals, i.e., 30, 60 and 90 min for etching under magnetic stirring, and 60 and 120 min for etching under no stirring, the detectors were taken out from the etchant and immediately rinsed by distilled water. The thickness of the residual active layer was again determined using Form Talysurf and revealed by the profile of the active-layer cliff over the substrate. Again, the mean value for the active-layer thickness is obtained by taking the average of the measurements for 10 different positions of the cliff. After measurements, the detectors were etched again for another period of time if necessary.

3. Results and discussions

The relationships between the thickness of residual active layer and the etching time are shown in Fig. 2 for the detectors etched under magnetic stirring and under no stirring. One can see in Fig. 2 that the bulk etch rate under magnetic stirring is much faster than that under no stirring. By fitting the linear relationship $y = A + Bx$ to the experimental data, where y(μm) is the thickness of the active layer and x(min) is
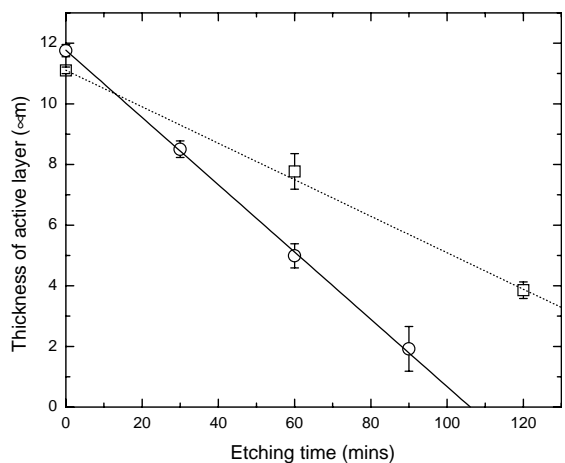

Fig. 2. Relationship between the thickness of the active layer of the LR 115 detector recorded by Form Talysurf and the etching time. Open circles and solid line: data obtained with etching under magnetic stirring and the corresponding best fit line; Open squares and dashed line: data obtained with etching under no stirring and the corresponding best fit line.

the etching duration, we have

$$A = 11.77 \pm 0.19, \quad \text{and} \quad B = -0.1109 \pm 0.0057$$

for etching under magnetic stirring,

$$A = 11.11 \pm 0.11, \quad \text{and} \quad B = -0.0602 \pm 0.0024$$

for etching under no stirring.

In other words, the bulk etch rates are 6.65 ± 0.34 μm h^{-1} and 3.61 ± 0.14 μm h^{-1} for etching under magnetic stirring and under no stirring, respectively. These values, in particular the one for etching under magnetic stirring, are different from the value of 3.27 ± 0.08 μm h^{-1} obtained by Nikezic and Janicijevic (2002) also under the same etchant conditions (10% aqueous NaOH at 60°C). Apparently, stirring has enhanced the bulk etch rate significantly. The enhancement of the bulk etch rate agrees with the model that a colloid layer is formed at the surface of the detector during etching (Enge et al., 1974; Gruhn et al., 1974).

Both our value 3.61 ± 0.14 μm h^{-1} and the value 3.27 ± 0.08 μm h^{-1} obtained by Nikezic and Janicijevic (2002) have been obtained under no stirring. The difference may be due to the different convection present in the etchants. The large enhancement in the bulk etch rate for etching under magnetic stirring also suggests a way to shorten the required etching time, which will be very useful if a large amount of detectors have to be measured within a short time, e.g., during a large-scale survey.

A second observation is that the initial thickness of the active layer might not be exactly 12 μm, as usually taken for granted. Although the initial thickness was determined by Nikezic and Janicijevic (2002) was 12 ± 0.01 μm, the value for the two pieces of detectors used in the present experiments were 11.77 ± 0.19 and 11.11 ± 0.11 μm, using exactly the same method as that by Nikezic and Janicijevic (2002). This implies that the thickness of the active layer of a piece of detector has to be measured each time before etching to facilitate an accurate determination of the thickness of removed layer during etching.

4. Conclusions

The surface profile measurement method using an instrument called Form Talysurf has been used to measure the thickness of the active layer of the LR 115 detectors. The bulk etch rate under magnetic stirring has been found to be 6.65 ± 0.34 μm h^{-1} and that under no stirring to be 3.61 ± 0.14 μm h^{-1} for the etching conditions: 10% aqueous solution of NaOH at 60°C. The initial thickness of the active layer before etching also varies.

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