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A fast method to measure the thickness of removed layer from etching of SSNTD based on EDXRF

C.W.Y. Yip, J.P.Y. Ho, D. Nikezic[1], K.N. Yu*

Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon Tong, Kowloon, Hong Kong

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Abstract

Solid state nuclear track detectors are commonly used for measurements of concentrations of radon gas and/or radon progeny. All these measurements depend critically on the thickness of the removed layer during etching. However, the thickness of removed layer calculated using the etching period does not necessarily provide a sufficiently accurate measure of the thickness. For example, the bulk etch rate depends on the strength of stirring during etching for the LR 115 detector. We propose here to measure the thickness of the removed layer by using energy-dispersive X-ray fluorescence spectrometry. In the present work, a reference silver nitrate pellet is placed beneath the LR 115 detector, and the fluorescence X-ray intensity for silver is then measured. We have found a linear relationship between the X-ray intensity and the thickness of the removed layer for LR 115 detector. This provides a fast method to measure the thickness of removed layer from etching of LR 115 detector. However, this method was found to be inapplicable for the CR-39 detector. Therefore, alternative methods have yet to be explored for the CR-39 detector.
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1. Introduction

It has been established a long time ago that the absorbed radon (^{222}Rn) dose in the human lung is not delivered by radon itself, but instead mostly caused by its short-lived progeny. However, until now, there is no widely accepted methods for long-term passive measurements of radon progeny concentrations, despite that short-term active measurements are relatively easy by air filtering and subsequent activity measurements on the filter. For this reason, exploration of long-term passive integration measurement methodologies regarding short-lived radon progeny has attracted much attention in the field of radon dosimetry.

There are a few different approaches to this problem, all of them being based on the usage of solid state nuclear track detectors (SSNTDs). 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. One of the directions nowadays is to exploit alpha spectroscopy (Mozzo et al., 1996) while another one is to explore particular etching conditions in such a way that only tracks with specified characteristics or in particular energy windows are seen on the detector (Baixeras et al., 1999). All these methods rely on an accurately defined thickness of etched layer of the detector.

For most studies using SSNTD, the control of the etched layer is usually specified by the etching conditions, which involve only the temperature and the concentration of the etchant, and the etching duration. However, stirring has been found to affect the bulk etch rate of LR 115 detector, so that control of the etching conditions is practically unrealizable (Yip et al., 2003), and monitoring of the active-layer thickness is necessary when using the LR 115 detector. This detector has been employed for radon measurements by many investigators (see e.g., Nikolaev and Ilic, 1999).

A convenient and accurate technique has been recently proposed to measure the thickness of the etched layer for the

* Corresponding author. Tel.: +852-2788-7812; fax: +852-2788-7830.

E-mail address: peter.yu@cityu.edu.hk (K.N. Yu).

[1] On leave from University of Kragujevac, Yugoslavia.

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LR 115 detector through the surface profile measurement using an instrument called "Form Talysurf" (Nikezic and Janicijevic, 2002). However, these profile measurements are relatively time-consuming and, most importantly, they are destructive (since the stylus scratches the detector surface during measurements), so the method can only be used after etching has been completed. After the measurements, no more etching can be carried out, so these measurements are referred to as a posteriori measurements.

However, it would be useful to devise methods to measure the thickness of the detectors before the completion of etching (hereafter referred to as a priori measurements of the thickness). These techniques should be fast and non-destructive. After such a measurement, if the thickness of a detector has not reached the desired value, further etching can still be carried out. In this way, the etched layer can be controlled. In the present study, the feasibility of using energy dispersive X-Ray fluorescence (EDXRF) as an a priori technique to determine the thickness of SSNTDs (including CR-39 and LR 115 detectors) will be investigated.

2. Experiment

2.1. Active-layer thickness measurements for LR 115 detectors

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. Seven pieces of LR 115 detectors were employed for our experiments, each with a size of about 1.5×1.5 cm^2.

To establish the correlation between the characteristic fluorescent X-ray intensities from the substrate and the thickness of the active layer of the LR 115 detector, the active-layer thickness should be accurately determined. The procedures for measuring the active-layer thickness followed those devised by Nikezic and Janicijevic (2002). Before etching, a portion of the red cellulose nitrate layer was first removed by a razor to expose the colorless polyester base, and the thickness of the active layer was revealed by the profile of this cliff of active layer over the substrate. The Form Talysurf (Taylor Hobson, Leicester, England) was employed to measure the active-layer thickness 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 mean value and the standard deviation for the thickness of the active layer are obtained through measurements for five different positions of the cliff.

The detectors were then etched separately in 2.5 N aqueous solution of NaOH maintained at a 60°C by a water bath, which is the most frequently used etching condition for LR 115 detector. The temperature was kept constant with an accuracy of ±1°C. The detectors were etched using a magnetic stirrer (Model No: SP72220-26, Barnstead/Thermolyne, Iowa, USA) for more uniform etching (Yip et al., 2003).

At selected time intervals, the detectors were removed 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. As before, the mean value and the standard deviation for the active-layer thickness were obtained through measurements for five different positions of the cliff.

2.2. Etched layer thickness measurements for CR-39 detectors

Seven pieces of CR-39 detectors (Page Mouldings Limited, England), each with a size of about 1.5×1.5 cm^2, were used in our experiments. For each detector, a part of its surface was masked with epoxy (Bostik Epoxy, Australia Pty Ltd, Australia) before being etched in 6 N aqueous solution of NaOH at 70°C maintained by a water bath. At selected time intervals, the detectors were removed from the etchant, rinsed by distilled water, and the epoxy peeled off from the masked surface. The profile of the masked part against the etched part is shown as a cliff under Form Talysurf, and the etched layer thickness of a CR-39 detector was then revealed by the height of the cliff. The mean value and the standard deviation for the etched thickness of the CR-39 detector were obtained through measurements for three positions of the cliff.

2.3. Measurements by using EDXRF

In our experiments, the idea is to detect the EDXRF signal from a chemical element present in a substrate placed beneath the SSNTDs. A chemical element is chosen in such a way that the characteristic fluorescent X-ray energy is high enough to avoid total absorption by the SSNTD but is also low enough to be sufficiently sensitive to the etched layer thickness of the detector. After several trials, we have found that Ag is such an appropriate element for the LR 115 detector.

As mentioned above, the EDXRF signals from Ag will be employed for the present experiments. EDXRF provides a rapid method for the analysis of trace elements in samples.

A silver nitrate pellet is required as a substrate. Each pellet contained 5 g of silver nitrate powder (Chemical formula: $AgNO_3$; Purity: 99.8%; Manufacturer: Panreac Quimica SA; Code: 131459) and 0.25 g of a blender (from Chemplex®, #600) for better binding properties. The materials were homogenized and then formed into stable pellets

using a SpectroPress® Systems press (Model no.: Automatic 50 TON PRESS, Chemplex Industries, Inc., USA) with die diameter of 1.25″ (32 mm) and the following settings: force = 30 ton, dwell time = 3 min and bleed time = 4 min. The pellet was placed beneath the LR 115 detector.

We measured the EDXRF K_β line intensities for the chemical element of Ag (energy = 2.984 keV). All measurements were carried out under vacuum, using an EDAX International DX-95 EDXRF spectrometer with a Mo target, equipped with a liquid nitrogen cooled Si(Li) detector for 300 live seconds. The maximum power of the instrument was 25 W. The incident and take-off angles were 45°, with a Be window thickness of 12.5 μm. The distance between the sample (exposed diameter of 22 mm) and the detector was 4.5 cm. The mean value and the standard deviation for the net Ag K_β line intensities (gross-background) are obtained from two measurements.

3. Results and discussion

3.1. LR 115 detector

The active-layer thickness for LR 115 measured by Form Talysurf and the corresponding net EDXRF K_β line intensity of Ag are comparatively shown in Table 1. The data are also shown in Fig. 1. An anti-correlation between the two variables is apparent. By (least-square) fitting the linear relationship $y = A + Bx$ to the experimental data, where y (μm) is the thickness of the active layer measured using Form Talysurf, and x (count per second) is the net intensity under the Ag K_β EDXRF line, we have $A = (40.7 \pm 0.2)$ and $B = -(0.227 \pm 0.002)$, with $R^2 = 0.9967$. This relationship with a large value of R^2 demonstrates that the active-layer thickness for LR 115 can be satisfactorily reflected by the net Ag K_β line intensity.

Moreover, it can be seen that the linearity is valid within the entire range of the present data set (active-layer thickness from 3.8 to 11.5 μm). For the standard etching conditions that are usually applied to the LR 115 detector, namely, the etchant of the 2.5 N aqueous solution of NaOH kept at 60°C and an etching duration of 2 h, the remaining active-layer thickness is expected to be around 5.5 μm (Nikezic and Janicijevic, 2002), which has been covered by the present data set. Therefore, the present relationship is valid for normal etching conditions. In this way, we have successfully established a fast a priori technique based on EDXRF to determine the thickness of LR 115 SSNTDs.

Table 1
The active-layer thickness for LR 115 measured by Form Talysurf and the corresponding EDXRF K_β line intensity of Ag

Active-layer thickness for LR 115 (μm)	EDXRF K_β line intensity of Ag (counts s^{-1})
11.5 ± 0.1	128.9 ± 0.8
10.1 ± 0.1	134.8 ± 0.9
10.0 ± 0.1	136.7 ± 1.5
8.2 ± 0.2	142.9 ± 0.3
7.0 ± 0.1	148.7 ± 0.6
5.7 ± 0.1	152.5 ± 1.3
3.8 ± 0.0	162.9 ± 0.5

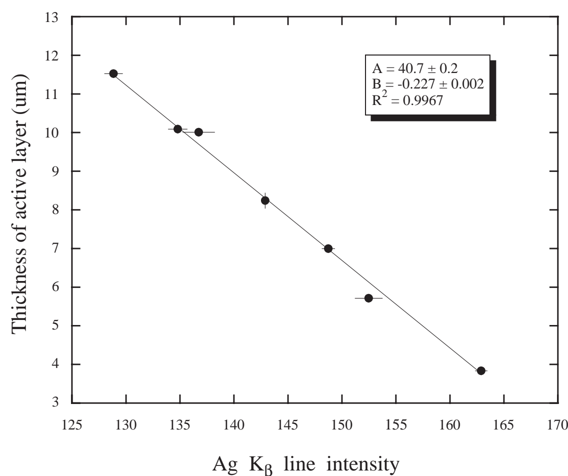

Fig. 1. Relationship between the active-layer thickness for LR 115 measured by Form Talysurf and the corresponding EDXRF K_β line intensity of Ag. Some error bars are smaller than the symbols. The solid line is the best linear least-square fit, with the parameters A and B shown.

It would also be desirable if further experiments can be devoted to ensure that X-ray radiation does not affect the track and bulk etching velocities if the thickness is controlled before and after etching. Infrared spectroscopy is also a non-destructive method, which has been used by Barillon et al. (2002) to quantify the loss of nitrate functions under ion irradiation. The technique can also be applied for thickness measurements.

3.2. CR-39 detector

When we applied the above methodology for LR 115 to the CR-39 detectors, no EDXRF K_β line intensities for the chemical element of Ag (energy = 2.984 keV) could be observed. The thickness of the CR-39 detector is about 1000 μm, and is several micrometers smaller after etching, so the detector effectively absorbs all the low energy XRF photons including those at the Ag K_β energy (2.984 keV). Higher energy XRF photons are needed to penetrate through the detector, but then they would not be sensitive enough to show observable different intensities before and after etching.

In other words, we failed to identify a chemical element whose characteristic fluorescent X-ray energy is high enough to avoid total absorption by the SSNTD but also low enough

to be sufficiently sensitive to the etched layer thickness of the detector. As a result, EDXRF is not an appropriate method for measuring the thickness of CR-39 detector. Alternative methods have yet to be explored for the CR-39 detector. Fortunately, however, it is likely that the thickness of removed layer in CR-39 detectors can be adequately surrogated by the etching period, so these methods might not be as critical as those for the LR 115 detectors (Ho et al., 2003).

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