

Sensitivity of LR 115 SSNTD in a diffusion chamber

S.Y.Y. Leung^a, D. Nikezic^{a,b}, J.K.C. Leung^c, K.N. Yu^{a,*}

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

^b Faculty of Sciences, University of Kragujevac, Serbia

^c Department of Physics, The University of Hong Kong, Pokfulam Road, Hong Kong

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Abstract

Solid-state nuclear track detectors (SSNTDs), such as LR 115, have been commonly used in diffusion chambers for long-term measurements of radon gas concentrations. For the LR 115 SSNTD, it has been found that the active layer removed during chemical etching is significantly affected by the presence and amount of stirring, and thus cannot be controlled easily. However, the sensitivity of the LR 115 detector inside a diffusion chamber to the radon and/or thoron gas concentration is dependent on the actual removed active layer thickness. This relationship is dependant on the geometry of the diffusion chamber and the deposition fraction of ^{218}Po in the diffusion chamber, as well as the V function for the LR 115 detector (V is the ratio between the track etch velocity V_t to the bulk etch velocity V_b). This paper presents the experimentally determined relationships between the sensitivity of the LR 115 detector inside a Karlsruhe diffusion chamber and the removed active layer thickness, for both radon and thoron. A V function was adjusted to simulate the relationships. In particular, for the case of ^{222}Rn , we have found $f \sim 0.5$, where f is the fraction of ^{218}Po which decays inside the diffusion chamber before deposition onto available inner surfaces of the chamber. In conclusion, we have found that the sensitivities critically depend on the actual removed active layer thickness, so this should be monitored and used in determining the sensitivities.

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1. Introduction

Radon ^{222}Rn is a naturally occurring radioactive inert gas with a half-life of 3.82 days. It is well established that short-lived radon progeny can cause lung cancers. The alpha track method is one of the standard methods for long-term measurements of radon gas concentrations. In this method, a piece of solid state nuclear track detector (SSNTD), the mostly commonly used being the LR 115 and CR-39 SSNTDs, is placed inside a diffusion chamber. Being a gas, radon can diffuse into the diffusion chamber. The radon gas, together with the progeny formed from radon decay, inside the diffusion chamber can irradiate the SSNTD and leave behind latent tracks. On chemical

etching, the tracks can be visualized and can be counted under an optical microscope with suitable magnification.

Thoron ^{220}Rn is a radioisotope of ^{222}Rn , and has a half-life of 55.6 s. Due to its short half-life, the concentration of thoron is usually much lower than that of radon in normal environments. However, there are cases where the thoron gas concentration cannot be neglected (see e.g. [1–3]). For example, the high ^{232}Th concentration in buildings and rock materials in Hong Kong [1] hinted that the thoron gas concentrations might not be negligible. Studies and surveys on the behavior of thoron and its progeny have been carried out [1,2,4]. To separate the contributions from radon and thoron, two diffusion chambers (the twin diffusion-chamber method [5–7]) are usually used, one for recording mainly the concentrations of radon while the other for recording the concentrations of (radon + thoron), so that the difference between the signals recorded by the

* Corresponding author. Tel.: +852 27887812; fax: +852 27887830.
E-mail address: peter.yu@cityu.edu.hk (K.N. Yu).

SSNTDs inside the two diffusion chambers can give the thoron concentration.

The technique of using diffusion chamber has been widely used and described in the literature [8–10], and a recent review on the SSNTDs has been given in [11]. In the present work, the SSNTD studied will be the LR 115 detector (from DOSIRAD, France).

In applying the alpha track method, the track density (number of tracks per unit area) per unit time on the LR 115 SSNTD is measured, which has the unit of $[m^{-2} s^{-1}]$. The average radon or thoron gas concentration (in $[Bq m^{-3}]$) during the exposure period is obtained by dividing the track density per unit time with the sensitivity (in $[m]$). However, it is apparent that different track densities, and thus different sensitivities, will be obtained for different removed active layer thickness of the LR 115 SSNTDs. The first objective of the present paper is to experimentally determine the sensitivities of the LR 115 SSNTD for different removed active layer thickness for ^{222}Rn and ^{220}Rn .

This relationship between the sensitivities and the removed active layer thickness is related to the geometry of the diffusion chamber and the deposition fraction of ^{218}Po in the diffusion chamber [12], as well as the V function for the LR 115 detector (V is the ratio between the track etch velocity V_t and the bulk etch velocity V_b). Therefore, the experimental data would also provide very useful information for examination and validation of the employed V function and the deposition fraction of ^{218}Po in the diffusion chamber. The latter forms the second objective of the present paper.

2. Experimental methodology

The diffusion chambers employed for the present study were conical, with the inner base radius of 2.35 cm, top radius of 3.35 cm and height of 4.8 cm, and are commonly referred to as the “Karlsruhe” diffusion chambers. There were holes in the brim of chamber, which allow air to flow in and out of the chamber (see Fig. 1). This type of diffu-

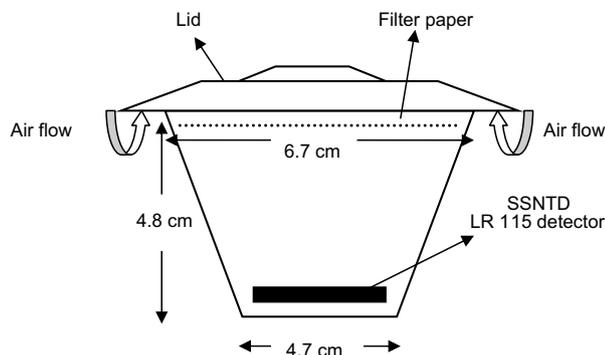


Fig. 1. The dimensions of the Karlsruhe diffusion chamber used in the present investigation with a top radius of 3.35 cm, inner base radius of 2.35 cm and height of 4.8 cm. The SSNTD and the filter paper were placed on the bottom and across the top of the diffusion chamber, respectively.

sion chamber has been used for routine measurements of radon concentrations.

The LR 115 detectors (Type 2, non-strippable) were purchased from DOSIRAD, France. The LR 115 detector consists of a 12 μm red cellulose nitrate active layer and 100 μm clear polyester base substrate as declared by the manufacturer. A piece of 3 cm \times 3 cm LR 115 detector was placed on the bottom of the diffusion chamber and a filter paper across the top to prevent radon/thoron progeny from getting inside the diffusion chamber.

The diffusion chambers with the SSNTDs were inserted into the radon/thoron exposure chamber [13] through an air lock. The experimental set-up for the exposures chamber is shown in Fig. 2. The exposures were carried out in a re-circulating mode at a flow rate of about 2 L min^{-1} for 1 h. Both the radon and thoron gas concentrations inside the exposure chamber were monitored by RAD-7 (DurrIDGE Company Inc., MA) with a sampling period of 30 min.

After the exposure of desired durations, the SSNTDs were removed from the diffusion chambers for chemical etching in 10% aqueous NaOH at 60 $^{\circ}C$ for approximately 1 h. The temperature was kept constant with an accuracy of ± 1 $^{\circ}C$. The detectors were etched using a magnetic stirrer (Model no: SP72220-26, Barnstead/ThermoLyne, Iowa, USA) so as to provide more uniform etching [14]. After etching, the detectors were taken out of the etchant, rinsed with de-ionized water and dried.

For the LR 115 SSNTD, it has been found that the removed active layer during chemical etching is significantly affected by the presence and amount of stirring, and thus cannot be controlled easily [14]. Different methods have been used to measure the active layer thickness of LR 115 detectors, e.g. surface profilometry [14,15], mass difference determination [16]; absorption of X-ray fluorescence photons [17], infrared absorption [18] and gray level determination [19]. The infrared absorption method [18] is adopted in the present work. The active layer thickness of the detector was obtained through the infrared absorption determined using a Perkin–Elmer Fourier transform infrared (FTIR) spectroscopy system (Model 16 PC FT-IR) for 10 cycles. The scanned diameter was 9 mm so the scanned area was 0.64 cm^2 . The exponential decay relationship between the infrared transmittance at the wave number

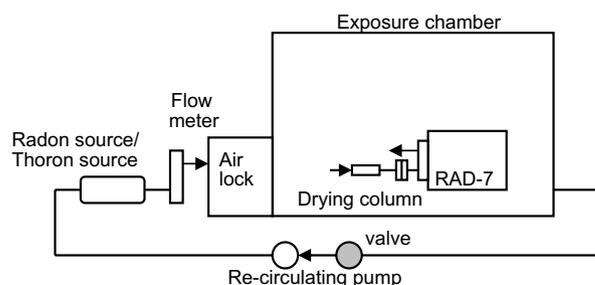


Fig. 2. The block diagram shows the experimental set-up for preparation of the exposure chamber.

at 1598 cm^{-1} (corresponding to the O–NO₂ bond in the cellulose nitrate) and the thickness of the active layer [18] was used for the present study. For each detector, the IR transmittance values were measured at six different locations to give the average values.

The alpha tracks registered by the detector were then counted under an optical microscope with 200× magnification and only those completely perforated the active layers of the LR 115 SSNTDs were counted. The above steps were repeated for different removed-layer thickness. Knowing the time of exposure and with the radon/thoron concentrations given by RAD7, the sensitivities corresponding to different removed active-layer thickness were determined.

3. Results and discussion

3.1. Experimental results for the sensitivities

The experimental results for the sensitivities of the LR 115 detector inside the Karlsruhe diffusion chamber for various removed active-layer thickness are shown in Figs. 3 and 4 for ²²²Rn and ²²⁰Rn, respectively. It can be observed that the thoron data have larger fluctuations when compared with the radon data. This might be due to the highly non-uniform distributions of ²²⁰Rn inside the exposure chamber caused by its very short half-life. These experimental results will be interpreted together with the results generated from computer simulations, which will be described in Section 3.2.

3.2. Computer simulations

As mentioned above, the relationship between the sensitivities and the removed layer is related to the V function of

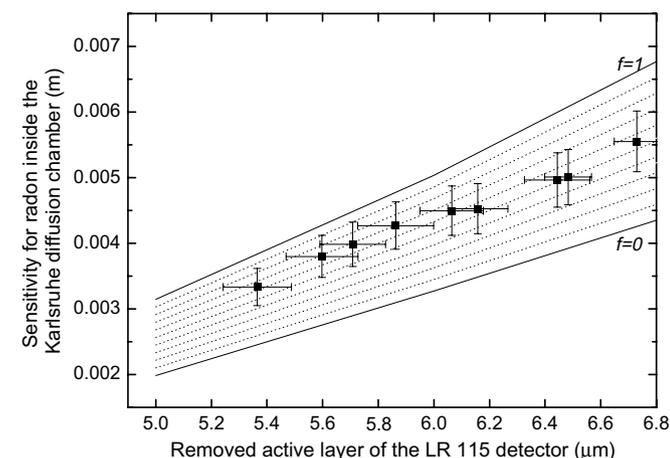


Fig. 3. The experimental data (solid squares) for the sensitivity of the LR 115 detector inside the Karlsruhe diffusion chamber for radon for different removed active-layer thickness, and the best fit lines generated using the V function from Barillon et al. [20] scaled up by a factor of 1.15, with f ranging from 0 to 1 in steps of 0.1.

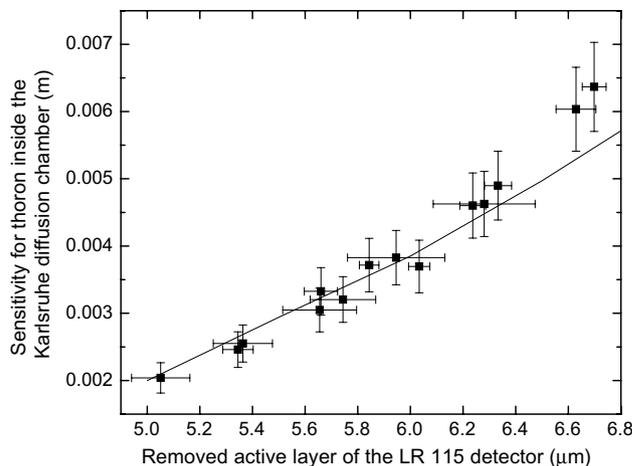


Fig. 4. The experimental data (solid squares) for the sensitivity of the LR 115 detector inside the Karlsruhe diffusion chamber for thoron for different removed active-layer thickness, and the best fit line generated using the V function from Barillon et al. [20] scaled up by a factor of 1.15.

the LR 115 detector. Barillon et al. [20] published a V function for alpha-particle tracks in LR 115 in the form

$$V_t = V_b + \frac{1}{a_1^2 + \left[a_2 R' - \frac{1}{a_3 R'} \right]^2} \quad (1)$$

with constants $a_1 = 0.23$, $a_2 = 0.032$, $a_3 = 3.8$ and R' is the residual range. This function will be used as the starting point in the present studies. From this V function, we can simulate the relationship between the sensitivities and the removed layer. In the case for ²²²Rn, however, this relationship is also related to the deposition fraction of ²¹⁸Po in the diffusion chamber. To minimize the uncertainty, we begin with the simulation for the case of ²²⁰Rn. To best match the experimental data, the V function has been scaled up slightly by a factor of 1.15. It is interesting to find that the simulated relationship is very sensitive to the V function. In other words, the experimental data can provide a very sensitive test of the V function. The relationship obtained using this scaled-up V function is shown in Fig. 4. It is exciting to see that the simulated curve reproduces the trend of the experimental data very satisfactorily.

The case for ²²²Rn is a bit more complicated. After ²²²Rn diffuses into the diffusion chamber, ²¹⁸Po will be formed. Due to its short half-life, a fraction f of ²¹⁸Po decays before deposition onto available inner surfaces of the chamber. Previously, we have determined f to be ~ 0.4 , which seems to be independent of the shape and dimensions of the diffusion chambers, the surface to volume ratios or the internal surface materials of the diffusion chambers [12,21].

By using the scaled-up V function, the relationships between the sensitivities and the removed layer for ²²²Rn have been calculated for $f = 0$ –1, with steps of 0.1. These curves are shown in Fig. 3 together with the experimental data. For best fitting the experimental data, f is likely to be ~ 0.5 (with an uncertainty of ~ 0.1). This is close to

the value of ~ 0.4 from our previous studies [12,21] (also with a typical uncertainty of about 0.1).

3.3. Practical considerations

The previous sections have shown that the sensitivities of LR 115 inside the diffusion chamber critically depends on the actual removed active layer thickness (see Figs. 3 and 4). Therefore, the correct sensitivities should be used to determine the radon and/or thoron concentrations from the track densities recorded on the SSNTD inside the diffusion chamber. It is noted that many investigators have adopted a fixed sensitivity value, which is either borrowed from other works or from their own “calibration”. However, such an approach has inherently assumed the same bulk etch rate V_b for the same etching temperature and the same etchant. However, as pointed out by Yip et al. [14], this is in general not true as V_b significantly depends on the amount of stirring during etching, which is very difficult, if not impossible, to control. For example, Yip et al. [14] found that the bulk etch rate for LR 115 detector etched using 2.5 N aqueous NaOH at 60 °C, with and without stirring as 6.65 and 3.61 $\mu\text{m h}^{-1}$, respectively. Even with magnetic stirring, we have monitored the bulk etch rates we have encountered in the present studies and have found that they have varied between about 5 and 7 $\mu\text{m/h}$.

Errors will occur from not controlling the removed active layer thickness. To show the magnitude of the possible errors, suppose we expect the removed layer is 5 μm but the actual removed layer is 6 μm . For ^{222}Rn , the expected sensitivity is 0.0026 m but the actual sensitivity should be 0.0042 m (determined from the curve for $f=0.5$ in Fig. 3). In this way, the determined ^{222}Rn concentration is overestimated by more than 60%. For ^{220}Rn , the expected sensitivity is 0.002 m but the actual sensitivity should be 0.0038 m (determined from Fig. 4). In this way, the determined ^{220}Rn concentration is overestimated by 90%. Similarly, if the expected removed layer is larger than the actual removed layer, there will be underestimation of the ^{222}Rn and ^{220}Rn concentrations. The large percentage errors advocate that the actual removed active layer thickness should be monitored and used in determining the sensitivities.

4. Conclusions

- (1) The sensitivities of the LR 115 SSNTD inside a Karlsruhe diffusion chamber have been experimentally determined for different removed active layer thickness for ^{222}Rn and ^{220}Rn , and for chemical etching using 10% aqueous NaOH at 60 °C with magnetic stirring. The thickness of the active layer was determined by the infrared transmittance at the wave number at 1598 cm^{-1} (corresponding to the O–NO₂ bond in the cellulose nitrate).
- (2) The V function for the LR 115 detector [20] was employed to simulate the relationship between the

sensitivities and the removed active layer thickness. We began with the case of ^{220}Rn . To best match the experimental data, the V function has been scaled up slightly by a factor of 1.15. The simulated curve reproduces the trend of the experimental data very satisfactorily.

- (3) By using the scaled-up V function, the relationships between the sensitivities and the removed layer for ^{222}Rn can be best fitted for $f \sim 0.5$, where f is the fraction of ^{218}Po which decays inside the diffusion chamber before deposition onto available inner surfaces of the chamber. This is close to the value of ~ 0.4 from previous studies [12,21].
- (4) The sensitivities of LR 115 inside the diffusion chamber critically depend on the actual removed active layer thickness. If the removed active layer thickness is not controlled or measured, erroneous results will be obtained. For example, over-estimations of 60% and 90% in the ^{222}Rn and ^{220}Rn concentrations, respectively, will result if the removed layer of 6 μm is underestimated as 5 μm . The large percentage errors advocate that the actual removed active layer thickness should be monitored and used in determining the sensitivities.

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