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Long-term measurements of equilibrium factor with electrochemically etched CR-39 SSNTD

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Abstract

Recently, our group proposed a method (proxy equilibrium factor method) using a bare LR 115 detector for long-term monitoring of the equilibrium factor. Due to the presence of an upper alpha-particle energy threshold for track formation in the LR 115 detector, the partial sensitivities to ^{222}Rn, ^{218}Po and ^{214}Po were the same, which made possible measurements of a proxy equilibrium factor F_p that was well correlated with the equilibrium factor. In the present work, the method is extended to CR-39 detectors which have better-controlled etching properties but do not have an upper energy threshold. An exposed bare CR-39 detector is first pre-etched in 6.25 N NaOH solution at 70 °C for 6 h, and then etched electrochemically in a 6.25 N NaOH solution with ac voltage of 400 V (peak to peak) and 5 kHz applied across the detectors for 1 h at room temperature. Under these conditions, for tracks corresponding to incident angles larger than or equal to 50°, the treeing efficiency is 0% and 100% for incident energies smaller than and larger than 4 MeV, respectively. A simple method is then proposed to obtain the total number of tracks formed below the upper energy threshold of 4 MeV, from which the proxy equilibrium factor method can apply.
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1. Introduction

Inhaled radon (^{222}Rn) progeny are the most important source of irradiation of the human respiratory tract. Methods for long-term monitoring of the ^{222}Rn gas itself are well established, such as through the use of solid-state nuclear track detectors (SSNTDs) (see e.g. [1,2] for surveys). However, it is established that radon gas measurements are not adequate for epidemiological studies and case control studies of radon induced lung cancer [3]. Methods for long-term monitoring of the concentrations of radon progeny, or the equilibrium factor (which surrogates the ratios of concentrations of radon progeny to the concentration of the ^{222}Rn gas), are still being explored. The equilibrium factor of radon progeny is defined as

$$F_{^{222}\mathrm{Rn}} = 0.105 f_{^{218}\mathrm{Po}} + 0.515 f_{^{214}\mathrm{Pb}} + 0.380 f_{^{214}\mathrm{Bi,Po}} \tag{1}$$

where f_i is the ratio of the activity concentration of the ith radon decay product to that of ^{222}Rn, i.e. $f_1 = f_{^{218}\mathrm{Po}}$, $f_2 = f_{^{214}\mathrm{Pb}}$ and $f_3 = f_{^{214}\mathrm{Bi,Po}}$.

Surveys of existing methods for determining the equilibrium factor and radon progeny concentrations have recently been made [2,4,5]. Recently, a new method was given by Amgarou et al. [4] who proposed measurements of the equilibrium factor through the so-called "reduced" equilibrium factor, which is defined as

$$F_{\mathrm{Red}} = 0.105 f_{^{218}\mathrm{Po}} + 0.380 f_{^{214}\mathrm{Bi,Po}}. \tag{2}$$

It was shown that the total equilibrium factor depended on F_{Red} in a very good manner.

More recently, our group has proposed a novel method (referred to as the proxy equilibrium factor method or simply the F_p method) using a bare LR 115 detector for

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long-term monitoring of the equilibrium factor [5–7]. We studied the partial sensitivities ρ_i of the bare LR 115 detector to ^{222}Rn and its alpha emitting short-lived progeny, ^{218}Po and ^{214}Po, i.e. the number of tracks per unit area per unit exposure (with the unit (m^{-2})/(Bq m^{-3} s) or just (m)). Due to the presence of an upper alpha-particle energy threshold for track formation in the LR 115 detector, the partial sensitivities to ^{222}Rn, ^{218}Po and ^{214}Po were all the same. This makes possible measurements of a proxy equilibrium factor F_p, which was defined as $(f_1 + f_3)$. In particular, $F_p = (\rho/\rho_i t C_0) - 1$, where ρ (track/m^2) is the total track density on the detector, t (s) is the exposure time and C_0 (Bq/m^3) is the concentration of ^{222}Rn. F_p was also found to be well correlated with the equilibrium factor between radon gas and its progeny.

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 [8]. Different methods have been used to measure the active layer thickness of LR 115 detectors [8–13]. For simpler applications, it is pertinent to explore the possibility of extending the proxy equilibrium factor method to other SSNTDs, of which the etching can be more easily controlled. In the present work, the method is extended to CR-39 detectors which have better-controlled etching properties [14]. However, the validity of the proxy equilibrium factor method relies on the presence of an upper alpha-particle energy threshold for track formation in the SSNTDs. Since CR-39 detectors do not have a relevant upper energy threshold, we explore in this paper a method, based on electrochemical etching (ECE), for quick determination of the number of tracks which are formed from alpha particles with incident energies above a chosen threshold. It is remarked here that the present work only serves as a feasibility study, so the etching conditions employed might not be the optimal ones and can be fine-tuned in the future.

2. Methodology

As described above, the main objective of the present study is to explore a method based on ECE to determine the number of tracks which are formed from alpha particles with incident energies above a chosen threshold. As a demonstration of the method, we have chosen this threshold as 4 MeV. As mentioned in the introduction, this is not necessarily the optimal choice. Other threshold energies can also be chosen, which will lead to different pre-etching conditions and different partial sensitivities of proxy equilibrium method. After choosing the threshold energy as 4 MeV, our first task will be to determine the optimum pre-etching time through chemical etching (CE) in such a way that all the tracks resulted from alpha particles with energies lower than 4 MeV become over-etched and will not form trees on ECE.

After determining the optimum pre-etching time for 4 MeV alpha particles during CE, we also need the ECE efficiency for alpha particles with incident energies at or above 4 MeV for different incident angles. This ensures that all the tracks from these alpha particles will be accounted for when calculating the number of tracks formed below the upper energy threshold.

The CR-39 detectors used in the present study were purchased from Page Mouldings (Perhsore) Limited (Worcestershire, England). The thickness of the CR detectors were ~150 μm.

2.1. Determination of the optimum pre-etching duration

The CR-39 detector for our studies were cut to a size of 2.2×2.2 cm^2. Alpha particles with 4 MeV were irradiated under normal incidence on a detector. The alpha source employed in the present study was a planar ^{241}Am source (main alpha energy = 5.49 MeV under vacuum). The energy of the alpha particles was varied by changing the source to detector distance through a collimator under atmospheric pressure. The relationship between the alpha energy and the air distance traveled by an alpha-particle with initial energy of 5.49 MeV from ^{241}Am was obtained by measuring the energies for alpha particles passing different distances through normal air using alpha spectroscopy systems (ORTEC Model 5030) with passivated implanted planar silicon (PIPS) detectors of areas of 300 mm^2.

The pre-etching treatment applied in this work was carried out using CE with 6.25 N NaOH solution at 70 °C. Irradiated CR-39 detectors were pre-etched for different durations which result in removing a surface layer with different thicknesses. After CE, the detectors were etched electrochemically in a 6.25 N NaOH solution. A high ac voltage with about 400 V (peak to peak) and 5 kHz was applied across the detectors for 1 h at room temperature. After electrochemical etching (ECE), the detectors were removed from the etchant, rinsed with distilled water and dried in air. The detectors were then scanned under an optical microscope in the transmission mode to count the tracks as well as the trees generated from ECE. The efficiency was obtained by the ratio of the number of trees divided by the total number of tracks from CE.

From the efficiency, an optimum pre-etching duration is determined. The ECE efficiencies for other incident alpha energies as well as other pre-etching durations are further studied to ensure the feasibility of the chosen pre-etching duration.

2.2. The ECE efficiency in different incident angles

After determining the optimum pre-etching duration for 4 MeV alpha particles during CE, the ECE efficiency of the 4 and 4.5 MeV alpha particles for different incident angles were also determined. Separate detectors (with area of 2.2×2.2 cm^2) were used to study the tracks for 4 and 4.5 MeV alpha particles. On the same detector, alpha particles with the same incident energy (4 or 4.5 MeV) were irradiated at 4 different regions, each region corresponding to one incident angle. The incident angles studied range from 30° to 90° with steps of 20°.

After pre-etching using CE for the optimum etching duration, the detectors were transferred to ECE treatment (with conditions the same as those described above). After ECE, the detectors were scanned under optical microscope to obtain the ECE efficiency.

3. Results and discussion

3.1. Determination of the optimum pre-etching duration

To determine the optimum pre-etching duration for 4 MeV alpha tracks, the corresponding ECE efficiencies were measured. Fig. 1 shows the variation of the ECE efficiency with the pre-etching duration for 4 MeV alpha-particle tracks with normal incidence. The ECE efficiency is 100% at the beginning, but it drops rapidly starting from 7 h of pre-etching. It is likely that the 4 MeV alpha-particle tracks start to become over-etched after 7 h of pre-etching. Rounded tracks in the over-etched phase fail to produce a sufficiently high electric field for the treeing phenomenon to occur. By also taking into consideration the ECE efficiencies for different incident angles of the alpha particles, the optimum pre-etching duration has been chosen as 6 h.

As mentioned above, to ensure the feasibility of choosing 6 h as the optimum pre-etching duration for 4 MeV alpha tracks, the ECE efficiencies for other incident alpha energies as well as other pre-etching durations are further studied.

Table 1 shows the ECE efficiencies for tracks from alpha particles with incident energies of 3.5, 4 and 4.5 MeV and normal incidence after 6–7 h pre-etching treatment. The data show that the ECE efficiency is almost 100% for alpha particles with incident energies of 4 and 4.5 MeV after 6–6.5 h pre-etching. In contrast, 3.5 MeV alpha-particle tracks fail to produce ECE trees and give 0% efficiency for 6–7 h pre-etching. Therefore, 6 h proves to be a feasible pre-etching duration for discrimination of tracks from normally incident alpha particles with incident energies below and above 4 MeV.

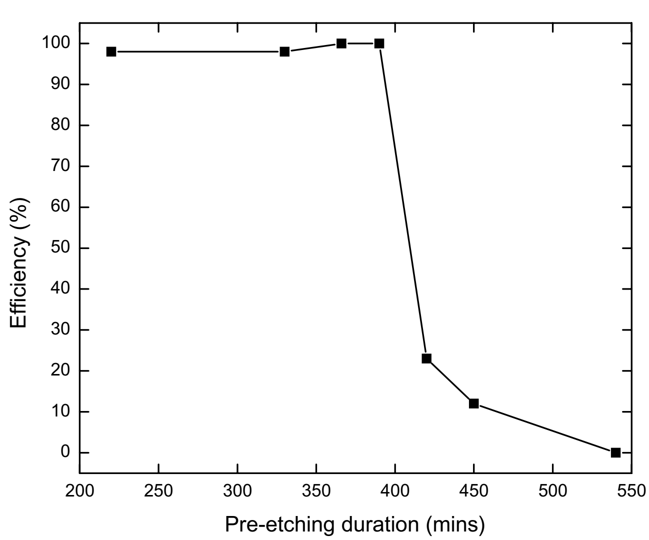

Fig. 1. Variation of the ECE efficiency with the pre-etching duration for 4 MeV alpha tracks with normal incidence.

Table 1
The ECE efficiencies for alpha particles with incident energies of 3.5, 4 and 4.5 MeV (for normal incidence)

Incident alpha energy (MeV)	6 h etching	6.5 h etching	7 h etching
3.5	0	0	0
4.0	100	100	23
4.5	100	100	97

3.2. The ECE efficiency for different incident angles

After choosing the pre-etching duration as 6 h, the ECE efficiency of the 4 and 4.5 MeV alpha particles for different incident angles were also determined. Figs. 2 and 3 show the images of the tracks from 4 and 4.5 MeV alpha particles, respectively, after 6 h pre-etching and ECE with focus

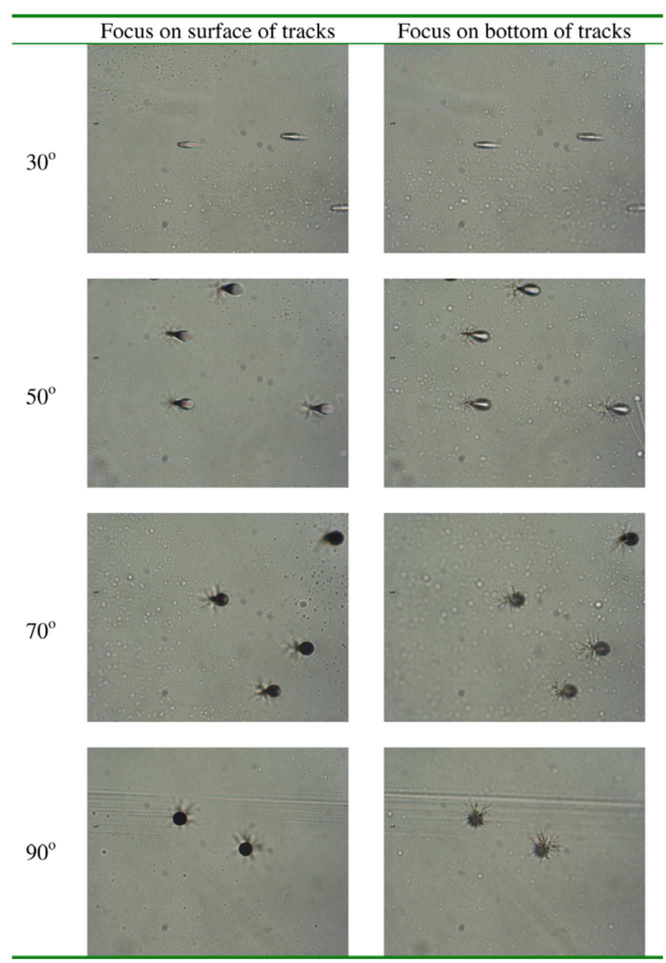

Fig. 2. Images of tracks from 4 MeV alpha particles with incident angles of 30°, 50°, 70° and 90°. Images in the left column refer to those with focus on the surface of the tracks, while those in the right column refer to those with focus on the bottom of the tracks.

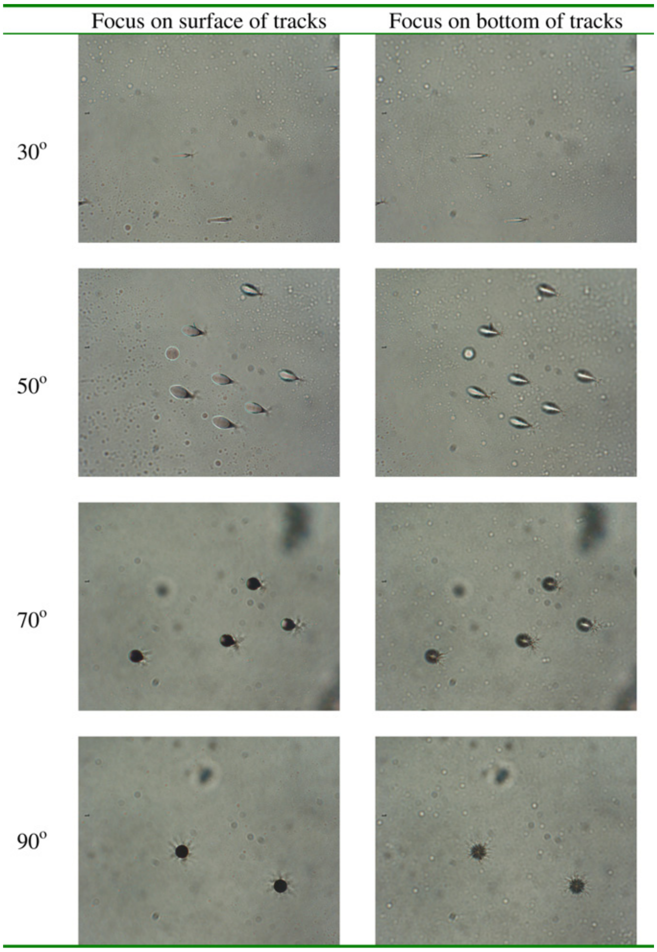

Fig. 3. Images of tracks from 4.5 MeV alpha particles with incident angles of 30°, 50°, 70° and 90°. Images in the left column refer to those with focus on the surface of the tracks, while those in the right column refer to those with focus on the bottom of the tracks.

on the surface and the bottom of the tracks. Except for the incident angle of 30°, trees from ECE are clearly seen to have formed, even if the focus is on the surface of the tracks. Identification of ECE trees for the incident angle of 30° requires greater effort and need focusing on the bottom of the tracks.

Table 2 shows the ECE efficiency of tracks from alpha particles with incident energies of 3.5, 4 and 4.5 MeV and different incident angles for 6 h pre-etching in 6.25 N NaOH solution at 70 °C. It is interesting to see that the ECE efficiencies go to extreme values (either 0% or 100%) for incident angles larger than or equal to 50°.

Table 2
The ECE efficiency of tracks from alpha particles with incident energies of 3.5, 4 and 4.5 MeV for incident for different incident angles (for 6 h pre-etching in 6.25 N NaOH solution at 70 °C)

Incident angle	3.5 MeV	4 MeV	4.5 MeV
30°	0%	0%	100%
50°	0%	100%	100%
70°	0%	100%	100%
90°	0%	100%	100%

3.3. Feasibility to extend the F_p method to CR-39 detectors with CE and ECE

In order to study the feasibility of extending the proxy equilibrium factor method to CR-39 detectors with CE and ECE, it is useful to look at the track parameters of tracks from 4 MeV alpha particles. In this feasibility study, we used our track etch function (V_t) previously determined for our detectors and etching conditions as

$$V = \frac{V_t}{V_b} = 1 + e^{-0.068R'+1.1784} - e^{-0.6513R'+1.1784} \qquad (3)$$

where R' is the residual range of alpha particles [15]. From this V_t function, and using a bulk etch rate of 1.2 μm/h [14] and an etching duration of 6 h, the track parameters, including the major axis, minor axis and track depth of the tracks from alpha particles with different incident energies and incident angles are calculated. There is no track formation for the incident angle of 10° and for many other cases. We also regard tracks with too small track depths (<0.5 μm) as unobservable. It is particularly noted that the tracks from 4.5 MeV alpha particles with an incident angle of 30° have a depth of only 0.59 μm, but it still has 100% ECE efficiency (see Table 2). All other observable tracks in Table 3 have larger depths. Therefore, it is reasonable to assume that all the observable tracks from alpha particles with incident alpha energies larger than 4.5 MeV will have 100% ECE efficiency because the incident angles are larger and the tracks have the same or larger depths, both favoring ECE treeing.

Table 3
Parameters for tracks from alpha particles with different incident energies (E) and incident angles and etched with 6.25 N NaOH solution at 70 °C for 6 h

E (MeV)	Incident angle						
	20°	30°	40°	50°	60°	70°	80°	90°
4.0	NO	10.82	11.27	9.93	9.07	8.53	8.23	8.13
		2.74	5.11	6.49	7.30	7.78	8.04	8.13
4.5	NO	6.79	10.12	8.23	8.98	7.76	7.49	7.41
		1.43	3.71	5.47	6.44	7.01	7.31	7.41
5.0	NO	NO	7.27	8.01	7.37	6.96	6.74	6.67
			2.07	4.34	5.53	6.20	6.55	6.67
5.5	NT	NT	NO	7.04	6.50	6.16	5.97	5.90
				2.98	4.55	5.36	5.77	5.90
6.0	NT	NT	NT	NO	5.65	5.36	5.20	5.15
					3.49	4.51	5.00	5.15
6.5	NT	NT	NT	NO	4.84	4.61	4.47	4.43
					2.28	3.65	4.25	4.43
7.0	NT	NT	NT	NT	NO	3.90	3.79	3.75
						2.79	3.54	3.75
7.5	NT	NT	NT	NT	NT	NO	3.15	3.13
							2.86	3.13
7.69	NT	NT	NT	NT	NT	NO	2.93	2.90
							2.62	2.90

There is no track formation for incident angle of 10°. For each incident energy, there are two rows of data: the first is the major axis (μm), and the second is the minor axis (μm). NO means no observation due to the very small track depth (<0.5 μm), while NT means no track development.

We propose the following methodology to determine the number of tracks which are formed from alpha particles with incident energies below the chosen threshold of 4 MeV. When we count the tracks with trees after ECE, we disregard all those with incident angles smaller than 50°. These tracks should be easily recognized since they have either exceptionally long major axis (>10 μm) or exceptionally short minor axis (<2.1 μm), and the number of these tracks should be relatively small (see Table 3). As mentioned before, the ECE trees around these tracks are more difficult to observe anyway, and may require changing the focus of the microscope to the bottom of the tracks. By disregarding all those alpha tracks with incident angles smaller than 50°, some economy is gained without a significant loss of information since the number of these tracks is relatively small anyway.

After getting the total number of tracks $N_{>50}$ corresponding to incident angles larger than or equal to 50° (all with incident energies larger than 4 MeV), the total number of tracks $N_{>30}$ corresponding to incident angles larger than or equal to 30° (also with incident energies larger than 4 MeV) can be theoretically calculated assuming homogeneity of radon gas and progeny atoms in air. This conversion can be easily calculated from the V_t function, the bulk etch rate and the etching duration, and should be simply related through a scale-up factor f, i.e. $N_{>30} = f \times N_{>50}$. The scale-up factor f needs to be computed only once and can be applied for all future uses. By subtracting $N_{>30}$ from the total number of tracks N_T obtained from CE, the number of tracks $N_{<4\ \mathrm{MeV}}$ formed below the upper energy threshold of 4 MeV can be obtained. Once $N_{<4\ \mathrm{MeV}}$ is obtained, the F_p method can be applied.

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