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Passive monitoring of the equilibrium factor inside a radon exposure chamber using bare LR 115 SSNTDs

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Abstract

In this paper, we propose and study a method using bare LR 115 SSNTDs for passive monitoring of the equilibrium factor F inside a radon exposure chamber. The method is based on the proxy equilibrium factor F_p which is defined as $F_p = \rho/(\rho_i C_0 t) - 1$, where ρ_i is the partial sensitivity (i.e., the number of tracks per unit area per unit exposure), C_0 is the radon gas concentration and t is the exposure time. F_p was found to be well correlated with F. LR 115 detectors were exposed in a huge exposure chamber with a volume of 43 m^3, with integrated radon exposure and equilibrium factor as 100 kBq m^{-3} h and 0.680, respectively. The relationship between ρ_i (m) and the removed active layer thickness x (μm) has been determined as $\rho_i = -0.0041 + 0.0011x$. The method has also been verified through determination of the equilibrium factor from the track densities on the LR 115 detectors. The mean of the mid points of the determined ranges of F for a few LR 115 SSNTDs can give a very good estimate of the true equilibrium factor.

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Keywords: Radon progeny; Equilibrium factor; LR 115 detector; Detector sensitivity

1. Introduction

It is well established that short-lived radon progeny contributes about half of the total exposure of human beings to ionizing radiation. It is also well known that the radon dose delivered to the lungs is not attributed to the radon gas itself, but instead to its short-lived progeny. Exposure to radon progeny is measured through the product of PAEC × t, where PAEC is the potential alpha energy concentration and t is the exposure time. The ratio of PAEC to the radon gas concentration can be surrogated by the equilibrium factor F, which is defined by

$$F = 0.105f_1 + 0.515f_2 + 0.380f_3, \quad (1)$$

where $f_i = C_i/C_0$, and C_0, C_1, C_2 and C_3 are the activitiy concentrations (in Bq m^{-3}) for ^{222}Rn, ^{218}Po, ^{214}Pb, and ^{214}Bi(Po), respectively.

In many epidemiological studies as well as in case control studies of the radon risk, the excess number of cancers are related to the radon gas exposure but not to the radon progeny exposure. However, it has been shown that radon gas measurements are not adequate for such studies [1]. Instead, measurements of radon progeny exposure, and in particular those long-term measurements, are important. Recent researches have proposed to employ solid-sate nuclear track detectors (SSNTDs) for long-term measurements of radon progeny exposure (see e.g., Refs. [2–4]). A review on SSNTDs and their applications for measurements of radon and its progeny can be found in Ref. [5].

The reliability of these methods invariably depends on the validity and accuracy of calibrations, for which exposure chambers are indispensable. While relatively small-size laboratory-based exposure chambers for ^{222}Rn gas are relatively easy to construct and operate [6], those for ^{222}Rn progeny are more complicated (e.g., Ref. [7]). In particular, the PAEC or F values inside the exposure chambers for ^{222}Rn progeny are usually monitored through active grab sampling, which can sometimes present

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problems since the aerosol concentrations (and thus the PAEC and F) can be significantly disturbed. And in order to avoid excessive disturbance of the conditions inside the exposure chamber, the grab sampling cannot be carried out too frequently. Therefore, some changes in the PAEC inside the exposure chamber might not be adequately reflected by such short-term grab samplings.

An alternative is to resort to a huge radon chamber, such as that of Health Protection Agency (UK), which has a volume of 43 m^3. In such a big chamber, the disturbance of the aerosol concentrations and thus the PAEC and F will be negligible during the active grab sampling. However, it would be more convenient and economical if the PAEC and F in laboratory-based exposure chambers for radon progeny can be correctly measured.

In this paper, we will propose and study a method based on the proxy equilibrium factor F_p for passive monitoring of the equilibrium factor inside a radon exposure chamber, which will not disturb the aerosol concentrations or the PAEC and which will give an integrated average value over the entire exposure period.

2. Proxy equilibrium factor F_p

Nikezic et al. [3] and Yu et al. [4] proposed to use the bare LR 115 detector for determining the airborne $^{218}Po + ^{214}Po$ concentration, and show that this concentration can be employed to give good estimates of the equilibrium factor in an environment. The LR 115 detector has an upper energy threshold for track formation, which is well below the energy of alpha particles emitted by the radon progeny plate out on the detector, i.e., plate-out progeny are not detected by LR 115.

The responses of the bare LR 115 detector to ^{222}Rn, ^{218}Po and ^{214}Po are expressed by the partial sensitivities ρ_i of the detector to these species (i.e., the number of tracks per unit area per unit exposure, i.e., the unit of $(m^{-2})/(Bq\,m^{-3}\,s)$ or just (m)). The partial sensitivities ρ_i were found to be the same for ^{222}Rn, ^{218}Po and ^{214}Po [3,4]. A simplified explanation is given as follows (neglecting some minor details such as the critical angle for track formation). The probability P of an alpha particle emitted at the point A (r, θ, φ) hitting a point-like detector at the origin of the coordinate system with a surface area S is given as $S\cos\theta/(4\pi r^2)$, so the average detection efficiency is proportional to:

$$\int P\,\mathrm{d}V = \iiint \frac{S\cos\theta}{4\pi r^2}\, r^2 \times \mathrm{d}r\,\sin\theta\,\mathrm{d}\theta\,\mathrm{d}\varphi \propto \int_{R_{\min}}^{R_{\max}} \mathrm{d}r \propto (R_{\max} - R_{\min}), \tag{2}$$

where $R_{\min}$ and $R_{\max}$ are the minimum and maximum distances, respectively, of the alpha emitters from which the alpha-particle energies will fall within the energy window of the LR 115 detector. It is apparent that the $(R_{\max}-R_{\min})$ values are the same for ^{222}Rn, ^{218}Po and ^{214}Po, since the $(R_{\max}-R_{\min})$ value is in fact controlled by the energy window of the LR 115 detector. Therefore, $\rho_i = \rho_{^{222}Rn} = \rho_{^{218}Po} = \rho_{^{214}Po}$. It is remarked here that the equality of partial sensitivities arises because of the presence of the upper energy threshold for recording alpha-particle tracks in LR 115, so the lower limit of integration in Eq. (2) is $R_{\min}$. For other detectors without the upper energy threshold, e.g., CR-39, the lower limit of integration in Eq. (2) becomes 0, so $\int P\,\mathrm{d}V$ will be proportional to $R_{\max}$ instead of $(R_{\max}-R_{\min})$, and the partial sensitivities will not be the same.

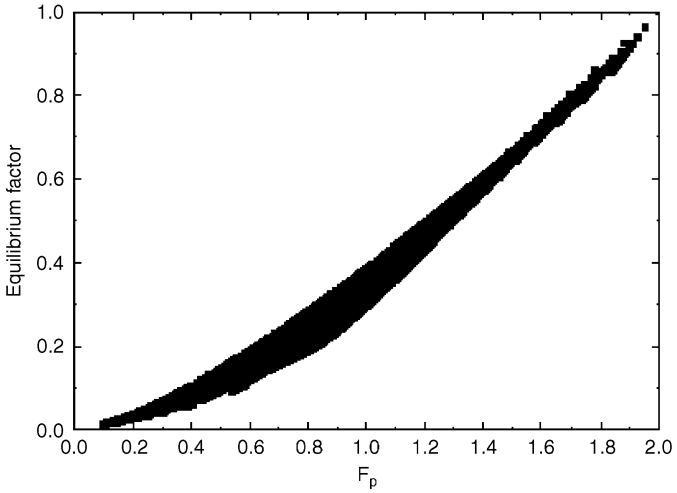

Fig. 1. The relationship between the equilibrium factor F and the proxy equilibrium factor F_p $(=f_1+f_3)$ (Ref. [4]).

Now that the partial sensitivities are equal, the total track density ρ (in track/m^2) on the detector is $\rho = \rho_i(C_0 + C_1 + C_3)t$, where t is the exposure time. The proxy equilibrium factor F_p was defined by [3,4]

$$F_p = f_1 + f_3 = \frac{C_1}{C_0} + \frac{C_3}{C_0} = \frac{\rho}{\rho_i t C_0} - 1. \tag{3}$$

Yu et al. [4] calculated F through the Jacobi room model [8] by systematically varying all parameters that influence the concentrations of radon and its progeny, and plotted them with F_p, and the results are shown in Fig. 1. The proxy equilibrium factor F_p was found to be well correlated with F.

3. Experimental determination of the partial sensitivity

To apply F_p for passive monitoring of the equilibrium factor inside a radon exposure chamber through Eq. (3), we have to know the partial sensitivity ρ_i. The present section will describe the procedures as well as the results on the determination of ρ_i. In particular, ρ_i will depend on the removed active layer thickness of the LR 115 detector.

3.1. Exposure of the LR 115 detectors

The LR 115 detectors (Type 2, non-strippable) used in the present studies were purchased from DOSIRAD,

France. The LR 115 SSNTD consists of a 12 μm red cellulose nitrate active layer on top of a 100 μm clear polyester base. The size of the detectors employed in our experiments was $3 \times 3\,cm^2$. A total of fourteen LR 115 detectors were sent to the Health Protection Agency (HPA), Chilton, UK (previously known as the National Radiological Protection Board or NRPB) for exposure in their walk-in exposure chamber with the volume of $43\,m^3$. The HPA staff adjusted the conditions inside the exposure chamber according to our requirements and the detectors were then placed inside. To ensure that the F_p method is valid, there should not be surfaces or objects at distances shorter than R_{max} in front of the detectors. The highest alpha energy in the radon chain is 7.69 MeV (from ^{218}Po) and the corresponding range in air is less than 70 mm [9]. The detectors were mounted onto a post placed at the centre of the exposure chamber, which was away from the walls much farther than 70 mm.

The reference value of the integrated radon exposure and the equilibrium factor inside the exposure chamber during the exposure period were provided by HPA as $100 \pm 5\,kBq\,m^{-3}\,h$ and 0.680 ± 0.068, respectively. After exposure, the detectors were returned back to our laboratory for analyses.

3.2. Chemical etching

The LR 115 detectors were etched in 10% aqueous solution of NaOH maintained at 60 °C by a water bath. The temperature was kept constant with an accuracy of ±1 °C. During the etching process, the etchant was stirred continuously using a magnetic stirrer (Model no: SP72220-26, Barnstead/Thermolyne, Iowa, USA) to provide more uniform etching [10]. After etching for ~1 h, the detectors were removed from the etchant, rinsed with de-ionized water and dried.

The removed active layer during chemical etching was significantly affected by the presence and amount of stirring, and thus cannot be controlled easily [10]. Different methods have been proposed to measure the active layer thickness of LR 115 detectors, e.g., surface profilometry [11,12], absorption of X-ray fluorescence photons [12], infrared absorption [13] and gray level determination [14]. The infrared absorption method was adopted in the present work.

The infrared transmittance was measured using a Perkin-Elmer Fourier Transform Infrared (FTIR) spectroscopy system (Model 16 PC FT-IR). The removed active layer of the LR 115 detector was determined using the exponential decay relationship between the active layer thickness and the infrared transmittance at the wave number $1598\,cm^{-1}$, which corresponded to the O–NO_2 bond in the cellulose nitrate [13]. The detectors were scanned for 10 cycles, with the scanned diameter of 9 mm and the scanned area of $0.64\,cm^2$. The wave number range employed was between 1700 and $1100\,cm^{-1}$ with a resolution of $4\,cm^{-1}$. For each detector, the IR transmittance values were measured at six different locations to give the average values.

3.3. Relationship between partial sensitivity and removed active layer thickness

As described in Eq. (3), the partial sensitivities can be determined from the track density ρ, the integrated radon exposure ($C_0 \times t$) and the proxy equilibrium factor F_p. After chemical etching and determination of the removed active layer thickness, the alpha-particle tracks registered by the LR 115 detector were counted under an optical microscope with 200× magnification. Only those tracks completely perforated the active layer of the LR 115 detector were counted. This visibility criterion has been introduced in order to minimize the level of subjectivity. The track densities ρ (number of tracks per unit area) were then found. The integrated exposure ($C_0 \times t$) has been directly provided by HPA as $100 \pm 5\,kBq\,m^{-3}\,h$. From the equilibrium factor of 0.680 ± 0.068 provided by HPA, the corresponding F_p value can be read from Fig. 1. For the mid-point value of 0.680, F_p is determined to be from 1.51 to 1.57, which is narrow, so we use the mid value of this range (1.54) for subsequent calculations. The equilibrium factor interval from (0.680−0.068) to (0.680 + 0.068) then correspond to an F_p interval from 1.37 to 1.67. Such an F_p interval is used in determining the relationship between the partial sensitivity and the removed active layer thickness as shown in Fig. 2.

Out of the fourteen LR 115 detectors sent to HPA for exposure, eight were randomly chosen for the determination of the partial sensitivity, while the remaining six were used for verification purposes as described in Section 4 below. From these eight pieces of LR 115 detectors, the experimental relationship between the partial sensitivity

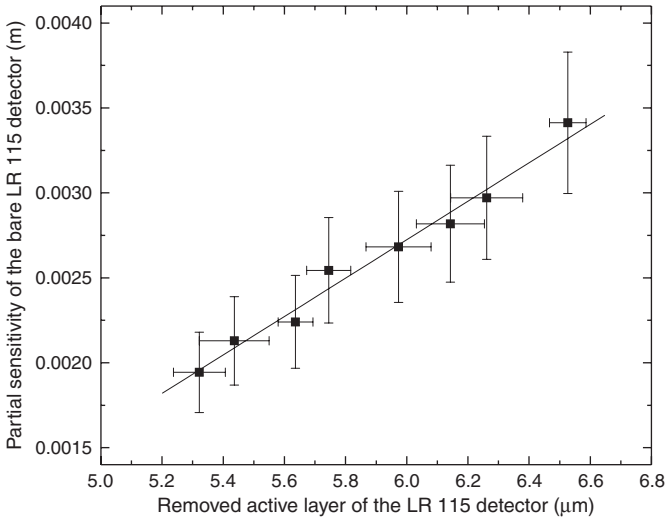

Fig. 2. Relationship between the partial sensitivity ρ_i (m) of the bare LR 115 detector and the removed active layer thickness x (μm). The solid line is the best-fit line represented by $\rho_i = -0.0041 + 0.0011x$, with an R^2 of 0.9895.

and the removed active layer thickness has been determined, which is shown in Fig. 2. It is remarked that the etching durations for the eight detectors have been adjusted to obtain a range of removed active layer thickness. The uncertainties in the partial sensitivity are derived from the range in F_p (from 1.37 to 1.67), which incorporates the uncertainties in the equilibrium factor of 0.680 ± 0.068 provided by HPA and the width of the curve in Fig. 1 (see Section 3.3). On the other hand, the uncertainties in the removed active layers are derived from uncertainties in FTIR measurements.

By fitting the linear relationship $\rho_i = A + Bx$ to the experimental data, where x (μm) is the removed active layer thickness of the LR 115 detector and ρ_i (m) is the partial sensitivity of the bare detector, we have obtained $A = -0.0041 \pm 0.0004$ and $B = 0.0011 \pm 0.0001$, with the coefficient of determination $R^2 = 0.9895$. The large value of R^2 shows that the variation in the partial sensitivity is well explained by the variation in the removed active layer thickness of the LR 115 detector. The fit is observed to be valid for the entire range of the present data set, in which the removed active layer thickness ranges from 5.3 to 6.6 μm.

4. Verification of the F_p method

Out of the fourteen LR 115 detectors sent to HPA for exposure, six were randomly chosen for verification of the F_p method for passive monitoring of F inside the exposure chamber. These six detectors have not been used for determining the relationship between the partial sensitivity and the removed active layer thickness.

Chemical etching, determination of the removed active layer thickness and the track densities were performed as described in Sections 3.2 and 3.3 above. From Section 3.3, the relationship between the partial sensitivity ρ_i (m) and the removed active layer thickness x (μm) has been determined as $\rho_i = (-0.0041 \pm 0.0004) + (0.0011 \pm 0.0001)x$, which is valid for the removed active layer thickness from 5.3 to 6.6 μm. The removed active layer thickness of these six LR 115 SSNTDs range from 5.4 to 6.5 μm, so the above relationship can be employed to determine the partial sensitivities. The results are shown in Table 1. It can be seen that all the determined ranges of F overlapped with the experimental value of 0.680 ± 0.068. In particular, the mid points of the ranges fall between 0.578 and 0.768, with a mean of 0.69. This shows that the mean of the mid points of the determined ranges of F for a few LR 115 SSNTDs can give a very good estimate of the true equilibrium factor.

Table 1
The removed layer, the partial sensitivities, the values of F_p and F determined for six LR 115 SSTNDs

Detector	Removed layer (μm)	Partial sensitivity (m)	F_p	F
1	5.418 ± 0.071	0.00206199	1.65	0.728–0.773
2	5.548 ± 0.083	0.00220968	1.40	0.534–0.622
3	5.600 ± 0.059	0.00226584	1.51	0.622–0.686
4	5.951 ± 0.122	0.00266462	1.63	0.713–0.761
5	6.095 ± 0.073	0.00282779	1.67	0.747–0.788
6	6.496 ± 0.102	0.00328036	1.55	0.649–0.711

The experimental value of F is 0.680 ± 0.068.

5. Conclusions and discussion

In the present paper, we have proposed and studied a method based on the proxy equilibrium factor F_p for passive monitoring of the equilibrium factor inside a radon exposure chamber. This method will not disturb the aerosol concentrations or the PAEC inside the exposure chamber and can give an integrated average value over the entire exposure period. This method might also be useful for exposure chambers for animals for epidemiological studies, for which the determination of radon progeny concentrations is also sometimes a challenge [15].

By exposing LR 115 detectors in a huge radon chamber at the Health Protection Agency (UK), which has a volume of 43 m^3, the relationship between the partial sensitivity ρ_i (m) and the removed active layer thickness x (μm) has been determined as $\rho_i = (-0.0041 \pm 0.0004) + (0.0011 \pm 0.0001)x$, which is valid for the removed active layer thickness from 5.3 to 6.6 μm. The method has also been verified by finding the equilibrium factor from track densities on the LR 115 detectors. In conclusion, the mean of the mid points of the determined ranges of F for a few LR 115 SSNTDs can give a very good estimate of the true equilibrium factor. If we combine the data for all fourteen LR 115 detectors, we can obtain the experimental relationship between the partial sensitivity and the removed active layer thickness as $\rho_i = (-0.0042 \pm 0.0013) + (0.0012 \pm 0.0002)x$, with the coefficient of determination $R^2 = 0.9836$. The coefficients are almost identical to those obtained from the eight SSNTDs.

From these relationships, we can see that the partial sensitivity increases sharply with the removed active layer thickness. In other words, the removed active layer thickness of the detector is critical for the calculation of F_p and hence F in real life experiments. Errors will occur from not controlling the removed active layer thickness accurately.

There are different possible error sources for F determined using the present method. From the equation $F_p = \rho/(\rho_i C_0 t) - 1$, we see that the percentage error of F_p is contributed by the percentage errors in the track density ρ, in the radon exposure $C_0 t$, and in the partial sensitivity ρ_i. The percentage error in the track density ρ mainly depends on the total number of tracks and is typically smaller than 3%, while that in the radon exposure $C_0 t$ depends on the radon gas concentration monitoring equipment and is typically smaller than 5%. The percentage error in the partial sensitivity ρ_i depends on our

regression results (Fig. 2) and is in general less than 4%. Taking these error sources all together, the error in F_p is typically smaller than 7%. This error will lead to an error of ~0.04 when F is equal to 0.1, ~0.1 when F is equal to 0.4 and 0.7.

The current method is designed for exposures only to ^{222}Rn and its progeny in radon chambers. In some cases, mixed exposures to ^{222}Rn, ^{220}Rn and their progeny are possible and needed. For such mixed exposures, the track density on the bare LR 115 SSNTD will be enhanced by the alpha particles from ^{220}Rn and its progeny. Such interference will be dealt with in a future study.

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