Retrospective radon progeny measurements through measurements of ^{210}Po activities on glass objects using stacked LR 115 detectors

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ABSTRACT

A stacked LR 115 detector consisting of two active layers was proposed for determining ^{210}Po activity in glass surfaces after deposition of short-lived radon progeny. The sensitivities of both active layers were calculated. Two glass samples were exposed in a chamber to determine the experimental calibration factors for the radon gas and progeny, which were then compared with the theoretical calibration factors from simulations. The experimental and the simulated calibration factors for radon progeny agreed well. The discrepancy between the calibration factors for radon gas was due to a much higher equilibrium factor used in the experimental calibration than the nominal value assumed in the simulation. A mini-survey of contemporary and retrospective radon progeny concentrations was carried out at 10 residential sites. A relationship between contemporary and retrospective radon progeny concentrations was not readily observable.

1. Introduction

It has been argued that case-control studies to estimate the risk of lung cancer due to radon exposure should preferably be based on long-term retrospective radon exposure assessments instead of contemporary radon exposure measurements [1–3] because of possible changes in exposures over time. Two methods of retrospective radon exposure assessment have been developed, namely, through surface traps (ST) and volume traps (VT). Both methods are based on determining the activity of ^{210}Pb, (a long-lived ^{222}Rn progeny with $T_{1/2}$ = 22.3 years) through measuring alpha particles emitted from its second successor ^{210}Po, either in ST such as the surface of solid media (mostly glass) [4–11] or in VT such as the bulk of porous media (mostly furniture filling sponges) [12].

In this paper, we only focus on the ST method, in particular using glass surfaces. Atoms of short-lived ^{222}Rn progeny, namely, ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po can accumulate and deposit on these surfaces. Due to the recoil after alpha decay of the ^{218}Po and ^{214}Po nuclides, some of the newly-formed atoms (^{214}Pb and ^{210}Pb) can be incorporated into the surface. The activity of the long-lived ^{210}Pb accumulated in a surface increases with the time of exposure. The ST method has been successfully applied in a number of previous surveys [3,13–16].

To determine the activity of ^{210}Po implanted in the surface of a glass object, the "(CR–LR) difference technique" [10,11] is commonly used. Here, two solid-state nuclear track detectors (SSNTDs), namely, LR 115 and CR-39, were fixed side by side on the glass object under study. Since the CR-39 detector does not have an upper energy threshold, it can detect alpha particles emitted from the surface layer (originating from implanted ^{210}Po) as well as from the volume of the glass object. The LR 115 detector has an upper energy threshold well below 5.3 MeV which is the energy of alpha particles emitted by ^{210}Po (except for very thick removed layers during chemical etching) and it detects only alpha particles emitted from the volume. Therefore, the difference between the track densities on these two detectors (after correction for different sensitivities) can be used for measurements of ^{210}Po implanted in the surface. The activity of ^{210}Po (Bq m^{-2}) is given in [10,11] as

$$A_{210_{Po}} = \frac{CR - B \times LR}{T \times K} \tag{1}$$

where CR is the net number of tracks per cm^2 on the CR-39 detector; LR is the net number of tracks per cm^2 on the LR 115 detector; B is the ratio of track densities recorded on a CR-39 detector to that on an LR 115 detector attached to a piece of unexposed glass; K is the sensitivity factor for the CR-39 detector to surface ^{210}Po activity (in the unit tracks cm^{-2} per Bq h m^{-2}) and T is the period time (h) for which the CR-39 and LR 115 detectors are mounted on the glass surface.

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A review of SSNTDs, including their use in retrospective radon dosimetry is given in [17]. The parameters B and K were determined in [18] for various removed layers from chemical etching for both the CR-39 and LR 115 detectors. Further studies have been carried out to relate the implanted ^{210}Po activity to the exposure to radon progeny [19,20], and a preliminary survey of retrospective radon progeny measurements for dwellings based on implanted ^{210}Po activities in glass objects has been carried out [21].

Although the (CR–LR) difference technique has become a default method for retrospective measurements of activities of ^{210}Po implanted in the surface of glass objects, some simplifications or improvements of the methodology are possible. First, the involvement of two different types of SSNTDs is inevitably more tedious and introduces extra uncertainties. For example, the etching conditions of the two types of SSNTDs are very different. Moreover, as explained in [18], for the (CR–LR) difference technique, the response of the CR-39 and LR 115 detectors to different alpha particle emitters in the volume of the glass objects, as well as the response of the CR-39 detectors to ^{210}Po on the surface of the glass objects should be determined, which relied on the availability of accurate V functions of these detectors. The function V is defined as the ratio between track etch rate V_t and the bulk etch rate V_b, i.e., $V = V_t/V_b$. It is noted that V functions are difficult to obtain and establish [22–25], and inevitably have uncertainties.

Secondly, for the (CR–LR) difference technique, the two SSNTDs are placed side by side on the glass surfaces and thus cover different positions. Although there were no detailed studies, the possibility of non-uniform background activities inside the glass objects cannot be ruled out. If such non-uniformity exists, applying separate detectors on two different positions on a glass object might lead to inaccurate results.

In the present work, a new method for determining the ^{210}Po activities in glass surfaces is proposed. Instead of requiring an LR 115 detector and a CR-39 detector to be fixed side by side on the examined glass object, we use only one stacked LR 115 detector mounted against a single location on the examined glass object. By using the LR 115 detector alone, only the uncertainty in the V function of this detector is relevant, which already reduces the total uncertainty of the method. Furthermore, now that only a single position of the glass surface is involved, the problem introduced by using two different positions is avoided.

2. Stacked LR 115 detectors and sensitivities of the active layers

The basic setup for a stacked LR 115 detector is shown in Fig. 1. Those used in the present studies (type 2, non-strippable) were purchased from DOSIRAD, France. They consist of a 12 μm red cellulose nitrate active layer and 100 μm clear polyester base substrate. In the present work, the stacked LR 115 detectors consist of two active layers. Active layer A is stripped from a piece of LR 115 detector with a thickness of 12 μm, and is placed in contact with the glass surfaces when in use. Active layer B is intact with its polyester base.

Active layer A registers only alpha particles emitted from the bulk glass volume. Due to the large energy of alpha particles emitted by ^{210}Po and small implantation depth (<0.1 μm), active layer A will not register any alpha particles emitted from the surface implanted ^{210}Po. Therefore, the response of active layer A can be written as

$$\rho_A = (\varepsilon_{A,V,U}C_U + \varepsilon_{A,V,T}C_T)t \tag{2}$$

where ρ_A is the track density recorded on the active layer A (in track/m^2), t is irradiation time (in s), $\varepsilon_{A,V,U}$ and $\varepsilon_{A,V,T}$ are the sensitivities of active layer A to alpha particles emitted in glass volume from the ^{238}U radioactive decay series (U) and the ^{232}Th decay ser-
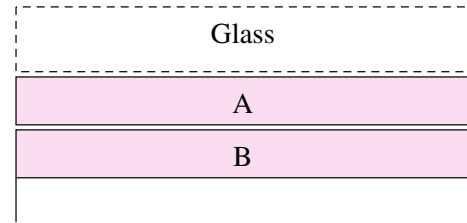

Fig. 1. Setup for a stacked LR 115 detector. Active layer A is stripped from a piece of LR 115 detector and will be placed in contact with the glass surfaces when in use. Active layer B is intact with its polyester base.

ies (T), respectively, both with the unit (track/m^2 s)/(Bq/kg), and C_U and C_T are the specific activities of ^{238}U and ^{232}Th in the glass (Bq/kg), respectively.

On the other hand, the active layer B registers alpha particles emitted from the bulk glass volume as well as alpha particles emitted from ^{210}Po implanted in the glass surface. In this way, the response of the active layer B is

$$\rho_B = (\varepsilon_{B,V,U}C_U + \varepsilon_{B,V,T}C_T + \varepsilon_{B,S,Po}A_{Po})t \tag{3}$$

where ρ_B is the track density recorded on the active layer B, $\varepsilon_{B,V,U}$ and $\varepsilon_{B,V,T}$ are the sensitivities of the active layer B to alpha particles emitted in glass volume from the ^{238}U decay series and the ^{232}Th decay series, respectively, and $\varepsilon_{B,S,Po}$ is the sensitivity of the active layer B to alpha particles emitted by ^{210}Po implanted into the surface, with the unit (tracks/m^{2}s)/(Bq/m^2), and A_{Po} is the surface activity of ^{210}Po (Bq/m^2).

Eqs. (2) and (3) have three unknown variables, namely, C_U, C_T and A_{Po}. Strictly speaking, it is not possible to solve this system without additional information. Fortunately, the problem can be simplified if we know the relationship between C_U and C_T. A number of different glass samples were measured using high-purity germanium spectrometry, and it was found that the ratio C_U:C_T was relatively constant at about 6:4. Therefore, if we write $C_{tot} = C_U + C_T$ and $C_U = kC_{tot}$, $k = C_U/(C_U + C_T) \approx 0.6$. With the notations $\varepsilon_A = k \times \varepsilon_{A,V,U} + (1 - k) \times \varepsilon_{A,V,T}$ and $\varepsilon_B = k \times \varepsilon_{B,V,U} + (1 - k) \times \varepsilon_{B,V,T}$, we can express the activity of ^{210}Po in the glass surface A_{Po} as

$$A_{Po} = \frac{\rho_B}{\varepsilon_{B,S,Po}t} - \varepsilon_B \frac{\rho_A}{\varepsilon_A \varepsilon_{B,S,Po}t} \tag{4}$$

To determine the ^{210}Po activity A_{Po}, two measurements are needed, i.e., the experimental track densities recorded on the active layers A and B, i.e., ρ_A and ρ_B, respectively.

3. Theoretical determination of sensitivities of the active layers

The coefficients ε_A, ε_B and $\varepsilon_{B,S,Po}$ in Eq. (4) are the sensitivities of the active layers of the LR 115 detectors, which can be derived if the V function is known. Here we use our recently obtained V function for the LR 115 detector:

$$V(R') = 1 + \left(A_1 e^{-A_2 R'} + A_3 e^{-A_4 R'}\right)\left(1 - e^{-R'}\right)$$

where A_1 = 14.23, A_2 = 0.48, A_3 = 5.9 and A_4 = 0.0773 [25], and R' is the residual range of the alpha particles. The sensitivities were then determined using Monte Carlo methods. The ranges of alpha particles in glass were determined using the SRIM program [26], with "glass soda lime" having a density of 2.3 g cm^3 as the target. The energy loss and alpha particle spectra needed for Monte Carlo calculations are presented in Figs. 2 and 3. The energy loss of alpha particles in 12 μm of the LR 115 detector active layer is shown. Alpha particles with energies up to 2.6 MeV are absorbed within the sensitive layer and lose all their energy – the linear part corresponds to this situation. These particles do not reach the detector