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Sensitivity of LR115 detector in diffusion chamber to ^{222}Rn in the presence of ^{220}Rn

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Abstract

Determination has been made of the sensitivity of LR115 type 2-track detectors (in units of m) to ^{222}Rn, measured in the presence of ^{220}Rn. Measurements have been made by means of a widely used diffusion chamber while Monte Carlo simulations have also been conducted. The experimentally derived sensitivities for ^{222}Rn and ^{220}Rn were found to be 0.470 ± 0.022 and 0.486 ± 0.042 m, respectively. For Monte Carlo simulations, the sensitivities to ^{222}Rn gas were found to range from 0.618×10^{-2} m (assuming that all ^{218}Po progeny decay before deposition onto the internal walls of the diffusion chamber) to 0.405×10^{-2} m (assuming that all ^{218}Po progeny are deposited on the internal walls of the same containment vessel before decaying). The sensitivity to ^{220}Rn gas of 0.465×10^{-2} m found from Monte Carlo simulations agrees to within uncertainty with experimental findings. The experimentally derived sensitivity value for ^{222}Rn indicates that 30% of the ^{218}Po progeny decay before deposition onto the internal walls of the diffusion chamber.

Keywords: Radon; Thoron; LR115 track detector; Detector sensitivity

1. Introduction

It is established that tracheobronchial deposition of ^{222}Rn (radon) progeny in the human body can lead to lung cancers, giving rise to considerable awareness of the ^{222}Rn problem. Various methods are available for measuring concentrations of ^{222}Rn, including use of the solid state nuclear track detector (SSNTD), LR115. In use of the cellulose nitrate material LR115, alpha particles emitted by ^{222}Rn and its progeny impinge upon the detector leaving latent tracks within it. The tracks can be made visible by chemical or electrochemical etching, the density of tracks being proportional to the average ^{222}Rn concentration during the period of exposure. The proportionality constant, denoted here by ε, is called the sensitivity of the detector.

An advantage of the LR115 detector, in comparison with one of the other commonly used SSNTDs, CR39, is that it does not detect the ^{222}Rn progeny which are deposited onto the detector itself, namely, the plate-out. However, the sensitivity of LR115 detector is 0.2–0.25 that of CR39. Only those alpha particles striking the surface with energies lower than a certain limit leave visible tracks and for LR115 the reduction in alpha energy required for this is more restrictive than for CR39. For most routine measurements, a detector in a diffusion chamber is used instead of a bare detector in order to minimize the influence of the unknown ratio between ^{222}Rn and its short-lived progeny on ^{222}Rn measurements. The method of using the detector in a diffusion chamber has been widely applied and well described in the literature (Frank and Benton, 1981; Khan et al., 1993; Durrani and Ilic, 1997). It is assumed that only gas will pass through the filter covering the diffusion chamber.

In recent years, increasing attention has been paid to the problem of ^{220}Rn. Research and surveys concerning

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the behavior of ^{220}Rn and its progeny have been carried out (Steinhäusler et al., 1994; Yu et al., 1999, 2000), showing that concentrations of ^{220}Rn and its progeny are not negligible compared to those of ^{222}Rn and its progeny. A particular concern is that alpha particles emitted from ^{220}Rn entering the diffusion chamber and progeny generated from ^{220}Rn within the diffusion chamber are detected by the LR115 detector, leading to incorrect results for measurements of ^{222}Rn concentrations (Nikezic and Yu, 2000). Since typically all recorded tracks are attributed to ^{222}Rn, measured ^{222}Rn concentrations are overestimated.

In the present paper, determination will be made of the sensitivity of LR115 detectors to ^{222}Rn in the presence of ^{220}Rn, in units of m, in a commonly used diffusion chamber. From this, the individual sensitivities to ^{222}Rn and ^{220}Rn will be determined. These sensitivities will also be determined through Monte Carlo simulations. The fraction of ^{218}Po progeny which decay before deposition onto the internal walls of the diffusion chamber will also be estimated.

2. Methodology

2.1. Experimental

The diffusion chambers employed for the present study were conical, with a base radius of 2.35 cm, a top radius of 3.35 cm and a height of 4.8 cm. This type of diffusion chamber is commonly used for routine measurements of radon concentrations, and it is for this reason that we have chosen this type of diffusion chamber for the study. The LR115 detectors were purchased from DOSIRAD, France (LR115 film, type 2, non-strippable). The detectors consist of 12 μm red cellulose nitrate on a 100 μm clear polyester base. Circular detectors with a radius of 2.35 cm were cut from the films, and fitted to the bottom of the diffusion chambers for exposure.

A specially designed ^{222}Rn/^{220}Rn exposure chamber capable of alpha spectrometry was employed for exposure of the LR115 detectors to an environment of known mixed amounts of ^{222}Rn and ^{220}Rn (Yu et al., 2002). A block diagram of the ^{222}Rn/^{220}Rn exposure chamber system is shown in Fig. 1. The ^{222}Rn source is ^{226}Ra while the ^{220}Rn source is ^{228}Th (both purchased from Isotope Products Laboratories, California). Both sources had an activity of 10 μCi (370 kBq) at the time of exposure. The gas from each source was driven by an individual diaphragm pump (GAST® DOA-P101-BN, from Gast Manufacturing, Inc., Michigan) through a membrane filter (pore size of 0.45 μm) into the exposure chamber. In the present investigation, the exposure chamber was operated in a recirculation mode, the output air from the exposure chamber being recirculated

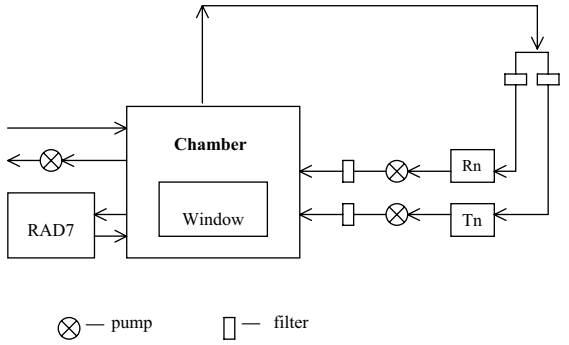

Fig. 1. Block diagram of the ^{222}Rn/^{220}Rn exposure chamber used in the present investigations.

through the ^{222}Rn and/or ^{220}Rn sources. The free air flow rate of each diaphragm pump was 1.6 m^3 h^{-1}. The ^{222}Rn and ^{220}Rn sources can be operated independently or simultaneously.

The instrument RAD7 (from Durridge Company Inc., MA) forms the basis of a comprehensive ^{222}Rn and ^{220}Rn monitor for our exposure chamber. The RAD7 spectrum provides a scale of alpha energies, converting the energy range from 0 to 10 MeV divided into 200 channels each of 50 keV width. The alpha energies associated with ^{222}Rn and ^{220}Rn reside within the range 6 to 9 MeV. These 200 channels are grouped into eight energy windows (labeled A to H). Windows A and B were used for our investigations. Window A which records 6.00 MeV alpha particles from ^{218}Po, the half-life of which is 3 min, is the window used for the ^{222}Rn sniff mode. Window B which records 6.78 MeV alpha particles from ^{216}Po, the half-life of which is 0.15 s, is the window used for the ^{220}Rn sniff mode. RAD7 allows different modes of measurement, including a normal mode and the sniff mode. For present purposes the sniff mode was employed. In the sniff mode, RAD7 uses ^{218}Po signals to determine ^{222}Rn concentrations and ^{216}Po signals to determine ^{220}Rn concentrations, ignoring subsequent and long-lived progeny.

Our RAD7 instrument was calibrated by the manufacturer as well as by ourselves. Cycle times of 10 min were adopted, with RAD7 providing a print out of the ^{220}Rn value as well as the ^{222}Rn value at the end of every cycle. The RAD7 includes an RS232 serial port which can be used for transfer of data into a file on a personal computer. The computer software (Capture 1.2.0) was provided by the manufacturer for this purpose. To evaluate the exposure conditions (in units of Bq h m^{-3}), a small, simple program has been written to perform time integration of ^{222}Rn/^{220}Rn concentrations.

For an unknown presence of ^{220}Rn, all tracks recorded by the LR115 detector are attributed to ^{222}Rn. We denote the ^{222}Rn sensitivity derived from

this process as the apparent ^{222}Rn sensitivity. In the present study, apparent ^{222}Rn sensitivities were determined for nine different ratios of ^{222}Rn to ^{220}Rn exposures, exposures being recorded in units of Bq h m^{-3}, and ratios ranging from ~100% (pure ^{222}Rn) to ~0%. The etching conditions were chosen to be a 2.5 N aqueous solution of NaOH at a temperature of 60°C, with an etching duration of 120 min, corresponding to a removed layer of about 6.7 μm of cellulose nitrate. This value was also adopted in the computer simulations described in Section 2.2.

2.2. Computer simulations

The methodology for calculating the theoretical sensitivity of the LR115 detector to the ^{222}Rn and ^{220}Rn chains has been presented elsewhere by Nikezic and Yu (2000), embodied in a computer program developed by Nikezic and Baixeras (1995). The program used a Monte Carlo method to simulate the propagation of alpha particles, and the Bethe–Bloch expression to calculate the stopping power of alpha particles in air and cellulose nitrate. The program also incorporates visibility criterion for the tracks. The sensitivities are lower for nuclides of larger alpha energy, the upper detection threshold for LR115 type 2 being in the interval of 4.1–4.6 MeV. This energy threshold depends on the thickness of the layer removed during etching (Nikezic and Baixeras 1996).

One of the major issues in calculation concerns the different irradiation geometries for alpha particles coming from progeny in the air within the diffusion chamber and from progeny deposited on the internal walls of the diffusion chamber.

For the ^{222}Rn chain, uncertainty exists regarding the fraction of ^{218}Po that decay in air within the diffusion chamber prior to deposition. Since ^{218}Po has a comparatively short half-life, this emitter will not be fully deposited before decaying. Conversely, ^{214}Bi has a long half-life and can be assumed to be completely deposited before decaying. For the ^{220}Rn chain, it is reasonable to assume that ^{216}Po completely decays in the air within the diffusion chamber before decaying, this being due to its very short half-life. As a result of the long half-life of ^{212}Po (10.64 h), the second and third ^{220}Rn generation progeny are completely deposited on the internal walls of the diffusion chamber before decaying. In this way, there is no uncertainty discernible in the ^{220}Rn chain detection due to unknown deposition fractions of its progeny.

3. Results and discussion

The results for the apparent ^{222}Rn sensitivities (in m) for nine different ratios (X) of ^{222}Rn and ^{220}Rn exposure

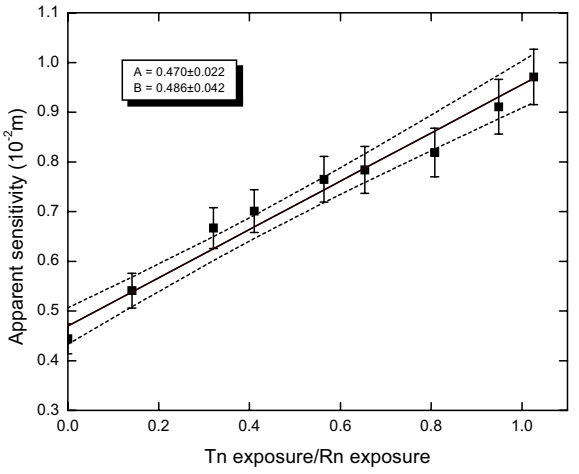

Fig. 2. The experimentally determined apparent sensitivity of LR115 detectors (in m), in a diffusion chamber, to levels of ^{222}Rn expressed as ratios of integrated exposure (in Bq h m^{-3}) to ^{222}Rn and ^{220}Rn. A linear relationship has been obtained between the apparent sensitivity and the ratio. The best fit to the data yields an intercept value of 0.470 ± 0.022 and a slope of 0.486 ± 0.042 m, respectively.

are shown in Fig. 2. A linear relationship has been obtained between the apparent sensitivity and values of the ratio, a situation which, as shown below, is to be expected. By definition, the apparent ^{222}Rn sensitivity ε^*, is given by

$$\varepsilon^* = [N(\mathrm{Rn}) + N(\mathrm{Tn})]/[A \times E(\mathrm{Rn})], \quad (1)$$

where N(Rn) and N(Tn) are the numbers of alpha tracks recorded on the LR115 detector that are attributable to ^{222}Rn and ^{220}Rn, respectively, E(Rn) is the integrated exposure of the detector to ^{222}Rn and A is the area of the detector. Theoretically, $N(\mathrm{Rn}) = \varepsilon(\mathrm{Rn})E(\mathrm{Rn})A$, and $N(\mathrm{Tn}) = \varepsilon(\mathrm{Tn})E(\mathrm{Tn})A$, where ε(Rn) and ε(Tn) are the true ^{222}Rn and ^{220}Rn sensitivities, respectively, and E(Tn) is the integrated exposure of the detector to ^{220}Rn. In this way, Eq. (1) becomes the linear relationship

$$\varepsilon^* = \varepsilon(\mathrm{Rn}) + \varepsilon(\mathrm{Tn}) \times X \quad (2)$$

with the intercept being the true ^{222}Rn sensitivity ε(Rn) and the slope the true ^{220}Rn sensitivity ε(Tn). From the best linear fit to the experimental data, $\varepsilon(\mathrm{Rn}) = (0.470 \pm 0.022) \times 10^{-2}$ m and $\varepsilon(\mathrm{Tn}) = (0.486 \pm 0.042) \times 10^{-2}$ m.

Monte Carlo simulations indicate that the sensitivity to ^{222}Rn gas ranges from 0.618×10^{-2} m (if all ^{218}Po progeny decay before deposition onto the internal walls of the diffusion chamber) to 0.405×10^{-2} m (if all ^{218}Po progeny are deposited before decaying), and that the sensitivity to ^{220}Rn gas is 0.465×10^{-2} m, agreeing to within uncertainty with the experimental results. The

experimentally derived value for ^{222}Rn indicates that 30% of the ^{218}Po progeny decay before deposition onto the internal walls of the diffusion chamber.

In normal indoor environments, ^{220}Rn can achieve levels of 10% of ^{222}Rn, or $E(\mathrm{Tn})/E(\mathrm{Rn}) = 0.1$. It follows that the relative error in normal indoor environments can be as much as $(0.486 \times 0.1)/0.470$ or about 10%. Cases of high indoor ^{220}Rn concentrations have previously been reported by Yu et al. (1999, 2000). In addition, if the source of ^{220}Rn is the wall material of the buildings, the ^{220}Rn concentration will decrease exponentially with distance from the wall (Katase et al., 1988). Under such circumstances, ^{222}Rn measurements performed at different distances from the wall will give different values. Therefore, ^{220}Rn in air will lead to uncertainty in ^{222}Rn measurements that are made using the LR115 detector. Corrections for this should be taken care of, particularly for environments with potentially high concentrations of ^{220}Rn.

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