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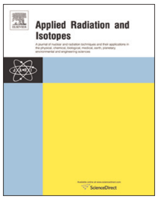

A comparative study between the dynamic method and passive can technique of radon exhalation measurements from samples

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HIGHLIGHTS

- Comparative study has been carried out between active and SSNTD based ‘can’ techniques.
- Study showed large difference in measured radon and thoron exhalation rates.
- Study unequivocally demonstrates the presence of intrinsic uncertainty in SSNTD based ‘can’ technique.
- The uncertainty has been analyzed through thoron interference and leak effect.

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ABSTRACT

A comparative study has been carried out between the SSNTD based ‘can’ technique and active monitors based dynamic method using nine different samples, eight of granite and one of phosphogypsum. Besides radon (^{222}Rn) exhalation, thoron(^{220}Rn) exhalation and ^{226}Ra and ^{232}Th content were also measured. The results are: (i) presence of significant thoron exhalation from samples and (ii) observation of thoron interference and leak ($\sim 0.05\ h^{-1}$) from the ‘can’ in the SSNTD based ‘can’ technqiue. The study unequivocally demonstrates the presence of intrinsic uncertainty in SSNTD based ‘can’ technique. Instead, dynamic method offers a more reliable and faster method.

1. Introduction

Measurement of radon (^{222}Rn) exhalation from samples of materials used in the construction of dwellings is important for assessing the sources contributing to indoor radon risks to human populations. Primarily, it provides information on the relative radon hazard potential of different building materials for the purpose of advising on their suitability for use prior to construction of a building. In addition, radon exhalation data generated for various samples are useful for validating diffusion based models for predicting radon emission flux from the walls and concentration in dwellings (Nazaroff and Nero, 1988; Sahoo et al., 2011). To a considerable measure, a resurgence of widespread interest on this topic follows from the relatively recent epidemiological findings of indoor radon risks (Darby et al., 2005; Krewski et al., 2005; Lubin et al., 2004) followed by its official recognition in a report by World Health Organization (WHO, 2009). A large number of publications on radon exhalation from building materials have appeared in journals in the past few years and the interest in exhalation measurements is likely to increase in future.

A simple and most widely used technique for radon exhalation measurements is the sealed ‘can’ technique using Solid State Nuclear Track Detectors (SSNTDs) (Abu-Jarad et al., 1980; Fleischer and Mogro-campero, 1978; Abu-Jarad, 1988). In this method, concentration of radon emitted from a sample placed inside a closed can is monitored in a time integrated manner by the SSNTDs, which is then converted to exhalation rates by the use of

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analytical formulae. Many previous studies (Khan et al., 1992; Singh et al., 1997; Kumar et al., 2003, 2005, 2008; Mahur et al., 2010; Gupta et al., 2010; Saad et al.,2013; Sharma and Virk, 2001) which used the can technique for radon exhalation measurements did not consider the possibility that the SSNTDs might also detect ^{220}Rn (thoron) atoms exhaled from the samples which would then be erroneuously interpreted as ^{222}Rn. In fact, in a recent study, Sahoo et al. (2014,) showed that SSNTDs placed at a distance of about 7 cm from the thoron source in a sealed 'can' do register tracks due to ^{220}Rn, thereby countering the generally held notion that thoron concentration will decay within a diffusion length of about 3 cm. Since most building materials may also contain significant amounts of ^{232}Th along with ^{226}Ra, one cannot rule out the possibility of interference due to thoron in radon measurements in SSNTD based 'can' techniques. Another important issue that has escaped attention is related to the effect of tiny leaks from the 'can'. Since the 'cans' are closed with gaskets (not hermetically sealed), it cannot guarantee zero leakage of the radon during the course of its build-up over a period of 90 days in the can. The presence of a small leak, say even as low as 0.01 h^{-1}, will not allow ^{222}Rn to build up to its full equilibrium value dictated by its radioactive decay constant (λ_R=0.00765 h^{-1}). As a consequesnce, the radioactive decay constant term occurring in the formula of Abu-Jarad (1988; Eq. 4 in this paper), which is generally used for converting the observed concentration to exhalation rates, needs to be replaced by an effective decay constant term, $\lambda_e = \lambda_R + \lambda_{leak}$. No previous study has examined the influence of leaks in sealed-can technique as a potential source of underestimation in radon exhalation measurements. Taken together, the twin effects of thoron interference and leak, will act in opposite direction in an unpredictable manner introducing uncertainties in the reported values of ^{222}Rn exhalation rates.

An alternative method of estimating radon exhalation from samples is based on the use of continuous radon monitors, rather than passive SSNTDs for measuring the radon concentration inside the can (Petropoulos et al., 2001; Sahoo et al., 2007; Chen et al., 2010). We refer to this as dynamic method in which the radon monitor, placed in a recirculating closed loop connected to the can and the pump, monitors the radon concentration at regular intervals. Since these instruments are designed to cut-off thoron response, thoron interference is automatically annulled in this technique. By analyzing the rate of rise of the concentration, this method makes it possible to estimate both the exhalation rate from the sample and the leak rate from the chamber, if any, in the apparently sealed system. The method also allows the measurement of thoron exhalation rates from samples by monitoring thoron concentration using RAD7 radon/thoron monitors (Durridge, USA). Another distinct advantage of the dynamic method is that it is possible to analyse samples within 1–2 days as compared to about 90 days using passive methods.

In this paper, we subject the SSNTD based passive, 'can' technique to experimental comparsion tests vis-à-vis the online monitor based dynamic method. We perform a series of experiments using 9 different samples of building materials. The experiments consist of estimating radon and thoron surface exhalation rates using the dynamic method for each sample and also estimating the apparent radon exhalation rate using the conventional 'can' technique. As additional support, the activity concentrations of ^{226}Ra and ^{232}Th were also estimated in these samples by gamma spectrometry. The data were subjected to statistical analysis to examine correlations between various quantities consistent with theoretical reasoning. The study unequivocally points out the influence of significant thoron emission from samples as well as the presence of leaks on the radon exhalation results interpreted from the 'can' technique.

2. Materials and methods

Radon and thoron exhalation rates were measured from 9 different samples of building materials collected from the state of Tamil Nadu, India. Eight of the samples were Indian granite while one sample was phosphogypsum sample. Commercial trade names of the samples along with sample IDs presented in Table 1. These samples were in the form of cuboidal blocks and hence, the radon exhalation from these samples were expressed in terms of radon activity released per unit surface area per unit time i.e., surface exhalation rate of radon. The exhalation rates were measured by two independent methods, viz., (i) active, dynamic method utilizing a continuous radon monitor and (ii) passive, 'can' technique based on Solid State Nuclear Track Detectors (SSNTDs). The measurement procedure followed in dynamic and the passive methods are briefly described below.

2.1. Dynamic method

2.1.1. ^{222}Rn exhalation measurements

Dynamic method is an active method which relates the rate of increase of radon concentration in a closed accumulation chamber to the radon exhalation rate of the sample enclosed in the chamber (Petropoulos et al., 2001; Sahoo et al., 2007; Chen et al., 2010). This is accomplished by monitoring the changes in radon concentrations at regular intervals using an active instrument, which is designed to exclude thoron interference. The photograph of the measurement set-up used for this study is given in Fig. 1. In this method, few blocks of building material samples were kept in an accumulation chamber, having a volume capacity of about 1.33 L. The chamber was sealed with a gasket at its mouth. The chamber was connected to a Continuous Radon Monitor, SRM (Gaware et al., 2011) in a closed loop through tubes and a pump system for re-circulation of air between accumulation chamber and detection chamber of SRM. The SRM radon monitor detects the alpha particles emitted from ^{222}Rn and its decay products (^{218}Po and ^{214}Po) in a ZnS(Ag) scintillation chamber. The gross scintillation counts obtained in a measurement cycle (e.g 15 min, 60 min etc) are converted to radon activity concentration (Bq m^{-3}) at the end of the each cycle by a built-in Micro-Controller based Smart Algorithm. Since the technology is purely based on direct scintillation counting of alpha particle with build-up/decay corrections achieved through software, the measurements are not affected by humidity or trace gases, unlike the case of electro-static collection based radon measurement systems. Hence, this monitor can be operated without use of additional drier system. This monitor is equipped with a pinhole based diffusion barrier to cutoff thoron

Table 1.
Measured ^{226}Ra and ^{232}Th activity concentrations in analyzed samples.

Sample name	Sample ID	^{226}Ra concentration (Bq Kg^{-1})	^{232}Th concentration (Bq Kg^{-1})
New imperial red granite	S1	150 ± 9	244 ± 8
Chocolate brown granite	S2	184 ± 10	246 ± 8
Bellary grey granite	S3	77 ± 10	193 ± 10
Bon red granite	S4	61 ± 8	196 ± 8
Imperial white granite	S5	58 ± 7	96 ± 6
Sentinel red granite	S6	25 ± 5	62 ± 5
Phosphogypsum	S7	160 ± 21	BDL
Imperial gold granite	S8	BDL	–
Oyster pearl granite	S9	BDL	133 ± 18

BDL: Below Detection Level.