Available online at www.sciencedirect.com
ScienceDirect
Nuclear Instruments and Methods in Physics Research B 263 (2007) 311–316
NIM B Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

A study of the polyethylene membrane used in diffusion chambers for radon gas concentration measurements

S.Y.Y. Leung [a], D. Nikezic [a,b], J.K.C. Leung [c], K.N. Yu [a,*]

[a] *Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon Tong, Hong Kong*
[b] *Faculty of Sciences, University of Kragujevac, Serbia*
[c] *Department of Physics, The University of Hong Kong, Pokfulam Road, Hong Kong*

Available online 14 April 2007

Abstract

Solid-state nuclear track detectors (SSNTDs) in diffusion chambers have been routinely used for long-term measurements of radon gas concentrations. In usual practice, a filter is added across the top of the diffusion chamber to stop the progeny from entering. Thoron can also be deterred from entering the diffusion chamber by using a polyethylene (PE) membrane. However, the thickness of the PE membrane is rarely specified in the literature. In this paper, we will present our experimental results for a radon exposure that the number of alpha-particle tracks registered by the LR 115 SSNTD in a Karlsruhe diffusion chamber covered with one layer of PE membrane is actually enhanced. This is explained by enhanced deposition of radon progeny on the outside surface of the PE membrane and the insufficient thickness of the PE membrane to stop the alpha particles emitted from these deposited radon progeny to reach the SSNTD. We will present the PE thickness which can stop the alpha particles emitted from the deposited radon or thoron progeny. For the "twin diffusion chambers method", one of the diffusion chambers is covered with PE membranes. The optimal number of thickness of PE membranes will be determined, which allows the largest amount of radon gas to diffuse into the diffusion chamber while at the same time screening out the largest amount of thoron gas.

PACS: 29.40; 23.60

Keywords: Diffusion chamber; Polyethylene membrane; Radon and thoron gas concentration

1. Introduction

The alpha track method is one of the most common methods employed for long-term measurements of radon (^{222}Rn) or thoron (^{220}Rn) gas concentrations. In this method, a diffusion chamber is employed in such a way that only radon and thoron gas, but not the progeny, can diffuse inside. A piece of solid-state nuclear track detector (SSNTD) is placed on the inner bottom of the diffusion chamber. The radon and thoron gas inside the diffusion chamber, as well as their progeny formed in the diffusion chamber, can emit alpha particles that strike and leave latent tracks in the SSNTD. On chemical etching, the tracks can be visualized and counted under an optical microscope with a suitable magnification. A review of different designs of the diffusion chamber can be found in [1] and large scale of surveys using different diffusion chambers have been carried out [2,3]. A recent review on SSNTDs can be found in [4].

When both radon and thoron gas are present in air, methods have to be devised to separate their signals obtained using the alpha track method. It has almost become a routine practice to use different covers for the diffusion chamber in order to get the different signal components. When the combined signal from radon and thoron is required, a filter paper is used as the cover. This setup can also be used alone for radon measurements if the thoron concentration is negligible. In general, if mainly the signal

* Corresponding author. Tel.: +852 27887812; fax: +852 27887830.
E-mail address: peter.yu@cityu.edu.hk (K.N. Yu).

0168-583X/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2007.04.120

from radon is wanted, a plastic membrane will be used as the cover, which acts as a barrier to delay the diffusion of short-lived thoron gas (half life of 55 s) into the diffusion chamber. In particular, polyethylene (PE) has been found a very appropriate anti-thoron filter [5] and has been used widely. Simultaneous measurements of radon and thoron gas concentrations can then be made using the so-called "twin diffusion chamber method" [6–8], with one diffusion chamber covered with filter paper (to record signals from radon + thoron) and the other covered with PE membrane (to mainly record the signals from radon). Despite the common use of PE as the cover, detailed studies on the PE thickness and precautions are not available in the literature.

In the present work, we will first present some interesting experimental results showing that, for radon exposures alone, the radon concentrations deduced from the alpha track method is significantly overestimated if the diffusion chamber is covered only by one piece of PE membrane, and will be correct if the diffusion chamber is covered by more than one piece of PE membrane or just a filter paper. The underlying reason will be explored. The present paper will show that the enhancement is due to the alpha particles emitted from deposited radon progeny on the outside surface of the PE membrane. A suggested solution is to increase the number or thickness of the PE membranes. The optimal number or thickness of PE membranes will be determined, which will stop all the alpha particles from deposited radon or thoron progeny on the outside surface of the PE membrane from reaching the SSNTD on the bottom of the diffusion chamber, while at the same time allowing the largest amount of radon to get inside and screening out the largest amount of thoron.

2. Methodology

2.1. Diffusion chamber and exposure chamber

The diffusion chamber used in the present studies is known as "Karlsruhe diffusion chamber", which is conical with a base radius of 2.35 cm, a top radius of 3.35 cm and a height of 4.8 cm (see Fig. 1). There are holes in the brim of the diffusion chamber, which let air flow in and out of the chamber.

An exposure chamber [9] is used to expose the diffusion chambers with SSNTDs inside. The radon and/or thoron concentrations inside the exposure chamber were measured using RAD-7 (Durridge Company Inc., MA) with a sampling period of 30 min (see Fig. 2), which can measure radon and thoron gas concentrations individually and simultaneously. Diffusion chambers are transferred into the exposure chamber through an air lock.

2.2. Etching LR 115 detector and counting alpha-particles tracks

The LR 115 detectors (Type 2, non-strippable) are purchased from DOSIRAD, France, and consist of a 12 μm red cellulose nitrate active layer on top of a 100 μm clear polyester base. After exposure, the LR 115 detector was removed from the diffusion chamber and 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. The detectors were etched using a magnetic stirrer (Model No: SP72220-26, Barnstead/Thermolyne, Iowa, USA) to provide more uniform etching [10]. After etching for 1 h, the detector was removed from the etchant, rinsed with de-ionized water and dried.

The alpha-particle tracks registered by the LR 115 detector were counted under an optical microscope with 200× magnification and only those tracks completely perforated the active layer were counted. The track densities (number of tracks per unit area) were then found, from which the radon or thoron gas concentrations could be calculated through the sensitivities. The sensitivities (track density per unit exposure, in m) for radon or thoron are dependent on the removed active layer thickness of the LR 115 detector [11]. The removed active layer of the LR 115 detector was determined using the 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 [12]. The infrared transmittance was measured using a Perkin–Elmer Fourier Transform Infrared (FTIR) spectroscopy system (Model 16 PC FT-IR). The detectors were scanned for 10 cycles,

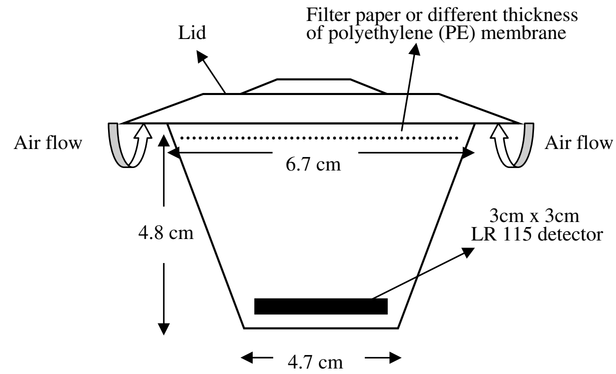

Fig. 1. The dimensions of the Karlsruhe diffusion chamber used in the present investigation with a top radius of 3.35 cm, base radius of 2.35 cm and a height of 4.8 cm.

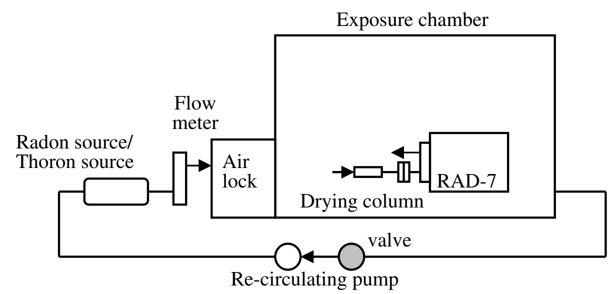

Fig. 2. The block diagram of the experimental set-up for the exposure chamber.

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 $4\ cm^{-1}$.

2.3. Experiment showing overestimation of radon concentration when the diffusion chamber is covered by one piece of PE membrane

In this section, we will compare the performance of LR 115 SSNTDs inside Karlsruhe diffusion chambers covered by (A) 1 filter paper, (B) 1 PE membrane and (C) 2 PE membranes. These diffusion chambers are transferred to an exposure chamber filled with radon gas only. The reference radon gas concentrations (RC) have been measured by the continuous radiation monitor AB-5 (PYLON, Canada). After the radon exposure, the track densities on the LR 115 detectors and the derived radon concentrations will be compared.

In order to explain the different results, we need two pieces of information. The first one is the thickness of the PE membrane employed for the present experiment and the second one is the residual range of alpha particles in air after passing through different number of PE membranes.

The thickness of the PE membrane can be revealed by the energy loss when alpha particles with a particular incident energy pass through the PE membrane. A planar ^{241}Am source (with the main alpha energy 5.49 MeV under vacuum, activity 3.34 kBq and active diameter of 5 mm) is employed to provide the incident alpha particles. The residual alpha energy was measured using an alpha spectroscopy system (ORTEC Model 5030) under vacuum. The alpha-particle detector was a passivated implanted planar silicon (PIPS) detector with an area of $300\ mm^2$. From the energy loss through the PE membrane, the thickness can be deduced if the range–energy relationship for alpha particles in PE is established. This is obtained from the stopping and range of ions in matter (SRIM) program (SRIM-2003 [13]). A full description of the calculations can be found in [14]. Similarly, the residual range of alpha particles in air after passing through PE membranes can be determined from the range–energy relationship for alpha particles in air, which can also be obtained using SRIM-2003 [13].

2.4. Optimal number or thickness of PE membranes for the "twin diffusion chambers method"

A solution to prevent enhancement of alpha track density on the SSNTD in the diffusion chamber due to alpha particles emitted from deposited radon/thoron progeny on the outside surface of the PE membrane is to increase the number or thickness of the PE membranes. As different from the experiments in Section 2.3, for a general consideration here, we have to consider both the deposited radon and thoron progeny on the outside surface of the PE membrane. From the thickness of the PE membrane determined as described in Section 2.3, and through calculations using the SRIM program, the number of PE membranes required for covering the diffusion chamber in order to prevent enhancement of alpha-particle track density due to deposition of radon or thoron progeny is determined.

For the "twin diffusion chambers method", one diffusion chamber is covered with filter paper to allow both radon and thoron to diffuse inside, while the other diffusion chamber is covered with PE membranes. Apart from preventing enhancement of alpha-particle track density due to deposition of radon or thoron progeny on the outside surface of PE membranes, the design should allow the largest amount of radon gas to diffuse into the diffusion chamber while at the same time screening out the largest amount of thoron gas. It is expected that the obtained track density will decrease with increasing PE thickness due to delay of diffusion of both radon and thoron into the chamber. The optimal number or thickness of PE membranes will be determined.

Finally, with this optimal number or thickness of PE membranes, the radon and thoron gas concentrations experimentally obtained by the "twin diffusion chambers method" are compared with the reference values.

3. Results and discussion

3.1. Experiment showing overestimation of radon concentration when the diffusion chamber is covered by one piece of PE membrane

Karlsruhe diffusion chambers covered by (A) 1 filter paper, (B) 1 PE membrane and (C) 2 PE membranes were exposed to radon gas only in the exposure chamber. The results are shown in Table 1. The numbers of alpha-particle tracks registered by the LR 115 SSNTD in the diffusion chamber for one PE membrane were actually enhanced in all the three exposures, while those for two PE membranes were similar to those for one filter.

This phenomenon can be explained by the enhanced deposition of radon progeny on the outside surface of the PE membrane (probably by static electricity on the membrane) and the insufficient thickness of a single PE membrane to stop the alpha particles emitted from these deposited radon progeny to reach the SSNTD. This conjecture can be supported if we can prove that the most energetic alpha particles from radon progeny can be totally stopped by two PE membranes and not by a single PE membrane.

Before going for this proof, the thickness of a PE membrane layer has to be known. As mentioned, this will be determined using alpha spectroscopy results together with SRIM calculations. The energies of alpha particles from ^{241}Am after passing through 1–3 PE membranes are shown in Table 2. The residual ranges in PE for these alpha energies have been determined from the range–energy relationship obtained from SRIM. These are also shown in Table 2. From these, the average thickness of a PE membrane

Table 1
Comparisons of track densities on the LR 115 SSNTDs in Karlsruhe diffusion chambers covered with (A) 1 filter paper, (B) 1 PE membrane and (C) 2 PE membranes

Exposure	Cover	Track density (10^6 m^{-2})	Removed active layer (μm)	Derived RC (Bqm^{-3})	Reference RC (Bqm^{-3})
1	A	1.589 ± 0.042	5.865 ± 0.108	1557 ± 161	1490 ± 100
	B	1.982 ± 0.047	5.956 ± 0.106	1874 ± 193	
	C	1.632 ± 0.043	5.946 ± 0.125	1549 ± 160	
2	A	1.688 ± 0.043	6.028 ± 0.065	1551 ± 160	1510 ± 100
	B	1.914 ± 0.046	5.994 ± 0.129	1784 ± 184	
	C	1.666 ± 0.043	6.061 ± 0.105	1509 ± 156	
3	A	1.592 ± 0.042	5.706 ± 0.124	1667 ± 173	1610 ± 110
	B	2.047 ± 0.048	5.815 ± 0.104	2047 ± 210	
	C	1.574 ± 0.042	5.690 ± 0.075	1660 ± 172	

The diffusion chambers are exposed to radon only. The removed active layer of the LR 115 SSNTDs measured by FTIR and the derived radon concentrations (RCs) are also given. Reference RC values are measured by the continuous radiation monitor AB-5.

Table 2
The experimentally measured peak energies of alpha particles (with incident energy of 5.5 MeV) after passing through different numbers of PE membrane layers under vacuum and the corresponding ranges in PE of the residual alpha energy (calculated using SRIM), and the derived thickness of the PE membrane

PE layers	Peak alpha energy (MeV)	Range in PE of the residual alpha energy (μm)	Average thickness of a PE layer (μm)
No PE	5.50	39.75	–
1 layer of PE	4.40	28.26	11.49
2 layers of PE	3.56	20.70	9.53
3 layers of PE	1.86	8.91	10.28

Table 3
Energy of alpha particles reaching the LR 115 detector in the diffusion chamber covered with different PE layers (calculated using SRIM)

Incident alpha energy	7.69 MeV		8.78 MeV		
Number of PE layers	1	2	2	3	4
Exiting alpha energy (E) from the PE layer(s) (MeV)	6.99	6.22	7.46	6.74	5.95
Range in air for exiting alpha energy E (mm)	56.37	46.93	62.61	53.21	43.75
Alpha energy incident on LR 115 detector (MeV)	1.72	–	2.76	1.04	–

is determined as about 10 μm, which will be used in subsequent calculations.

With the ranges of alpha particles in air and in PE generated by SRIM, and knowing that the distance between the PE membrane and the SSNTD is 48 mm, the minimum PE thickness required to completely stop the alpha particles emitted from the deposited radon progeny on the outside surface of the PE membrane to reach the SSNTD has been calculated as 13.52 μm. In other words, two PE membranes can completely stop all the alpha particles while a single PE membrane cannot, which agrees with the experimental findings. The agreement shows that the enhancement of alpha track density on the SSNTD in a diffusion chamber covered by one piece of PE membrane for a radon exposure is due to the alpha particles emitted from deposited radon progeny on the outside surface of the PE membrane.

3.2. Optimal number or thickness of PE membranes

Table 3 lists the energy of alpha particles (incident energies of 7.69 MeV for the radon chain and 8.78 MeV for the thoron chain) reaching the LR 115 detector after passing through different number of PE membranes, which are calculated using SRIM. For the incident alpha energy of 7.69 MeV, the alpha energy exiting from 2 PE membranes is 6.22 MeV which corresponds to a range of 46.93 mm in air. On the other hand, for the incident alpha energy of 8.78 MeV, the alpha energy exiting from 4 PE membranes is 5.95 MeV which corresponds to a range of 43.75 mm in air. Under both conditions, the ranges in air are smaller than the height of the diffusion chamber of 48 mm, so these alpha particles will not be registered by the LR 115 detector. For other PE membrane setups, i.e. 1 PE membrane for the case of radon exposure and 2 or 3 PE membranes for the case of thoron exposure, the alpha particles emitted by the progeny deposited on the outside surface of the PE membrane reaching the LR 115 SSNTD are all energetic enough to be registered. Under such circumstances, enhancement of alpha-particle track density will be resulted due to the enhanced deposition of radon or thoron progeny.

From the above discussion, we conclude that using 4 PE membranes will stop alpha particles from the deposited radon or thoron progeny from being registered. Therefore, we start with using 4 PE membranes to see which thickness (or equivalently the optimal number of PE membranes) can allow the maximum amount of radon gas to enter the diffusion chamber and at the same time screen out the largest amount of thoron gas.

For both 4 and 5 PE membranes, five measurements with different removed active layers were made. Table 4 shows the percentage of radon and thoron gas that can

Table 4
Percentage of radon and thoron passing through 4 and 5 PE membranes determined for different active layer thickness of the LR 115 detector, the latter being measured by FTIR

Layers of PE membranes	% of radon getting into the diffusion chamber and the active layer thickness	% of thoron getting into the diffusion chamber and the active layer thickness
4	97.9 (5.62 μm)	8.26 (5.35 μm)
	100.7 (5.72 μm)	8.12 (5.47 μm)
	100.7 (6.06 μm)	7.81 (5.52 μm)
	100.4 (6.07 μm)	7.59 (5.75 μm)
	92.4 (6.70 μm)	7.23 (6.12 μm)
Average	98.4 ± 3.5	7.80 ± 0.41
5	81.7 (5.77 μm)	7.75 (5.42 μm)
	89.3 (5.80 μm)	5.13 (5.51 μm)
	87.8 (6.11 μm)	8.21 (5.79 μm)
	83.9 (6.45 μm)	5.39 (6.17 μm)
	86.2 (6.66 μm)	6.51 (6.52 μm)
Average	85.8 ± 3.0	6.60 ± 1.37

diffuse into the diffusion chamber with different removed active layer thickness. The average percentage of radon gas that can get into the diffusion chamber was 98.4 ± 3.5 and 85.8 ± 3.0% for 4 and 5 PE membranes, respectively. On the other hand, the average percentage of thoron gas that can get into the diffusion chamber was 7.80 ± 0.41 and 6.60 ± 1.37% for 4 and 5 PE membranes, respectively.

With 4 PE membranes, only about 7.8% of thoron gas can get into the diffusion chamber while only about 2% of radon gas has been screened out. For 5 PE membranes, despite a little bit less thoron gas (i.e. about 6.6%) can get inside, a much larger amount of radon gas (14%) has been screened out. In this way, 4 PE membranes should be the optimal number, which corresponds to about 40 μm of PE. The actual radon and thoron concentrations can be determined by solving the simultaneous equations

$$X\varepsilon_1 + Y\eta_1 = A, \quad \text{for the diffusion chamber covered with filter paper}$$

$$0.984X\varepsilon_2 + 0.078Y\eta_2 = B,$$

for the diffusion chamber covered with 4 PE membranes

where X and Y are the radon and thoron concentrations, respectively; ε_{11} and η_1 are the sensitivities for radon and thoron, respectively, for the diffusion chamber covered with filter paper; ε_2 and η_2 are the sensitivities for radon and thoron, respectively, for the diffusion chamber covered with 4 PE membranes [11]; and A and B are the total alpha track density per unit time registered by the LR 115 SSNTD in the diffusion chambers covered by a filter paper and 4 PE membranes, respectively.

Table 5 shows the comparison of radon and thoron gas concentrations obtained using this "twin diffusion chambers method" and the reference values measured by RAD-7. The results show that the "twin diffusion chambers method" can accurately determine the radon and thoron gas concentrations at the same time.

Table 5
Comparison between the radon and thoron gas concentrations obtained using the "twin diffusion chambers method" and the reference values obtained by RAD-7

	Exposure 1	Exposure 2
Thoron gas concentration obtained by twin diffusion chamber method (Bqm^{-3})	877 ± 93	997 ± 105
Reference thoron gas concentration (Bqm^{-3})	828 ± 124	932 ± 140
Radon gas concentration obtained by twin diffusion chamber method (Bqm^{-3})	148 ± 20	164 ± 22
Reference radon gas concentration (Bqm^{-3})	152 ± 23	171 ± 26

4. Conclusions

(1) For exposures to radon gas only, the numbers of alpha-particle tracks registered by the LR 115 SSNTD in the Karlsruhe diffusion chamber covered with one PE membrane (about 10 μm thick) were enhanced, while those covered with two PE membranes or one filter gave correct radon exposure values. This can be explained by the enhanced deposition of radon progeny on the outside surface of the PE membrane together with the insufficient thickness of a single PE membrane to stop the alpha particles emitted from these deposited radon progeny to reach the SSNTD. Calculations using SRIM show that a PE thickness of 13.52 μm is needed to completely stop the alpha particles from reaching the SSNTD.
(2) For exposures to mixtures of radon and thoron gas, calculations using SRIM show that 4 PE membranes (with a total thickness of about 40 μm) will stop all alpha particles from the radon or thoron progeny deposited on the outside surface of the PE membranes from being registered on the LR 115 SSNTD in the Karlsruhe diffusion chamber.
(3) For the "twin diffusion chambers method", one of the diffusion chambers is covered with PE membranes. It has been found that 4 PE membranes provide the optimal situation which allows the largest amount of radon gas (about 98.4%) to diffuse into the diffusion chamber while at the same time screening out the largest amount of thoron gas (about 92.2%). The method has been validated through experiments.

Acknowledgement

This research was supported by a grant from the Research Grants Council of the Hong Kong Special Administrative Region, China (Project No. CityU 103204).

References

[1] V.A. Nikolaev, R. Ilic, Radiat. Meas. 30 (1999) 1.
[2] D. Chen, X. You, R. Hu, Radiat. Meas. 39 (2005) 205.

[3] J. Swakon, K. Kozak, M. Paszkowski, R. Gradzinski, J. Loskiewicz, J. Mazur, M. Janik, J. Bogacz, T. Horwacik, P. Olko, J. Environ. Radioact. 78 (2005) 137.
[4] D. Nikezic, K.N. Yu, Mater. Sci. Eng. R 46 (2004) 51.
[5] Abdel-Fattah Hafez, G. Somogyi, Nucl. Tracks 12 (1986) 697.
[6] H.S. Virk, N. Sharma, Appl. Radiat. Isotopes 52 (2000) 137.
[7] D. Barooah, I. Laskar, A.K. Goswami, T.V. Ramachandran, K.S.V. Nambi, Radiat. Meas. 36 (2003) 461.
[8] M. Sreenath Reddy, P. Yadagiri Reddy, K. Rama Reddy, K.P. Eappen, T.V. Ramachandran, Y.S. Mayya, J. Environ. Radioact. 73 (2004) 21.
[9] J.K.C. Leung, D. Jia, M.Y.W. Tso, Nucl. Instr. Meth. Phys. Res. A 350 (1994) 566.
[10] C.W.Y. Yip, J.P.Y. Ho, V.S.Y. Koo, D. Nikezic, K.N. Yu, Radiat. Meas. 37 (2003) 197.
[11] S.Y.Y. Leung, D. Nikezic, J.K.C. Leung, K.N. Yu, Nucl. Instr. and Meth. B, in press, doi:10.1016/j.nimb.2007.04.116.
[12] F.M.F. Ng, C.W.Y. Yip, J.P.Y. Ho, D. Nikezic, K.N. Yu, Radiat. Meas. 38 (2004) 1.
[13] J.F. Ziegler, SRIM-2003.26, http://www.srim.org/, 2003.
[14] J.F. Ziegler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solids, Pergamon, New York, 1985.