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**A straightforward approach for assessing the effectiveness of
membrane materials as radon (^{222}Rn) barriers**

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A straightforward approach for assessing the effectiveness of membrane materials as radon (^{222}Rn) barriers

Abstract: The ubiquitous presence of the radioisotope radon (^{222}Rn) and its short-lived progeny (^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po) is challenging in two respects: (i) Radon is a major issue regarding health-related problems due to potentially elevated radiation exposure of humans in dwellings, and (ii) due to the mobility of radon the short-lived progeny may cause complications in radionuclide detection in laboratories. Polymer membranes are an appropriate means for effectively preventing unwanted radon migration. However, most of the published literature focusses on robust membranes made for the large-scale sealing of dwelling substructures. Membranes that are suitable (at small-scale) for sealing purposes in radionuclide detection applications are only rarely discussed. In this paper, we present a straightforward practical approach that allows the effectiveness of any membrane to be assessed for any purpose related to radon sealing. Executing the approach requires only (i) a suitable container with inlet and outlet ports, (ii) a mobile radon detector, and (iii) any type of radon source material. The approach provides a tool that allows testing any available membrane for its applicability as radon barrier sheeting.

Keywords: membrane; natural radioactivity; permeation; radio ecology; radon; radon barrier; Rn-222; sealing

1. Introduction

Due to the omnipresence of ^{238}U in nature and due to the gaseous nature of ^{222}Rn (in the following referred to as ‘radon’), which allows its easy migration, both radon itself and its progeny are ubiquitously present in nature. On the one hand, this omnipresence is of advantage as it allows radon to be used as environmental tracer in groundwater migration

investigations [e.g. 1], in groundwater/surface water interaction studies [e.g. 2,3], in investigations that aim at forecasting tectonic events [e.g. 4,5] and in subsurface contamination evaluations [e.g. 6,7]. On the other hand, radon (^{222}Rn) and its short-lived progeny (^{218}Po , ^{214}Pb , ^{214}Bi , ^{214}Po) may cause two general problems, namely elevated radiation exposure of humans in dwellings [e.g. 8–11] and complications in radionuclide detection routines in laboratories [e.g. 12,13].

A common method to tackle these problems (i.e. to prevent (i) radon diffusion into dwellings, (ii) diffusive radon loss from sample capsules, or (iii) plate-out of short-lived radon progeny onto radiation detector components) is to seal the items of concern. Membranes made of (coated) polymer material have proven suitable as effective radon barriers. Numerous studies that focussed on the sealing of dwelling substructures have been executed in the past applying a wide range of approaches. The results of these studies give an overview of the suitability of several types of membranes [e.g. 13–16 and citations therein].

Despite this substantial number of published data, the study presented here was executed for two reasons: (1) Most of the published literature focusses on robust membranes that are meant for sealing dwelling substructures. Membranes suitable for tackling the abovementioned problems related to radionuclide detection in laboratories are only rarely of concern. (2) Purchasing an (expensive) industrial membrane exclusively produced for achieving the latter might be avoidable if an alternative (less expensive) membrane turns out to be adequate for a specific purpose. Hence, case-specific individual tests for approximating the effectiveness of a certain membrane material as a radon barrier might be desirable for end-users.

Instrumental setups for executing such tests that are presented in the literature are often complex. The major reason for that might be that the procedures have to meet

accreditation requirements by the respective national legislative [e.g. 17]. Here we present a straightforward practicable approach that allows approximating the effectiveness of any membrane material requiring only (i) a suitable (self-made) container with inlet and outlet ports, (ii) a (lendable) mobile radon monitor, and (iii) any (natural) radon source material.

2. Material and methods

2.1. Polymer permeability – State of the art in brief

Gas transport through polymers in general has been the subject of investigation for several decades starting in the mid-20th century [e.g. 18,19]. These original approaches have been reviewed extensively in more recent years [e.g. 20–22]. From the resulting data, it can be concluded that the predominant influencing factor for gas permeation through polymeric systems is the structure of the polymer. Here, the literature distinguishes generally between hard ('glassy') and soft ('rubbery') polymers. Related radon-specific data indicate that rubbery polymers allow much higher gas permeation rates than glassy polymers [23–25].

The gas permeation rate is furthermore governed by the size and shape of the permeating gas molecules (or atoms) and by the solubility of the gas in the polymer material, which, in turn, is a function of temperature. The latter dependencies are quantitatively less understood, since gas-specific permeation rates have been determined for only a few gases. It can generally be stated, that the gas permeation rate as well as its solubility is inversely proportional to the molecular/atomic radius of the gas.

For radon, a radioactive gas, the diffusion length allows the gas permeation rate to be evaluated. The diffusion length (d_e) is defined as the diffusion distance (under steady-state conditions), over which the radon concentration is reduced by a factor of

$1 - e^{-1}$ (i.e. by ca. 63 %). The radon diffusion length can be calculated individually for each polymer by applying Equation (1), where D (m^2/s) is the polymer-specific radon diffusion coefficient and λ the radon decay constant ($2.1 \cdot 10^{-6} \text{ s}^{-1}$).

$$d_e = \sqrt{D/\lambda} \quad (1)$$

The polymer-specific wall thickness that would be required for a ‘radon-tight’ membrane can be determined based on the radon diffusion length. Four times d_e results in a reduction of the radon concentration within the polymer layer to about 1 % of its initial value.

Wojcik [25] has published a set of radon specific data. For rubbery polymers the study revealed radon diffusion lengths of between 0.5 mm (for ‘butyl rubber’) and 2.2 mm (for ‘rubber soft’) (Figure 1). This indicates that even for the ‘radon-tightest’ of the materials tested (‘butyl rubber’), a wall thickness of about 2 mm would be required to prevent radon passage through the material. Hence, it can generally be stated that uncoated rubbery polymer materials are not suitable as effective radon barriers.

2.2. Experimental setting

The complete experimental setup applied in our study is illustrated in Figure 2. For the experiments, the inner volume of a desiccator (22.2 L) was divided by the membrane of concern into a radon source volume (lower part of the desiccator; 17.2 L) and a radon receiving volume (upper part of the desiccator; 5.0 L). Both volumes were equipped with inlet and outlet ports. An AlphaGuard mobile radon monitor (Bertin GmbH, Germany) was used for radon measurement in the receiving volume. Before each experimental run, a volume of radon-rich air (about 25–100 mL) was injected into the source volume via the inlet port. A flow-through radon calibration standard (Pylon, Canada) was used as radon source. The resulting source activity concentrations ranged

between 30 and 423 kBq/m³ (*cf.* Table 1). For recording the radon activity concentration in the source volume a second radon detector ('Radon Scout home', Sarad, Germany) was added to the setup. Still, this additional device is not mandatory. The radon source activity concentration can alternatively be measured at the end of the experiment using the mobile radon monitor that was used for radon measurement in the receiving volume. Furthermore, it should be pointed out that any other comparable container, mobile radon monitor or radon source material (e.g. mineral sand rich in ²²⁶Ra) would be appropriate.

The exposed membrane area was 0.125 m². At least 10 cm of membrane material was allowed in excess all around the desiccator rim in order to prevent radon leak-flow from the source volume into the receiving volume. The membrane was tightly attached to the desiccator's glass rim by means of Vaseline.

For the experiments, the AlphaGuard (with a detection chamber volume of 0.6 L) was connected to inlet and outlet port of the receiving desiccator volume by means of Tygon[®] tubing, and a closed loop gas flow (0.5 L/min) was started by means of the AlphaGuard internal gas pump. The radon concentration in this closed (receiving) gas loop was recorded with a 10 min counting cycle. The radon concentration in the source volume was recorded with a 60 min counting cycle. Potential leakage of radon from the system during the experiments was checked by comparing the monitored overall radon inventory of the system to the $^{222}\text{Rn} = f(t)$ function that would be expected due to decay of the initial radon inventory alone.

Five exemplary membrane specimens were investigated, namely (1) a 0.01 mm HDPE membrane (cut from a garbage bag; Swirl–Melitta Group GmbH, Minden, Germany, 'HDPE'), (2) a 0.1 mm robust membrane produced from LDPE recycling granulates with a high degree of crystallinity (Polifilm GmbH, Cologne, Germany,

'LDPE-1'), (3) a fourfold layered (i.e. 0.4 mm) stack of the latter LDPE-1 material, (4) a 2 mm soft LDPE sheet with a low degree of crystallinity (sold as desk pad, 'LDPE-2'), and (5) an aluminium-coated 0.48 mm robust textile (ALUJET Climajet SD100; ALUJET GmbH, Mammendorf, Germany, 'Al-coated'). Two experimental runs were executed with each of the five membrane specimens, resulting in five pairs of results. For all five pairs of experiments the overall experimental setting and execution differed only in the source activity concentration and the ambient air temperature (Table 1; since the experiments were not run in an air-conditioned room ambient temperature could not be kept constant). Between two experiments with the same membrane specimen several days were allowed for purging the membrane (by diffusion) in a radon-free environment. Thus, it was made sure, that no 'old' radon from the previous experiment remained in the membrane material.

2.3. *Data evaluation approach*

Each experiment started with a 100 % radon concentration gradient between source volume and receiving volume, resulting in an initially linear increase of the radon concentration in the receiving volume. Calculation of the radon permeation rate was done based on two parameters: (i) the initial radon source concentration and (ii) the slope of the initially linear increase of the radon concentration in the receiving volume [25]. Figure 3 displays an exemplary dataset. The plot reveals that the onset of the linear section of the slope is (in the given case) somewhat delayed. This membrane-specific delay depends (in addition to the ambient temperature as well as the size and shape of the permeating gas molecules or atoms) on the structure of the membrane polymer and the thickness of the membrane. While very thin membranes (e.g. the tested 0.01 mm HDPE membrane) result in a virtually immediate onset of the linear slope, the exemplary plot in Figure 3 reveals a time lag of about 5 hours.

3. Results and discussion

A precondition for the successful execution of the experiments is the radon-tightness of the system. This was controlled by comparing the recorded radon inventory of the overall system (i.e. the summed inventories of source volume and receiving volume including the AlphaGuard detection chamber) to the theoretical decay of the initial inventory. The results revealed that radon leakage from the system was generally about 5 % during a three-day experiment and thus negligible (in particular for the phase of the linear slope). Still, the results of some experiments carried out with the fourfold layered stack of the LDPE-1 membrane had to be neglected because the Vaseline caused puckers in the stacked LDPE layers resulting in leaks and thus uncontrolled radon loss from the system.

The experimental results reveal that the Al-coated material did not allow any radon permeation at all. The radon concentration in the receiving volume stayed at background level (12 Bq/m^3) even though a very high source activity concentration of 423 kBq/m^3 was applied. In fact, the initial (i.e. ambient) radon background concentration in the receiving volume decreased over time in accordance with the radon decay constant thereby confirming the radon-tightness of the receiving volume (Figure 4).

In contrast to the Al-coated material, the eight experiments executed with the four polymer membrane specimens revealed radon permeation through the tested membranes. Table 2 summarizes the experimental results. Displayed are the resulting mean values for each set of experiments ($n = 2$). To allow a comparison of the detected radon permeation rates (i.e. linear slopes) that were experimentally determined with different source activity concentrations, we normalized each radon permeation rate ($\text{Bq/m}^2\text{s}$) to the source activity resulting in a slope/source ratio (m/s). In order to make

the resulting values more manageable we applied a factor of 10^{-7} to the resulting slope/source ratios.

The data in Table 2 reveal the following general effects of an increase in membrane thickness (see also Figure 5 A–D): (i) the slope/source ratio decreases, (ii) the time lag increases and (iii) the determination coefficient of the linear slope decreases. These trends are briefly discussed in the following paragraphs.

Both slope/source ratio and time lag depend directly on the polymer-specific radon permeation rate and allow therefore an evaluation of the suitability of the membrane to block radon permeation. Generally, the data reveal a proportional relationship between membrane thickness and time lag (Figure 6A) and inverse proportionality between membrane thickness and slope/source ratio (Figure 6B). Specifically, the data show that none of the tested specimens (except the Al-coated material discussed above) block radon permeation satisfactorily. Even the 2 mm LDPE sheet allows radon permeation of $0.0188 \text{ Bq/m}^2\text{s}$ if a source activity concentration of 1 kBq/m^3 is applied.

The experimental variance of the slope/source ratios (with $n = 2$) is most pronounced for the fourfold layered stack of the LDPE-1 material. The results of the two experiments differ by 18 % from their mean value. On the other hand, the slope/source ratios of the two experiments executed with the 0.01 mm HDPE membrane are practically identical. Generally, the variance in data resulting from experiments with an identical membrane can be attributed to the fact that the radon permeation rate is influenced by the ambient temperature. Since the temperature could not be kept constant during all experiments, it was continuously recorded. Generally, it can be stated that otherwise identical experiments yielded higher slope/source ratios if they were executed at higher temperatures (*cf.* Table 1). Hence, it can be assumed that the observed

variances in the slope/source ratios for otherwise identical experiments are due to temperature differences. The unusual high variance observed for the 0.4 mm LDPE-1 material (18 %) is probably due to the unusual material property of the specimen (fourfold layered stack).

The determination coefficients summarized in Table 2 indicate the uncertainty of the individual results. The data show that greater uncertainties are generally due to lower activity concentrations in the receiving volume. Figures 5 A–D illustrate this observation in four exemplary plots. The plots show clearly how onset time lag and uncertainty of the recorded linear slope increase with increasing membrane thickness.

4. Conclusion

The paper introduces a simple experimental setup and a related data evaluation approach that allows assessing the effectiveness of membrane materials as radon barriers. Potential areas of application include the radon sealing of dwelling basements as well as radon sealing of detection equipment in radionuclide laboratory applications. The presented method allows not only a qualitative assessment of any membrane material but also a quantitative evaluation of its radon permeation rate. Besides confirming the general applicability of the developed approach, the experimental results revealed, that only one of the five tested membrane specimens is applicable as radon barrier. It was shown that, besides polymer structure and thickness of the membrane, the ambient temperature plays a noteworthy role. Because of its simple experimental setup, the presented method gives anyone a tool at hand that allows testing any available membrane for its applicability as radon barrier sheeting.

Disclosure statement

No potential conflict of interest was reported by the authors.

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Figure captions

Figure 1. ^{222}Rn diffusion coefficient (D) vs. ^{222}Rn diffusion length (d_e) illustrated for four soft polymers (based on Wojcik [25]).

Figure 2. Experimental setup.

Figure 3. Exemplary dataset illustrating the $f(t)$ increase of the radon concentration in the receiving volume for a thick membrane (0.4 mm LDPE-2).

Figure 4. Radon concentration in the receiving volume during the experiment with the aluminium-coated material compared to the theoretical concentration decrease by decay.

Figure 5 A–D: Exemplary datasets illustrating time lag and linear slope (incl. variance) of the increasing radon concentration (dotted line) for individual experiments executed with the membrane specimens (A) ‘HDPE’, (B) 0.1mm ‘LDPE-1’, (C) 0.4mm ‘LDPE-1’ and (D) ‘LDPE-2’. Data points of the linear sections are marked with diamonds, data points of the time lag sections with crosses (no such section in Figure 5A).

Figure 6: Membrane thickness and time lag (A) and membrane thickness and slope/source ratio (B) illustrated for the tested membrane types.

Table 1. Setting for 5×2 experiments executed with five different membrane specimens.

Membrane thickness	Initial source activity [kBq/m ³]	Temperature [°C]
HDPE (0.01 mm)	42	12.2
	59	17.0
LDPE-2 (0.1 mm)	235	14.4
	167	20.8
LDPE-2 (0.4 mm)	88	17.7
	127	20.9
LDPE-1 (2 mm)	221	18.1
	125	19.7
Al-coated (0.48 mm)	423	19.8
	120	20.0

Table 2. Experimental results for polymer membranes (mean values, n=2).

Membrane material		Slope/source ratio $\times 10^{-7}$ (m/s)*	Time lag (h)	Determination coefficient of linear slope (R^2)
HDPE (0.01 mm)		$5.9342 \pm 0.05 \%$	0.1	1.00
LDPE-2 (0.1 mm)		$0.5069 \pm 10 \%$	0.3	0.99
LDPE-2 (0.4 mm)		$0.1210 \pm 18 \%$	4.8	0.94
LDPE-1 (2 mm)		$0.0188 \pm 5.3 \%$	36.5	0.86

*The unit (m/s) indicates a radon permeation rate (Bq/m²s) normalized to the source activity concentration.





