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^{226}Ra measurement via gamma-ray spectrometry of ^{222}Rn progeny – quantification of radon losses from sample capsules

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Abstract

The detection of radium-226 (^{226}Ra) in soil and sediment samples is generally executed by means of gamma-ray spectrometry. Data evaluation relies (besides the actual ^{226}Ra gamma peak at 186.2 keV) on the combined analysis of major gamma peaks that are produced by the two short-lived radon (^{222}Rn) daughter nuclides ^{214}Pb and ^{214}Bi. Precondition for this detection approach is equilibrium decay of all members of the decay chain between ^{226}Ra and ^{214}Bi. In closed systems, this decay equilibrium is reached after about five half-lives of ^{222}Rn, i.e. after about 20 days. However, a closed system can only be guaranteed if the capsule which contains the sample prevents diffusive escape of the noble gas radon. Such radon-tightness cannot be guaranteed for a wide range of plastic materials. Due to its polymer structure, plastic material generally tends to allow radon diffusion and hence radon loss from the enclosed sample resulting in a disturbance of the required decay equilibrium. The paper introduces an approach that allows quantifying radon loss from sample capsules by direct radon measurements using mobile radon detection equipment. The experimental findings are supported by theoretical considerations. An examined alternative approach based on the offset of the 186.2 keV data point from an efficiency function that is calculated exclusively from short-lived radon progeny peaks from ^{214}Pb and ^{214}Bi in the gamma-ray spectrum did not prove to be applicable due to a lack of supporting peaks in the low-energy section of the spectrum.

Keywords: environmental tracer; gamma-ray spectrometry; isotope measurements methods and equipment; natural radioactivity; quantifying radon loss; radium-226; radon-222

1. Introduction

Radium (^{226}Ra, $t_{1/2}$ = 1600 a) is the parent nuclide of the naturally occurring noble gas radon (^{222}Rn, $t_{1/2}$ = 3.8 d) within the uranium-238 decay chain. Radium is ubiquitously present in soils and sediments with activity concentrations ranging in general between about 5 and 50 Bq/kg. Radium activity concentrations are of interest in several fields of applied geosciences. A main research area is radiation protection related to radium in subsoil or building materials giving rise to elevated concentrations of radon and its progeny in indoor air [e.g. 1,2]. Another main field of application is the combined use of radium and radon as naturally occurring tracers for studying hydrological processes [e.g. 3–5].

Radium activity concentrations in soils and sediment samples are commonly determined by means of gamma-ray spectrometry. The decay of ^{226}Ra into its immediate daughter ^{222}Rn results in a gamma-ray peak at 186.2 keV. However, the associated gamma line has a relatively low emission probability of only 3.5 % and it is furthermore overlapping with the ^{235}U gamma peak at 185.7 keV (emission probability 57.2 %). Thus, radium activities revealed by the 186.2 keV gamma line are usually confirmed by cross-evaluating gamma lines that originate from the decay of two short-lived ^{222}Rn daughters, namely ^{214}Pb and ^{214}Bi. Precondition for this combined data evaluation is a decay equilibrium between ^{226}Ra, ^{214}Pb and ^{214}Bi. In a closed system this equilibrium is practically reached after five ^{222}Rn half-lives, i.e. after about 20 days.

However, since the noble gas ^{222}Rn escapes quickly and easily from soil or sediment samples, the decay equilibrium is generally disturbed in any kind of freshly taken sample. To allow establishment of the equilibrium for proper gamma spectrometry, soil or sediment samples have to be stored in sample containers which are ‘radon-tight’ (and also suitable as measuring geometry for the gamma detector in use) [6].

The material of such sample container has to be chosen carefully. Glass is considered to be radon-tight. However, due to its fragility and its potential radium content glass is generally less appropriate for the purpose. Sample containers made of plastic material are more common. Still, since most polymeric materials do not block radon diffusion satisfactorily [e.g. 7], potential radon loss from the samples has to be considered and quantified when the gamma spectrometry data is evaluated [6].

Some fundamental studies on the permeation of radon through polymeric materials have been published (for details see Sect. 4). Sinn [8] and Müller [9] measured both diffusion coefficient and solubility of radon in commercially available plastic materials. Their work was continued by Wojcik [10]. Radon permeabilities were determined experimentally for other plastic materials by Arafa [11]. Yet, when the available datasets are looked at in detail, inconsistencies are noticeable, which can be attributed to the imprecise characterization of the studied polymer types in terms of composition, porosity and/or homogeneity. These deficiencies result in a limited comparability of the data available so far. Nevertheless, the published datasets allow the semi-quantitative conclusion that dense plastic materials (such as high-density polyethylene ‘HDPE’, polyethylene terephthalate ‘PET’ or polymethyl methacrylate ‘PMMA’) block diffusive radon losses more efficiently than materials with a lower density (such as low-density polyethylene ‘LDPE’ or silicone rubber). These theoretical results were confirmed by more applied tests that proved the suitability of PET and polylactic acid ‘PLA’ for storing water samples with only limited radon loss [12]. Still, for sound assessment of gamma spectrometric data that results from measurements applying plastic sample containers as part of the measurement geometry, more specific quantitative information regarding the material-specific radon loss is compulsory.

Besides choosing suitable sample containers made of a suitable material, the sealing the lid of the containers, i.e. keeping the containers leakage proof during storage and measurement, has been discussed in the literature. It was shown that sealing the lid after filling the container with the sample improves the radon-tightness of the sample significantly. Options to seals the lid include the use of PVC tape, aluminium tape or two-component epoxy adhesive [13] or the complete vacuum packaging of the sample container in an aluminium lined bag [14]. Another influential parameter is the surface/volume ratio of the containers. The higher it is, the higher is the likelihood of diffusive radon loss [11]. Consequentially, Marinelli beakers show a higher potential for loosing radon than cylindrical capsules [13].

In the paper, we discuss an approach for quantification of radon losses from sample capsules by direct measurement using a radon-in-air monitor attached to a simple experimental setup. Furthermore, the possibility of quantifying radon losses indirectly based on the recorded gamma spectrometry data is critically evaluated. The paper is completed with theoretical considerations of radon diffusion through plastic materials that help understanding and estimating any material-specific radon loss results.

2. Material and methods

For exemplarily quantifying radon losses from plastic sample capsules, we used cylindrical capsules of two types. ‘Type 1’ is 3.2 cm in height and 6.7 cm in diameter (volume 113 cm^3); ‘Type 2’ is 1.9 cm in height and 4.8 cm in diameter (volume 34 cm^3). The capsules have a wall thickness of 1.5 mm. The material can best be described as heat-resistant, opaque, non-flexible plastic that comes close to a material known under the trade name ‘Bakelite’ [15].

For preparation of standardized specimens, the capsules were filled with homogenized uranium tailings, i.e. fine grained residuals that remain after crushing and leaching of uranium ore. The tailings material can be characterized (based on 15 randomly taken samples) as well-sorted medium grained sand with a ^{226}Ra activity concentration of A_{Ra} = 2.66 ± 0.27 Bq/g, a dry density of δ = 1.65 g/cm^3, a radon emanation coefficient of ε = 0.23 and a pore space of n = 0.4 [16]. Hence, apart from the ^{226}Ra activity concentration the material can be can be characterized as comparable to average soil or sediment samples.

Five standardized specimens were prepared for the experiments by filling the tailings material into three Type 1 capsules (named ‘1A’, ‘1B’ and ‘1S’) and two Type 2 capsules (‘2A’ and ‘2B’). The capsules were closed with tightly fitting lids of identical plastic material. Flexible sticky tape (Cellpack Premio 233) was wrapped around the rim of the lids to hamper radon diffusion. The interstice between capsule body and tightly fitting lid of specimen ‘1S’ was additionally sealed with a flexible glue (Hylomar® M) in order to most efficiently block radon diffusion along this pathway. Subsequently the five specimens were stored for 20 days in a low radon environment (open lab shelf) to reach radioactive equilibrium between ^{226}Ra,

^{214}Pb and ^{214}Bi. Table 1 summarizes the activities of ^{226}Ra and its short-lived progeny and other relevant characteristics of the five specimens. The ^{222}Rn production rate within the capsules was calculated based on the ^{226}Ra activity concentration (A_{Ra}). The ^{222}Rn activity concentration in the pore space (C_{Rn}) was calculated based on the parameters given above (i.e. A_{Ra}, δ, ε and n) based on Equation (1).

$$C_{Rn} = \frac{A_{Ra} \times \delta \times \varepsilon}{n} \tag{1}$$

3. Experimental

Direct radon loss measurements were carried out by means of the experimental setup illustrated in Figure 1. All detection equipment was chosen and assembled carefully in order to avoid radon losses through the detection equipment. An AlphaGuard® radon monitor (Bertin Technologies, France) was used for radon detection. The closed air loop was kept circulating by means of an AlphaPump®, i.e. an air pump that has been designed specifically for pumping air (with a pump rate of up to 1 l/min) allowing only negligible loss of radon. All parts of the experimental setup were connected with Tygon® tubing (Saint Gobain, France). For the purpose the tubing type Tygon R-3603 was chosen since the R-3603 resin is the most suitable regarding radon tightness. Still, the plasticizer added to the resin in order to make the tubing material flexible might offer a minor possibility of radon permeation. In order to strictly minimize radon losses by diffusion through the tubing walls, all tube connections between the parts of the experimental setup were kept as short as possible. Hence, radon losses through the detection equipment could be considered insignificant.

For the measurements, the five specimens were individually placed in a radon-tight radium-free glass container, which was equipped with an inlet and an outlet port. After placing a specimen in the container, the container was tightly closed and sealed. The container was additionally sealed from the outside air by putting it under water. Subsequently air was pumped in a closed loop though the specimen container and the radon-in-air monitor. The pump rate was kept constant at 1 l/min. The total volume of gas circulating through the closed system was 1600 cm^3.

For each of the five specimens the radon inventory of the closed air loop (I_{Rn}; [Bq]) was recorded continuously as time series in 10-minutes counting intervals. Each measurement started with a radon background concentration of about 30 Bq/m^3, i.e. a radon background inventory of the closed system of $I_{Rn} = 0.05$ Bq. The initial radon background concentration represented the ^{222}Rn concentration in the ambient air. The detector internal background was subtracted beforehand.

The diffusive radon loss from the specimens resulted in a gradual increase of I_{Rn} (Figure 2). For investigating the diffusive radon loss from the capsules, two parameters were recorded: (1) the virtually linear slope of the I_{Rn} increase during the initial phase of each experiment and (2) the equilibrium of I_{Rn} that was finally reached (Figure 2). Three identical measurements were executed with each of the five specimens in order to increase statistical significance of the results.

Since the described direct measurement approach is rather time-consuming (and requires a mobile radon detector, which might not be available), the possibility of deducing the diffusive radon loss indirectly by evaluating the recorded gamma-ray spectrum was considered. This examined indirect evaluation approach is based on the following idea: The short-lived ^{222}Rn progeny ^{214}Pb and ^{214}Bi produce at least 25 distinctive gamma lines with satisfactory emission probabilities (incl. the low energy x-ray line of ^{214}Pb at 77.1 keV), which are all indirectly indicative of the ^{226}Ra in the sample. If there is no loss of radon from a sample capsule, the ^{226}Ra data point at 186.2 keV (being subject to a traceable ^{235}U interference, see below) must fit to an efficiency function that is derived exclusively from the ^{222}Rn progeny peaks. If, on the other hand, there is radon loss from the capsule, the 186.2 keV data point will plot above this fitted function, with the distance between the measured and the fitted 186.2 keV data points quantitatively indicating the radon loss from the individual sample capsule.

For all five specimens, the ^{235}U contribution to the 186.2 keV peak was calculated and allowed for based on the ^{235}U peaks at 143.9, 163.4 and 205.3 keV. The resulting ^{235}U activity was furthermore checked for reasonability by analysing the peak of the immediate ^{238}U daughter ^{234m}Pa (at 1001 keV; half-life: 1.17 min) and comparing the resulting experimental ^{238}U/^{235}U activity with the naturally occurring undisturbed ^{238}U/^{235}U ratio (21.72 ± 0.15) that can be expected for the used tailings material.

The five specimens were each measured for at least 24 hours by low-level gamma-ray spectrometry using a shielded coaxial HPGe detector, P-type (ORTEC), with an active volume of 107 cm^3. The spectra were analysed with the software GAMMA-W® [17]. All gamma-ray data used in this paper are taken from [18].

4. Theoretical considerations

Diffusive gas transport through polymers has been studied since the mid-20th century [e.g. 19,20]. The originally published datasets have been reviewed extensively in more recent years [e.g. 11,21,23]. The resulting findings reveal generally that the structure of the polymer is the predominantly influencing factor for gas permeation through it. Regarding this structure, polymers can in principle be classified as rather hard (‘glassy’) or rather soft (‘rubbery’). As a rule of thumb, soft materials show a much higher gas diffusion coefficient (D; [m^2s^{-1}]) than hard polymers [8–10].

In addition to the structure of the polymer, gas diffusion is governed by the gas-specific gas solubility in the polymer material. Although gas solubility data has been published for only a few polymer materials, it can generally be stated that the gas solubility (and also the gas-specific diffusion coefficient) shows a trend that goes inverse to the size (and shape) of the permeating gas molecules or atoms.

Due to the radioactivity of radon, its permeation rate through polymer materials can be quantified by the radon diffusion length (d_e). Generally, the diffusion length is defined as the steady-state diffusive migration distance in any (polymer) material, over which the radon concentration decreases by a factor of $1–e^{-1}$ (i.e. by ca. 63 %). Equation (2) allows calculation of d_e individually for each polymer based on the polymer-specific radon diffusion coefficient (D) and the radon decay constant (λ; 2.1×10^{-6} s^{-1}).

$$d_e = \sqrt{D/\lambda} \tag{2}$$

The diffusion length allows to assess the polymer-specific wall thickness that is required for blocking diffusive radon loss completely. Four times d_e results in a reduction of the radon concentration within the polymer down to about 1 % of its source value. Wojcik [10] specified a radon diffusion length of $d_e = 0.11$ mm for two hard polymers, namely the polyamid ‘Supronyl’ and the polyacrylate ‘Plexi Glass’. This indicates that a wall thickness of at least about 0.5 mm would prevent diffusive radon loss for either of these two polymers. For two other hard polymers, namely polyurethane (PU) and polyvinylchloride (PVC), the required wall thickness was determined to be around 3 mm, also based on Wojcik’s [10] results.

No radon diffusion coefficient is available from the literature for Bakelite, which complicates assessing the material-specific radon diffusion length in the case of our study. However, based on related data that is known for other gases (and allowing for the radon atomic diameter) it is possible to at least estimate the radon diffusion coefficient for Bakelite. Barrer [24] determined the permeation velocities of H_2 and N_2 through Bakelite at room temperature. He did not report any solubility data, which complicates the quantification of the related diffusion coefficients. Still, the reported permeation velocity ratio of H_2 and N_2 (10/1) and the ratio of their molecule diameters (0.271 nm / 0.357 nm; [25]) allow an estimate regarding radon. With consideration of the much larger atomic diameter of radon (0.520 nm) it can at least be approximated that its permeation through Bakelite must be significantly slower than that of N_2.

Another indirect way to obtain a reasonable estimate of the radon diffusion coefficient in Bakelite is based on the ‘lag time’ necessary for reaching steady state conditions of radon transport (i.e. I_{Rn} equilibrium in our case; *cf*. Figure 2) through a polymer layer (i.e. the capsule wall in our case). The approach is based on an empiric relationship suggested by Wojcik [10] (Equation (3)) that implies that the lag time is solely determined by the diffusion coefficient (D; [m^2s^{-1}]) and the thickness of the polymer layer (z; [m]).

$$t = z^2/6D \tag{3}$$

It has to be mentioned, though, that the experimental concept applied in our case differs from the one used by Wojcik [10]. In both approaches the radon concentration within the polymer layer is initially at zero. In the experimental arrangement applied by Wojcik [10] the concentration at the face through which the diffusing radon emerges is maintained ‘at low concentration levels compared to the opposite face’ throughout the whole experiment. In contrast, in our case the concentration at the ‘off-site’ face rises steadily. However, even in steady state (i.e. after about 4.5 days; *cf*. Figure 2B) the concentration gradient is still around two orders of magnitude, i.e. ‘at low concentration levels compared to the opposite face’. That makes our experimental concept comparable to the one discussed by Wojcik [10]. Furthermore, Wojcik [10] determined the lag time by back-extrapolation from a plot ‘cumulative radon flux vs. time’ (*cf*. Figure 1 in [10]). We, on the other hand, defined it as the time after which steady state concertation is reached in the closed air loop. Still, in both cases the lag time is the time after which the radon flux across the polymer layer (i.e. membrane and capsule wall, respectively) is in steady-state, which makes both experimental concepts comparable. Hence, we consider Equation (3) appropriate for obtaining at least a reasonable estimate of the radon diffusion coefficient in Bakelite even though the experimental setup used in [10] differs from ours.

5. Results

5.1. Direct radon loss measurement

Averaged over all 3 x 5 experiments it was found that the I_{Rn} increase in the beginning of each experiment is almost linear for about five hours. The I_{Rn} equilibrium value is reached after a lag time of about 4.5 days (*cf*. Fig. 2).

Table 2 summarizes for each of the five specimens the radon loss, i.e. the ‘radon flux’ (F) from the capsules, distinguishing between (i) the flux occurring during the phase of initial linear I_{Rn} increase (F_{slope}) and (ii) the flux after reaching the phase of I_{Rn} equilibrium (F_{eq}). The resulting losses are given both as absolute radon flux rates [Bq/s] and as rates relative [%] to the specimen-specific radon production rate (P_{tot}).

During the initial phase of linear I_{Rn} increase, the maximum radon concentration gradient between specimen and air loop triggered maximum radon flux (as quantified by F_{slope}). The ratio F_{slope}/P_{tot} allows quantifying the relative diffusive radon loss, i.e. the diffusive radon loss related to the radon production rate. The F_{slope} scenario represents the situation of a virtually radon-free outer environment, i.e. the situation met by any sample stored within a plastic capsule. Thus, F_{slope} can be used as quantitative indicator for the radon loss in practical applications.

After radon equilibration, the radon flux decreased to the rate required for balancing radon decay in the closed air loop (F_{eq}). This scenario does not reflect any situation of practical relevance. Still, the observed lag time that elapsed until radon equilibrium is reached can be used as parameter for obtaining a reasonable estimate of the radon diffusion coefficient in Bakelite. Based on Equation (3), our capsule wall thickness ($z = 0.15$ cm) and the determined lag time (about 4.5 days) we can assume a radon diffusion coefficient of about $D = 10^{-8}$ cm² s^{-1}. Using this estimate for D leads (based on Equation (2)) to the conclusion that a wall thickness of around 3 mm (four times d_e) would be necessary to make a Bakelite container radon-tight. This required thickness is similar to that of hard PU and PVC (see Sect. 4), i.e. makes Bakelite regarding radon diffusion comparable to hard PU and PVC.

The result supports the assumption that the major radon leakage pathway was diffusion through the capsule plastic material (besides diffusive leakage through the interstice between capsule body and tightly fitting lid). The measurement results suggest this pathway for two more reasons: (1) Larger relative losses were detected for the capsule Type 2. Although the Type 2 capsules are smaller (thus showing a smaller surface area), their surface area / volume ratio is larger than the corresponding Type 1 value. (2) The additional sealing of the lid of the Type 1 capsule (Type 1S) reduced the radon loss only from about 33 to 22 %, thus suggesting diffusion through the capsule walls as a major migration pathway.

In summary, the results in Table 2 reveal that the 1.5 mm Bakelite plastic material does not block radon diffusion. The Type 1 (large) and Type 2 (small) capsules lose about 33 and 39 % of their total radon production, respectively. Since diffusion through the capsule walls is the main pathway of radon loss, additional sealing of the capsule does not block radon loss completely. The sealed specimen 1S still loses 2/3 of the radon that is lost by the unsealed but otherwise identical specimens 1A/B.

5.2. Indirect determination of radon losses based on gamma spectrometry data

An individual efficiency function was constructed for each of the five recorded spectra based exclusively on the ^{214}Pb and ^{214}Bi datapoints. Over 25 peaks were used for the construction of each function. The efficiency value resulting from the ^{226}Ra peak (at 186.2 keV) was also calculated. As expected, for all five specimens the measured (and ^{235}U-corrected) 186.2 keV data points lie significantly ($>1\sigma$) above the fitted efficiency function. This confirms the conclusion of the radon loss experiments that radon is escaping from the tested Bakelite capsules (Sect. 5.1).

However, all efforts to analyse the datasets in more detail revealed that a quantitative evaluation of the radon losses based on the determined ratios of the fitted/measured efficiency values for the 186.2 keV peak is not possible. While the measured efficiency data points at 186.2 keV could be determined with sufficient precision, the corresponding fitted values could not be determined with the required accuracy. The main reason for this is the lack of low energy datapoints for reliably constructing the radon progeny efficiency function. The only reasonable data point for the energy range below 186.2 keV comes from the rather weak x-ray line of ^{214}Pb at 77.1 keV. This lack of data points supporting the efficiency function in its low-energy section resulted in an uncertainty of the fitted function around 186.2 keV that did not allow reliable quantitative conclusions based on the difference between the measured and the fitted efficiency values at 186.2 keV.

6. Conclusions

The data resulting from our study of diffusive radon loss from plastic capsules reveal the following conclusions:

(1) The introduced method for quantifying radon losses from sample capsules that are applied as part of a gamma detection geometry proved appropriate.

(2) The Bakelite capsules that were only tape-sealed allowed substantial diffusive radon losses of up to 40 %. Additional tight sealing of the interstice between capsule body and lid reduced the radon loss but still did not block it completely. Consequently, we could identify leakage via diffusion through the capsule walls as main reason for the radon loss. Theoretical considerations on radon permeation through plastic materials support this finding.

(3) The examined straightforward approach of quantifying radon losses based on the evaluation of the gamma spectra data did not prove to be applicable because of lacking datapoints supporting the efficiency function in its low-energy section.

Generally, it can be stated that any user should be aware that radon losses from sample capsules might occur and might be significant. Hence, checking for potential radon loss is advisable for any established gamma detection setup.

Disclosure statement

No potential conflict of interest was reported by the authors.

References

[1] Font L, Baixeras C. The RAGENA dynamic model of radon generation, entry and accumulation indoors. Sci Total Environ. 2003;307:55–69.

[2] Appleton JD, Cave MR, Miles JCH et al. Soil radium, soil gas radon and indoor radon empirical relationships to assist in post-closure impact assessment related to near-surface radioactive waste disposal. J Environ Radioac. 2011;102(3):221–234.

[3] Moore WS, Sarmiento JL, Key RM. Submarine groundwater discharge revealed by ^{228}Ra distribution in the upper Atlantic Ocean. Nat Geosci. 2008;1:309–311.

[4] Moore WS. Sources and fluxes of submarine groundwater discharge delineated by radium isotopes. Biogeochemistry. 2003;66:75–93.

[5] Rocha C, Wilson J, Scholten J et al. Retention and fate of groundwater-borne nitrogen in a coastal bay (Kinvarra Bay, Western Ireland) during summer. Biogeochemistry. 2015;125:275–299.

[6] Scholten JC, Osvath I, Pham MK. ^{226}Ra measurements through gamma spectrometric counting of radon progenies: How significant is the loss of radon?. Mar Chem. 2013;156:146–152.

[7] Schubert M, Altendorf D, Weiß H. A straightforward approach for assessing the effectiveness of membrane materials as radon (^{222}Rn) barriers. Isot Environ Health Stud. 2022;58:301–310.

[8] Sinn D. Diffusionskonstante, Löslichkeit und Permeabilität von Radon in handelsüblichen Kunststoffen [Staatsexamensarbeit]. Institut für Umweltphysik, Universität Heidelberg, Germany; 1978. German.

[9] Müller W. Die Diffusions-, Löslichkeits- und Permeabilitätskoeffizienten von Radon-222 in handelsüblichen Kunststoffen [Staatsexamensarbeit]. Institut für Umweltphysik, Universität Heidelberg, Germany; 1978. German.

[10] Wojcik M. Measurement of radon diffusion and solubility constants in membranes. Nucl Instr Meth B. 1991;61:8–11.

[11] Arafa W. Permeability of radon-222 through some materials. Radiat Meas. 2002;35:207–211.

[12] Lucchetti C, De Simone G, Galli G, et al. Evaluating radon loss from water during storage in standard PET, bio-based PET, and PLA bottles. Radiat Meas. 2016;84:1–8.

[13] Bonczyk M, Samolej K. Testing of the radon tightness of beakers and different types of sealing used in gamma-ray spectrometry for ^{226}Ra concentration determination in NORM. J Environ Radioact. 2019;205/206:55–60.

[14] Mauring A, Gäfvert T. Radon tightness of different sample sealing methods for gamma spectrometric measurements of ^{226}Ra. Appl Radiat Isot. 2013;81:92–95.

[15] Baekeland LH. Bakelit, ein neues synthetisches Harz. Chemiker-Zeitung. 1909;35(33):317–318. German.

[16] Schubert M. Erfassung von Untergrundkontaminationen durch Non-Aqueous Phase-Liquids (NAPLs) mit Hilfe der Bestimmung des Radongehalts der Bodenluft [PhD thesis] Georg-August-Universität, Göttingen; 2000. (UFZ-Bericht; 1/2001). German.

[17] GAMMA-W. Software for the high-precision analysis of HPGe spectra. Gesellschaft für Kernspektrometrie mbH, Ebsdorfergrund (Germany); 2017.

[18] Westmeier W, Reus U. Catalogue of gamma-rays from radioactive decay, 2nd edition, unpublished; 2000.

[19] Barrer RM. Diffusion in and through solids. Cambridge: Cambridge University Press; 1941.

[20] Crank J. Park GS. Diffusion in polymers. London and New York: Academic Press; 1968.

[21] Klopffer MH, Flaconnèche B. Transport properties of gases in polymers: Bibliographic review. Oil Gas Sci Technol (Rev IFP). 2001;56(3):223–244.

[22] Flaconnèche B, Martin J, Klopffer MH. Transport properties of gases in polymers: Experimental methods. Oil Gas Sci Technol (Rev IFP). 2001;56(3):245–259.

[23] George SC, Thomas S. Transport phenomena through polymeric systems. Prog Polym Sci. 2001;26:985–1017.

[24] Barrer RM. Permeability of organic polymers. Trans Faraday Soc. 1940;35,644–648.

[25] D’Ans J, Lax E. Taschenbuch für Chemiker und Physiker, Vol. 1, 4th ed. Berlin: Springer, 1992.

Table1. Specimen characteristics.

specimen	Volume (cm³)	Weight (g)	activity of ^{226}Ra and progeny (Bq)	^{222}Rn production (Bq/s)	^{222}Rn in pore space (kBq/m³)
1A	113	182.4	485 ± 49	1.02 ± 0.10E-3	2410 ± 24
1B	113	187.0	497 ± 50	1.04 ± 0.11E-3	2530 ± 25
1S	113	183.0	487 ± 49	1.02 ± 0.10E-3	2530 ± 25
2A	34	54.1	144 ± 14	3.02 ± 0.31E-4	2350 ± 24
2B	34	55.2	147 ± 15	3.08 ± 0.31E-4	2440 ± 24

Table 2. Radon losses from defined specimens during the phase of initial linear I_{Rn} increase and after reaching I_{Rn} equilibrium; all data achieved by direct radon loss measurement.

specimen (n = 3)	Rn loss initial phase (Bq/s)	Rn loss initial phase (%)	Rn loss equilibrium (Bq/s)	Rn loss equilibrium (%)
1A	3.40 ± 0.35E-04	33.4 ± 5.0	8.38 ± 0.84E-05	8.2 ± 1.0
1B	3.51 ± 0.35E-04	33.6 ± 4.5	8.77 ± 0.88E-05	8.4 ± 1.0
1S	2.28 ± 0.23E-04	22.4 ± 3.3	5.91 ± 0.59E-05	5.8 ± 0.7
2A	1.25 ± 0.13E-04	41.3 ± 6.8	2.84 ± 0.28E-05	9.4 ± 1.3
2B	1.14 ± 0.12E-04	37.1 ± 6.1	2.98 ± 0.30E-05	9.7 ± 1.3

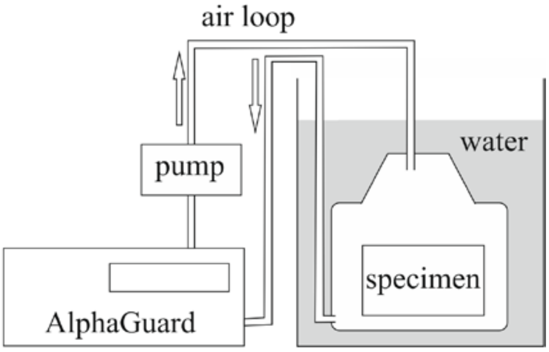

Figure 1. Experimental setup for direct measurement of diffusive radon losses.

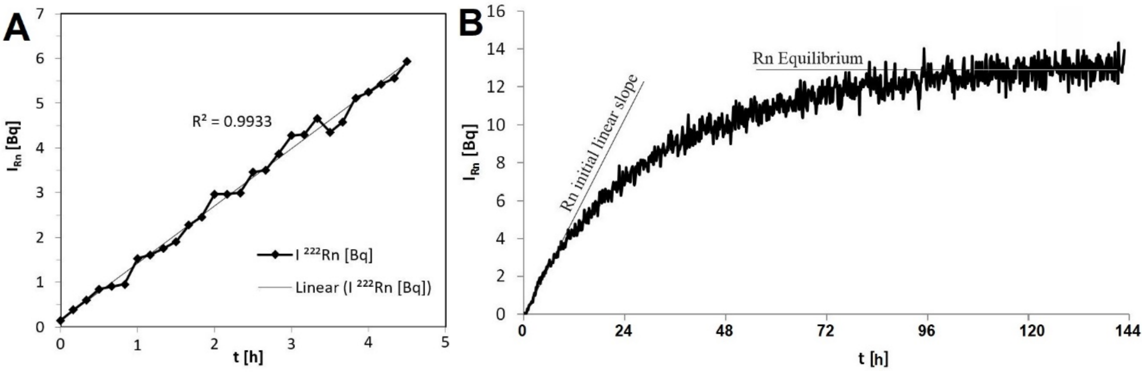

Figure 2. Exemplary dataset ‘RADON inventory’ vs. Time; A: initial linear slope; B: development of equilibrium plateau.