



## Review

## Radon diffusion coefficient measurement in waterproofings – A review of methods and an analysis of differences in results

Kateřina Rovenská<sup>a,\*</sup>, Martin Jiránek<sup>b</sup><sup>a</sup> Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Břehová 7, 11000, Praha 1, Czech Republic<sup>b</sup> Faculty of Civil Engineering, Czech Technical University in Prague, Thákurova 7, Praha 6, 166 29, Czech Republic

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## ABSTRACT

This paper summarizes information about 16 measuring methods for determining the radon diffusion coefficient of waterproofing materials. We have found that the differences in results for identical membranes, which can be as high as two orders of magnitude, can mainly be attributed to insufficient duration of the tests, insufficient radon concentration to which the samples are exposed, and the use of steady state calculation procedures for data measured under non-steady state conditions.

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## 1. Introduction

If they are placed over the entire surface of the substructure of a house, suitable waterproof membranes can prevent soil radon from entering buildings. In many countries, membranes are therefore considered as a basic measure for protecting new buildings. According to UK building regulations (Coskeran et al., 2007), all houses in areas where more than 3% of properties exceed the action level of 200 Bq/m<sup>3</sup> have had to be fitted with a membrane since 1999. In the Czech Republic, a membrane has had to be installed in all houses built in radon-prone areas since 1995 (CSN 73 0601, 2006). The effectiveness of membranes in

reducing radon transport from the soil depends mainly on their chemical composition, on their mechanical properties, on their ability to create airtight joints, on pipe penetrations and on sufficient durability.

The barrier properties of membranes against radon penetration are usually expressed in terms of the radon diffusion coefficient. The values of this parameter in 360 waterproofing materials are summarized in Jiránek and Kotrbatá (2011), where it is shown that there are great differences in the diffusion properties. Depending on the chemical composition, the radon diffusion coefficients in waterproofings widely used for protecting houses vary within eight orders from 10<sup>−15</sup> to 10<sup>−8</sup> m<sup>2</sup>/s. The lowest values were obtained for bitumen membranes with an Al carrier, irrespective of whether the bitumen was modified. On the other hand, the highest radon diffusion coefficient values were discovered for sodium bentonite membranes, rubber membranes

\* Corresponding author.

E-mail address: [katerina.rovenska@suro.cz](mailto:katerina.rovenska@suro.cz) (K. Rovenská).

made of EPDM, and polymer cement coatings. The radon diffusion coefficients for PVC, HDPE, LDPE, polypropylene and bitumen membranes vary in the range from  $3 \times 10^{-12}$  to  $3 \times 10^{-11}$  m<sup>2</sup>/s.

In general, we find three different approaches to the use of the radon diffusion coefficient. In Ireland, the upper limit for the radon diffusion coefficient is applied. According to *Building Regulations* (1997), the maximum radon diffusion coefficient value is set to  $12 \times 10^{-12}$  m<sup>2</sup>/s. The second approach compares the thickness of the membrane with the radon diffusion length calculated as  $l = (D/\lambda)^{1/2}$ . For example, in Germany a membrane can be considered as radon-tight if it is at least three times thicker than the radon diffusion length  $l$  (Keller et al., 2001). And, lastly, in the Czech Republic and in Slovakia the radon diffusion coefficient is used for calculating the minimum thickness of the membrane for each house (Jiraneck and Hulka, 2001; Jiránek, 2004; CSN 73 0601, 2006), taking into account other parameters such as the soil gas radon concentration at the building site, the characteristics of the house (ventilation rate, area in contact with the soil), etc.

Although the diffusion coefficient is applied differently from country to country, it has been found to be a suitable parameter for selecting the effective radon barriers from among the membranes available on the building materials market. A need has therefore arisen for comparable results for the radon diffusion coefficient value for a single type of membrane measured and certified by different laboratories around the world.

A theoretical comparison of different methods used for radon diffusion coefficient measurement is presented in this paper. The results of 1st International comparative measurement of the radon diffusion coefficient in polymer membranes (Rovenska and Jiránek, 2011), supported by this review, led to the decision to prepare a unified method for measuring the radon diffusion coefficient.

It should be noted that other less usual names can be found in the literature for the radon diffusion coefficient, such as radon resistance, permeability coefficient (SP-Sweden, 2003), permeation coefficient (Labed et al., 1997), etc. In the present work, the radon diffusion coefficient is an effective parameter that includes not only diffusion through the membrane but also other physical processes, such as solubility and adsorption of radon and diffusion of radon through pinholes and pores.

## 2. A summary of methods for determining the radon diffusion coefficient in waterproofing materials

For the purposes of this paper, we have summarized 16 measuring techniques described in scientific journals or protocols. Most of the methods share the same principle: A sample of the tested material is placed between the source container and the receiver container. A special type of container-sample-container design is the measurement system proposed by Perry et al. (1996), in which the alpha particle counter serves as the radon detector. Due to the short range of alpha particles in air, the distance between the sample and the detector is approx. 18 mm (Perry et al., 1996), which means that the volume of the receiver container is very small.

The source is either an artificial source of radon (LER, 2009; Ďurčík and Havlík, 1996; Jiraneck and Fronka, 2008; Wojcik et al., 2000; Arafa, 2002) or concrete or another material with a high content of radium (<sup>226</sup>Ra) which decays into radon (<sup>222</sup>Rn) (SP-Sweden, 2003). The radon concentration generated in the source container varies over a wide range, from kilobecquerels (Ďurčík and Havlík, 1996; Sarad Geolab GMBH, 2009; Arafa, 2002; Daoud and Renken, 2001) up to megabecquerels (Jiraneck and Fronka, 2008; Quindos Poncela, 2005; Gulabyants, 2009; Perry et al.,

1996). The radon concentration in the two containers is monitored by various measuring devices attached directly to the containers, or connected to the containers by flexible piping, or the containers themselves serve as measuring devices. All known concepts use scintillation (Quindos Poncela, 2005; Ďurčík and Havlík, 1996; Wojcik et al., 2000) or semiconductor detectors (Rad7 (Cosma, 2009), gamma spectrometry (Pressyanov et al., 2009), Sarad (Sarad Geolab GMBH, 2009)) or ionization chambers (current mode (Jiraneck and Fronka, 2008), AlphaGuard (LER, 2009)) as measuring units. The surface area of the samples varies from laboratory to laboratory, and according to the measuring device, but the sample area is typically tens to hundreds of cm<sup>2</sup>. The volume of the receiver container also depends on the type of measuring device, but a typical volume is several tenths of litres to units of litres. The duration of the measurement varies from several tens of hours up to several weeks. Monitoring of the ambient parameters is usually not documented in the literature. Ďurčík and Havlík (1996) specify measurements of temperature, relative humidity and pressure, and the measured diffusion coefficient is corrected on the basis of the results. Jiraneck and Fronka (2008) measure the ambient pressure and temperature, but no correction is made.

The information presented in Table 1 leads to the conclusion that the radon diffusion coefficient is in most cases measured under non-stationary conditions. The decisive measurement of radon concentrations (the measurement used for calculating the radon diffusion coefficient) starts immediately after radon has been admitted into the source container. During this type of measurement, the radon concentrations in the two containers change with time, and as a consequence the radon concentration within the tested sample and the radon exhalation rate from the sample into the receiver container also change.

Using various mathematical techniques (simple steady state or complex numerical solutions), the radon diffusion coefficient is then calculated from the time-dependent courses of the radon concentrations measured in the source and receiver containers, and the area and thickness of the tested sample. The theory is based on Fick's law. The differential equations that are used for calculating the diffusion coefficient or the diffusion length are usually provided without the boundary conditions or any description of further application. Measurement and calculation in steady state is used in Sarad Geolab GMBH (2009), Fernandez et al. (2004), but in some laboratories the diffusion coefficient is calculated from stationary equations, though the measurement is provided under non-steady state conditions (SP-Sweden, 2003; LER, 2009). LER (2009) and SP-Sweden (2003) laboratories calculate the radon permeability under the assumption of a linear concentration profile within the sample, though the measurement is provided in non-steady state.

There are two exceptions to the common container-sample-container design. The first of these, described by Quindos Poncela (2005) and Gulabyants (2009), has the form of a chamber which is sealed by the sample. At one moment high radon activity is introduced into the chamber and the subsequent decrease in radon concentration caused by radon decay and its diffusion into the sample is monitored. From the known radon concentration at  $t=0$ , it is possible to calculate the decay curve. The difference between the measured curve and the theoretical curve is used by Quindos Poncela (2005) and Gulabyants (2009) for calculating the diffusion coefficient. The weakness of this method is the unknown ventilation rate of the chamber, and the unknown depth of radon diffusion into the sample.

The second method, proposed by Pressyanov et al. (2009), is based on measuring the radon desorption from the sample into a huge free air chamber. A gamma-spectrometry measurement of <sup>214</sup>Pb in the sample is used. The method calculates the diffusion

**Table 1**

Details on various radon diffusion coefficient measurement techniques obtained from the literature.

Ref.	<i>T</i>	<i>S.surf.</i>	<i>V</i>	<i>SC<sub>Rn</sub></i>	Rn detector	<i>Q</i>	Regime
(LER, 2009)	–	590 cm <sup>2</sup>	9.5 l	–	AlphaGuard	<i>k</i> , <i>P</i> <sup>*</sup>	–
(Hosoda et al., 2009)	–	121 cm <sup>2</sup>	–	Tens kBq/m <sup>3</sup>	Pylon	<i>D<sub>ef</sub></i>	NSD/C
(Steiner et al., 2009)	Days	–	–	Various sources strength	Rad7	<i>L</i>	SD
(Malki et al., 2010)	21 day	–	–	300 kBq/m <sup>3</sup>	Electrets, Activated charcoal	<i>L</i>	SD
(Đurčák and Havlík, 1996)	Days (weeks)	From 50 to 600 cm <sup>2</sup>	30 l	200 kBq/m <sup>3</sup>	Scintillation chamber	<i>D</i>	C
(Pressyanov et al., 2009)	66 h	–	50 l	1600 Bq/l	Gamma spectrometry	<i>L</i>	–
(Cosma, 2009)	48–98 h	400 cm <sup>2</sup>	–	Hundreds of kBq/m <sup>3</sup>	Rad7, Radim-Eman	<i>D</i>	C
(Sarad Geolab GmbH, 2009)	Days (weeks)	78 cm <sup>2</sup> resp. 225 m <sup>2</sup>	–	Hundreds of kBq/m <sup>3</sup>	Radon Monitor RM 2000	<i>P</i>	SD
(Quindos Poncela, 2005)	Tens of hours	–	–	Hundreds of MBq/m <sup>3</sup>	Scintillation chamber	<i>D</i>	–
(Fernandez et al., 2004)	Days, 5 × rel. time	40 cm <sup>2</sup>	8 l	–	–	<i>D</i>	SD
(Gulabyants, 2009)	Hours	Depends	Depends	Hundreds to thousands kBq/m <sup>3</sup>	Scintillation chamber	<i>D</i>	–
(Wojcik et al., 2000)	Tens to hundreds of hours	22.9 cm <sup>2</sup>	0.028 l	18.5 kBq <sup>226</sup> Ra	Receiver cont. is scintillation chamber	<i>D</i>	NSD
(Jiranek and Fronka, 2008)	Tens to hundreds of hours	152 cm <sup>2</sup>	2.319 l	Thousands of kBq/m <sup>3</sup>	Receiver cont. is ionizing chamber in current mode	<i>D</i>	NSD
(Arafa, 2002)	4 day	81 cm <sup>2</sup>	0.0162 l	Approx. 1850 Bq/m <sup>3</sup>	Activated charcoal	<i>K</i>	NSD
(Daoud and Renken, 2001)	Time to steady state calculated using time-lag method	80 cm <sup>2</sup>	–	From 36 to 432 kBq/m <sup>3</sup>	Continuous monitor	<i>D</i>	SD
(Perry et al., 1996)	Days	230 cm <sup>2</sup>	–	3 MBq/m <sup>3</sup>	Alpha particle detector	<i>D</i>	SD*

*T* – Duration of measurement.*S.surf* – Sample surface.*V* – Receiver container volume.*SC<sub>Rn</sub>* – Source radon concentration.*Q* – Measured quantity:*D* – Diffusion coefficient (m<sup>2</sup>/s).*P* – Radon permeability ( $P = \frac{f_{idl}}{\sin(d/L)}$  according (Fernandez et al., 2004)).*L* – Diffusion length (m).*k* – Radon permeability (m<sup>2</sup>/s), *P*<sup>\*</sup> – radon transmittance (m/s),  $k = P^* \cdot d$ , where *d* is thickness of sample (LER, 2009).*K* – Radon permeability (Arafa 2002).**Regime** – Regime of diffusion (steady state – SD, Non-steady state – NSD), Regime of source (Constant – C), SD\* the steady state assumption is valid only for very thin samples (Perry et al., 1996).

coefficient and the solubility of radon in the sample. The effective diffusion coefficient is obtained by multiplying these quantities. The mathematical solution is described in Pressyanov et al. (2009).

Arafa (2002) uses two cans with activated charcoal. The first can is open face; the second can is tightly sealed with the membrane sample. Both of them are enclosed in box of much larger volume than the volume of the cans. Approximately 4 day after admitting the radon gas into the large chamber, the cans are measured after establishing the equilibrium between radon and progenies by shielded NaI(Tl). The net count from the energy range of 295 to 609 keV representing <sup>214</sup>Pb and <sup>214</sup>Bi is calculated.

In the past years, the radon diffusion coefficient became important in low background experiments. Special and very precise techniques are reported by Zuzel (2005) and Rau (2005).

### 3. Consequences of different measuring approaches

The summary of measuring methods shows that there are no clearly-defined, well-established or standardized techniques for determining the radon diffusion coefficient in radon barrier materials. Since different requirements for radon-proof membranes are applied in different countries (not only within the EU member states), manufacturers and suppliers of membranes must undergo testing in each country in order to comply with local requirements. As a result of this impractical situation, different test protocols with extraordinarily different results can be found for the same membrane. During the last 15 years, since measurement of the radon diffusion coefficient has been required in the Czech Republic by the CSN 730601 standard, we have had several opportunities to compare our values with the results stated in the test protocols produced by foreign laboratories. In general, the

differences between the results have sometimes been extremely high for the same membrane, so that they cannot be neglected. For example, in the category of bitumen membranes the differences vary within 2 orders of magnitude, and in the category of cement coatings even higher differences have been found. To shed some light on the sources of these divergences, international comparative measurements of the radon diffusion coefficient in waterproofing membranes were jointly organized in 2009 and 2010 by the National Radiation Protection Institute in Prague and the Faculty of Civil Engineering of the Czech Technical University in Prague. Eleven laboratories from Europe, Asia and the USA participated in these measurements (some of the measuring techniques are described above), but not all participants submitted their results. The aim of the comparative measurements was to determine the radon diffusion coefficient in three different types of LDPE and HDPE membranes. The radon diffusion coefficient values obtained from the comparison varied for HDPE membranes from  $2.2 \times 10^{-13}$  to  $7.1 \times 10^{-12}$  m<sup>2</sup>/s, and for LDPE membranes from  $9.6 \times 10^{-13}$  to  $2.2 \times 10^{-11}$  m<sup>2</sup>/s (Rovenska and Jiránek, 2011). Thus the above-mentioned discrepancies were also confirmed for membranes of quite different chemical composition. A detailed description of the comparative measurements, together with an analysis of the results, is presented in a separate article (Rovenska and Jiránek, 2011). Here we would like to concentrate on explaining the reasons for the differences.

### 4. Analysis of reasons responsible for differences between results

Section 2 shows that the methods differ in a great number of important factors, such as the mathematical processing of the

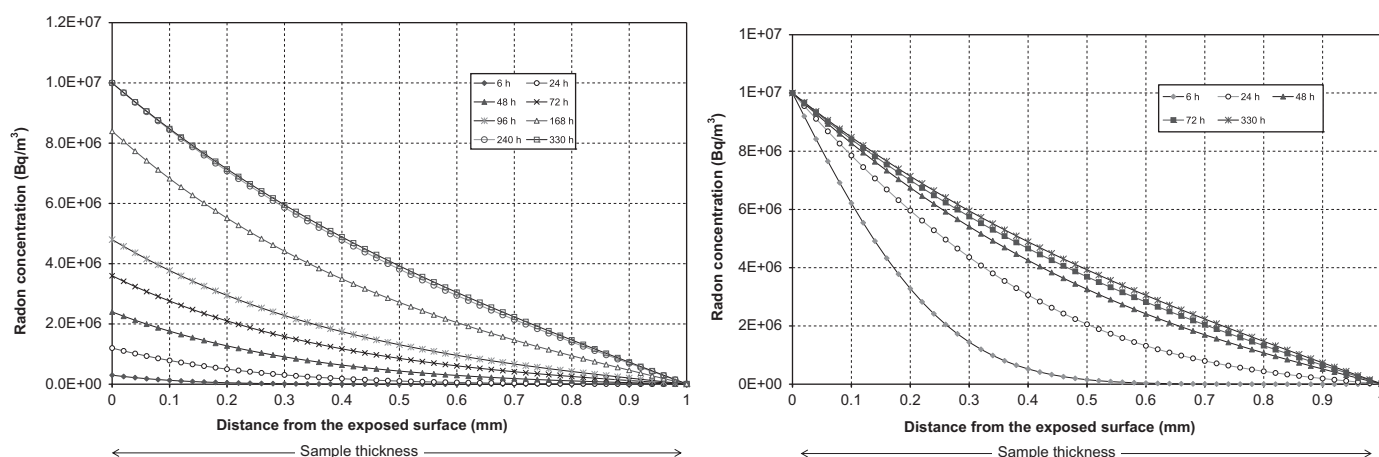
measured data, the boundary conditions under which the test is carried out (stationary or non-stationary), the duration of the test (from several tens of hours up to one month), the radon concentration to which the samples are exposed, the assumed radon distribution within the membrane, the shape of the increase in radon concentration in the two containers, the measuring technique used for monitoring the radon concentrations, etc. The main problem is that some of the assumptions that have been adopted are not generally valid, but are valid only for certain materials and/or in certain conditions. Examples of assumptions that lead to errors, if they are used universally, include:

- linear radon distribution within the measured sample,
- a linear build-up of radon concentration in the receiver container,
- the use of stationary numerical techniques under non-stationary conditions

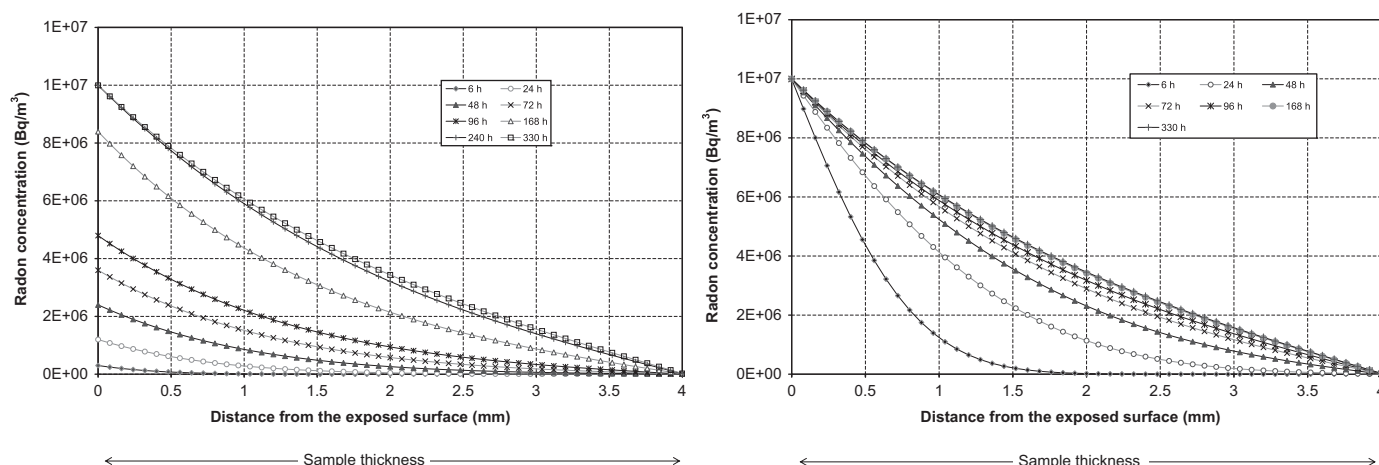
These assumptions were analysed by the TransRn universal numerical program (Jiranek and Svoboda, 2009), which simulates the whole time-dependent measuring process. The program is based on the well-known partial differential equation describing one-dimensional non-stationary radon transport by diffusion.

#### 4.1. Linear radon distribution within the measured sample

The radon concentration profile within the tested sample generally has the shape of an exponential function. It is only in some special cases, e.g., for very thin membranes with the radon diffusion coefficient close to the order  $10^{-10} \text{ m}^2/\text{s}$  or higher, that the radon distribution can be almost linear. However, this is not a typical situation because membranes with such a high radon diffusion coefficient are too permeable for use as efficient radon-proof barriers. Time-dependent changes in radon concentration within the samples for two typical representatives of membranes (a 1-mm thick polymeric membrane with  $D=1 \times 10^{-12} \text{ m}^2/\text{s}$  and a 4-mm thick bitumen membrane with  $D=1 \times 10^{-11} \text{ m}^2/\text{s}$ ) are presented in Figs. 1 and 2. During the calculation it was assumed that the radon concentration in the source container either has a stable value of  $10 \text{ MBq/m}^3$ , or increases at a rate of  $50 \text{ kBq/m}^3$  per day, and the steady state concentration in the source container of  $10 \text{ MBq/m}^3$  is attained at time  $t=200 \text{ h}$  after radon has been admitted into the source container. Both figures confirm the exponential shape of the radon concentration profile within the membrane, even in the steady state. In addition, it is evident that the time needed to establish the steady state profile is proportional to the rate of radon concentration increase in the source container.



**Fig. 1.** Radon distribution within a membrane of  $D=1 \times 10^{-12} \text{ m}^2/\text{s}$  for gradually increasing radon concentration in the source container (left) and the steady state concentration in the source container of  $10 \text{ MBq/m}^3$  (right).



**Fig. 2.** Radon distribution within a membrane of  $D=1 \times 10^{-11} \text{ m}^2/\text{s}$  for gradually increasing radon concentration in the source container (left) and the steady state concentration in the source container of  $10 \text{ MBq/m}^3$  (right).

#### 4.2. Linear build-up of radon concentration in the receiver container

If the decisive measurement of the radon concentration in the receiver container starts immediately after radon has been admitted into the source container, the initial part of the build-up curve is not linear. This is because, at the beginning of the test, the membrane is not fully saturated with radon and hence the radon exhalation rate from the membrane into the receiver container is not steady. The significance of this non-linear shape increases as the thickness of the membrane increases and the radon diffusion coefficient decreases. Another important factor influencing the initial shape of the curve is the rate of the radon concentration increase in the source container. The effect of all of these factors is shown in Fig. 3. The figure indicates that the initial non-linear part can be observed within the first 1 to 10 day after the test has commenced. A linear approximation of the measured data in this phase, especially if the duration of the test is short, can lead to serious mistakes and to underestimation of the diffusion coefficient.

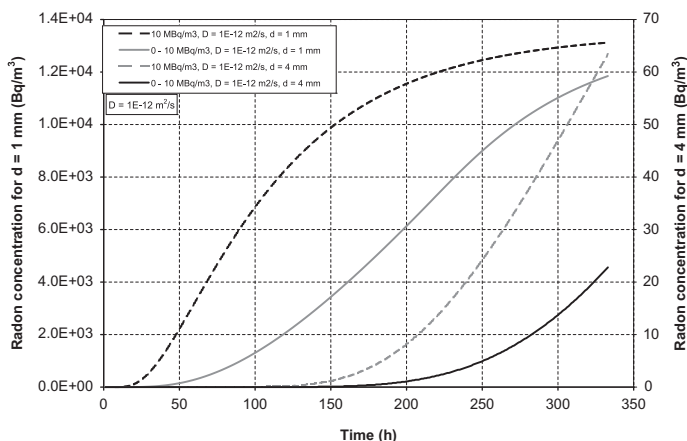
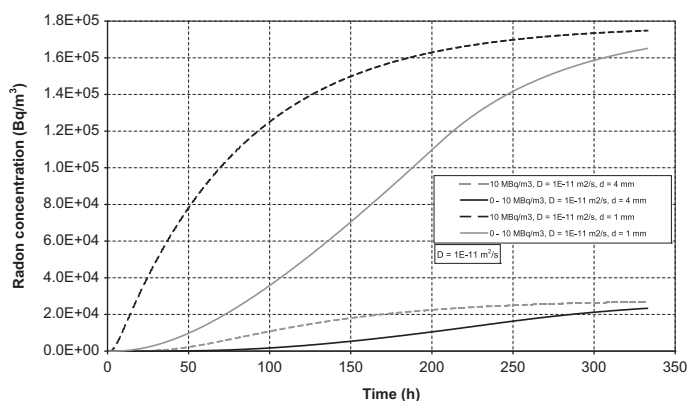
#### 4.3. Use of stationary numerical techniques under non-stationary conditions

Simple steady state calculation procedures can lead to accurate results only when all the assumptions are fulfilled (stable, i.e., time-independent, radon distribution within the sample and a

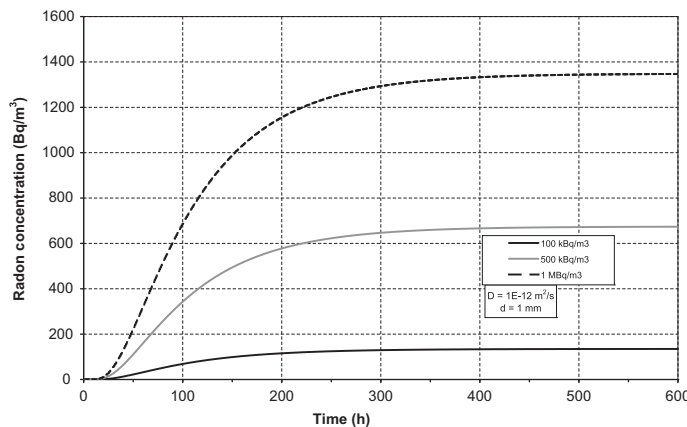
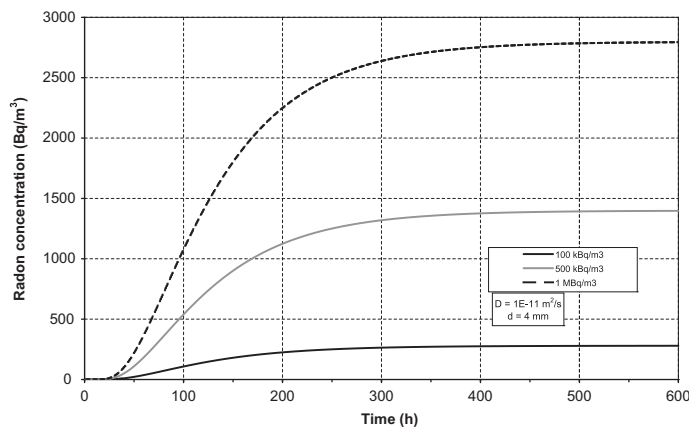
stable radon exhalation rate from the sample into the receiver container). This usually requires a sufficient amount of time, especially in the case of thick membranes with a low radon diffusion coefficient – as will be shown below. The duration of the tests stated in Table 1 indicates that the measurement is often stopped prematurely before attaining stationary conditions, and subsequent use of stationary calculation techniques will result in a considerable loss of accuracy for the calculated radon diffusion coefficient. Complex numerical modelling is the only way to overcome this problem, in cases when it is not convenient to wait until steady state conditions are established.

#### 4.4. Low radon concentration in a source container

That cannot generate sufficiently high radon concentration (i.e., well detectable by the measurement technique that is applied) in the receiver container is the other important factor that may result in significant errors. Correct prediction of the minimum radon concentration in the source container is a crucial prerequisite for ensuring that the measurement uncertainties will be as low as possible. For the two typical representatives of membranes mentioned above, Fig. 4 shows how the radon concentration in the receiver container is influenced by the radon concentration in the source container. It can be seen, for example, that for a membrane with a radon diffusion coefficient of  $1.10^{-12} \text{ m}^2/\text{s}$  and 1 mm in thickness, exposed to concentrations



**Fig. 3.** The shape of the built-up curves of the radon concentration in the receiver container in dependence on the radon diffusion coefficient, membrane thickness and rate of radon concentration increase in the source container. It was assumed that the samples are exposed either to the steady state radon concentration in the source container of  $10 \text{ MBq/m}^3$ , or to a gradual increase in the radon concentration in the source container at a rate of  $50 \text{ kBq/m}^3$  per day.



**Fig. 4.** Radon concentration in the receiver container for the two typical representatives of membranes in dependence on the steady state radon concentration in the source container.



of 100 kBq/m<sup>3</sup> or 500 kBq/m<sup>3</sup>, respectively, it takes 80 h or 37 h for the radon concentration in the receiver container to reach 50 Bq/m<sup>3</sup>. If we consider that a value of 1 kBq/m<sup>3</sup> in the receiver container in the steady state provides well detectable concentrations in the receiver container even at the beginning of the measurement, this membrane should be exposed to a concentration of at least 750 kBq/m<sup>3</sup>. The thickness of the membrane has a major influence on this assessment. For a membrane with the same radon diffusion coefficient but four times greater thickness (4 mm), the radon concentration in the source container must be more than 10 times higher (see Fig. 3 – for 10 MBq/m<sup>3</sup> in the source container, the radon concentration in the receiver container reaches 50 Bq/m<sup>3</sup> after a period of 307 h).

Accurate determination of the radon diffusion coefficient is also affected by inaccuracies arising during the measurement of radon concentrations in the containers. Typical sources of errors are as follows: (a) long time intervals between readings of the radon concentrations in the containers (intervals longer than 6 h cannot provide a precise description of the build-up curves), (b) alternate measurement of radon concentrations in the source container and in the receiver container by a single monitor (the concentrations in the containers at the same time steps are not known) and (c) the assumption that radon is distributed uniformly in the containers. This assumption is mostly not valid, and therefore measured radon concentration values depend on the ratio between the area of the sample and the volume of each container, and on the location of the detector within the container. The potential errors increase with increasing distance between the detector and the surface of the sample. The greatest differences between the measured values and the real concentration can be expected for devices with radon monitors located separately from the containers. A delayed response of detectors connected to the containers or located separately from the containers is a further source of errors. These disadvantages are almost removed when the containers serve at the same time as the radon detector.

## 5. Conclusion

A review of radon diffusion coefficient measurement methods confirmed that there is at present no standardised technique, at international level or even at national level, for determining the radon diffusion coefficient in waterproofing materials. Measurement techniques and methods differ in a great number of important factors, and in the boundary conditions under which the tests are carried out. As a result of this impractical situation, different test protocols with extraordinarily different results can be found for the same waterproofing material. The 1st international comparative measurement of this coefficient, organized in 2009 and 2010 (Rovenská and Jiránek, 2011), showed that the differences between particular laboratories could be as high as two orders of magnitude.

Detailed time-dependent numerical analysis of the measurement procedures performed by the TransRn universal numerical program showed that these differences can mainly be attributed to three factors – insufficient duration of the tests, insufficient radon concentration to which the samples are exposed, and the use of steady state calculation procedures for data measured under non-steady state conditions.

This analysis indicates a clear need to develop a unified test method on an international level for determining the radon diffusion coefficient in waterproofings which will ensure comparable results. This will be very helpful, especially in situations where several different measurement methods exist in Europe and where measurement of the radon diffusion coefficient of

waterproofings is required by various national technical standards in cases where the waterproofing is to act as a radon-proof membrane.

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