Ruvira-Quintana, B.; García-Fayos, B.; Juste Vidal, BJ.; Arnal Arnal, JM.; Verdú Martín, GJ. (2022). Experimental estimation of the diffusion coefficient in radon barrier materials based on ISO/TS 11665-13:2017. Radiation Physics and Chemistry. 193:1-8. https://doi.org/10.1016/j.radphyschem.2022.109993

Experimental estimation of the diffusion coefficient in radon barrier materials based on ISO/TS 11665-13:2017

Beatriz Ruvira[a,*], Beatriz García-Fayos[a], Belén Juste[a], José Miguel Arnal[a], Gumersindo Verdú[a]

[a]Institute for Industrial, Radiophysical and Environmental Safety (ISIRYM), Universitat Politècnica de València, Camino de Vera s/n. Valencia, 46022, Spain. E-mail: *bearuqui@upv.es

Abstract

Due to the health risks posed by high radon concentrations, many regulations impose concentration restrictions and set out several techniques to prevent radon entry into enclosed spaces. One of the most effective technique to protect new buildings from radon entry is to install a radon barrier over the entire surface of the ground beneath the foundation. Whether a barrier is suitable for protection is determined by the diffusion coefficient, the methodology which is standardised in ISO/TS 11665-13:2017. These barriers are typically made of polymeric or bitumen materials, which have diffusion coefficients between 10^{-11} and 10^{-13} m^2/s. By applying a modification to the ISO standard that allows shorter test times, the diffusion coefficients of a commercial membrane ($6.21 \pm 0.75 \cdot 10^{-13}$), polyethene ($1.14 \pm 0.14 \cdot 10^{-12}$), polypropylene ($4.35 \pm 0.53 \cdot 10^{-14}$), polyvinyl chloride ($3.55 \pm 0.43 \cdot 10^{-14}$), polystyrene ($4.96 \pm 0.60 \cdot 10^{-13}$) and polymethyl methacrylate ($4.07 \pm 0.49 \cdot 10^{-13}$) have been measured, the latter two material values not being published so far. The results validate the developed methodology since the diffusion coefficients provided are comparable with those of the literature. In all cases, a concentration reduction of 99% is obtained. A relationship is observed between the amount of radon adsorbed by the material and its thickness and chemical composition.

Keywords: radon, diffusion coefficient, radon barrier, polymeric material

1. Introduction

Radon-222 is a radioactive gas that comes from the uranium-238 decay chain (Brill et al., 1994; Donohoe and Royal, 1996; Khan et al., 2019). When radon disintegrates, it emits an alpha particle and starts a series of short-live descendants. It can be inhaled and transported into the lungs and if its progeny disintegration occurs there, alpha particles can damage the tissue and originate lung cancer (Donohoe and Royal, 1996; Khan et al., 2019; World Health Organization, 2009).

Radon originates in soils and rocks with radium content and because of its gaseous properties and its half-life of 3.82 days, it can travel through the pores and cracks and enter the buildings by diffusion and/or differences in pressure and temperature (Brill et al., 1994; Fucic et al., 2011; Gandolfo et al., 2017; Khan et al., 2019). Radon also exhales from the building materials, like concrete or bricks (Donohoe and Royal, 1996; Pacheco-Torgal, 2012).

The World Health Organization Handbook on indoor radon (World Health Organization, 2009) states that the radon mitigation strategies for new buildings are active and passive soil depressurization, sealing surfaces, barriers and membranes, and ventilation of occupied and unoccupied spaces. According to (Daoud and Renken, 2001) and (Khan et al., 2019), the most important mitigation method for new buildings is the installation of a radon-proof membrane across the foundation, which has to be airtight and continuous (Pacheco-Torgal, 2012) and must function properly during their service time, so the materials should have long endurance and high resistance to ageing (Jiránek and Hůlka, 2001).

Over the past few years, regulations in many European countries have required the installation of radon barriers to protect the newly constructed buildings if soil exhalation requires it (Holmgren and Arvela, 2012). In Spain, the transposition of the Euratom 2013/59 directive (Council of the European Union, 2013) has led to the modification of the Technical Building Code, including the obligation to install these barriers in new buildings in category I zones, which slow down the radon diffusion so that the concentration remains under 300 Bq/m^3 inside dwellings and workplaces (Spanish Ministry of Public Works and Transport, 2019).

The radon diffusion coefficient is a property that defines radon transport through the material, so it can be used to know the suitability of a material as a radon-proof barrier (Jiránek et al., 2008). The materials used as insulation in houses and buildings have radon diffusion coefficients between 10^{-8} and 10^{-15} m^2/s, and the most frequently used materials include bitumen membranes, polymeric membranes and some coatings. The lowest values correspond to bitumen membranes with an aluminium film and ethylene vinyl acetate (EVA) membranes, whereas the highest values are for sodium bentonite membranes, rubber membranes and polymer cement coatings. Waterproof materials commonly used for protecting buildings, i.e., polyethene (PE), polypropylene (PP), polyvinyl chloride (PVC) and bitumen membranes, have a radon diffusion coefficient in the range from 10^{-11} and 10^{-13} m^2/s (Jiránek and Hůlka, 2001, 2000; Jiránek and Kotrbatá, 2011).

The radon diffusion coefficient depends on material chemical composition as well as manufacturing, material density, raw materials, concentration gradient on both sides of the barrier, etc. so even for the same type of material some discrepancies in radon diffusion coefficient values can be found (Daoud and Renken, 2001; Jiránek and Hůlka, 2000; Jiránek and Kotrbatá, 2011; Szajerski and Zimny, 2020).

In addition to diffusion through the material, radon adsorption on the barrier and back diffusion can also occur. The effect of adsorption can be important in typical materials used for insulation as radon has good solubility in polymeric materials and very good solubility in materials containing more aromatic hydrocarbons. Back diffusion is a minor effect, mainly concerning materials with high thicknesses. If back diffusion is not determined, radon transport processes, which are mainly the radon exhalation rate, may be underestimated (Szajerski and Zimny, 2020).

Therefore, it is interesting to study materials that can be used as radon barriers. This work aims to validate an experimental methodology for radon diffusion coefficient determination based on the ISO/TS 11665-13:2017 norm (International Standardization Organization (ISO), 2017) comparing the values of the radon diffusion coefficient calculated with a control (Sisalex® 871). Then, the radon diffusion coefficients values obtained for LDPE, PP and PVC will be compared with those obtained from the literature. In addition, the methodology will be used to determine the diffusion coefficient of new materials (polystyrene [PS] and polymethyl methacrylate [PMMA]). Finally, the influence of thickness and chemical structure on the radon behaviour of the material will also be studied.

2. Materials and methods

2.1. Measuring system

The design of the measuring system is based on the guidelines established in the ISO/TS 11665-13:2017 norm "*Determination of the diffusion coefficient in waterproof materials: membrane two-side activity concentration test method*". The measuring system consists of two air-tight containers (source and receiver) one above the other with a volume (V) of $8.01 \cdot 10^{-4}$ m^3 each, made of stainless steel AISI316L and a thickness of 2 mm. The test barrier area (S) is $7.85 \cdot 10^{-3}$ m^2 and it is surrounded by a flange with screws to seal the barrier sample, which is located between the two containers. Each container has a couple of valves to attach the measuring instrument. The radon source is a sealed static pitchblende stone with a 1.469 kBq activity, which is located at the bottom of the source container.

The measuring devices that monitor radon activity are two continuous detectors (DURRIDGE RAD7), which have a maximum standard relative uncertainty of 10% and can measure the temperature and relative humidity of the air flowing into the detector. To keep a low relative humidity, a DURRIDGE DRYSTIK 144-ADS-3R unit is connected between the source chamber and the air inlet to one RAD7, while a desiccant unit with drierite is connected between the receiver chamber and the air inlet to the second RAD7. The tubes connecting the different devices are made of vinyl and have a thickness of 1.41 mm for the thin tubes and 1.82 mm for the thick ones. A closed-loop is maintained as the air that is removed from the container for radon

concentration analysis is returned to the container. Figure 1 shows a diagram and a picture of the experimental system set-up.

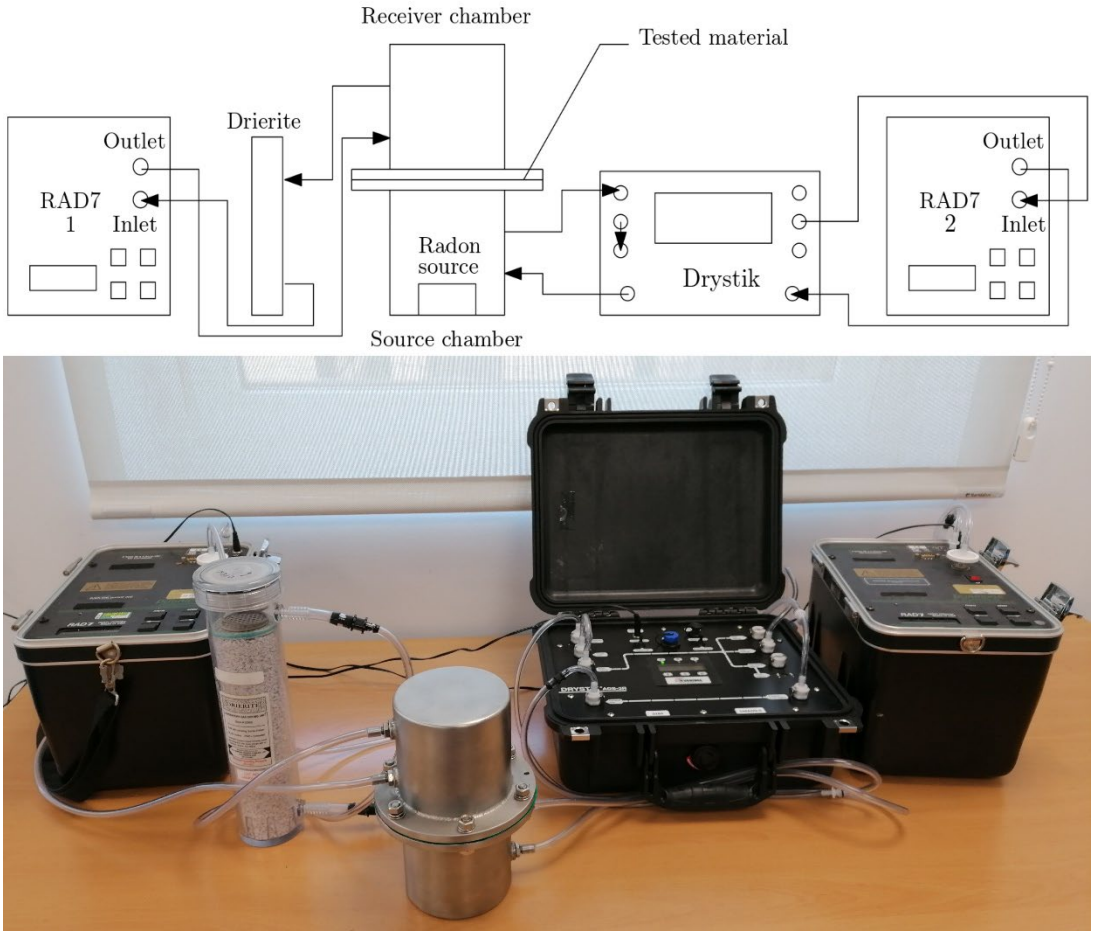

Figure 1. Experimental system set-up.

The RAD7 uses a solid-state alpha detector, which consists of a semiconductor material (silicon) capable of converting alpha radiation into an electrical signal. It registers the alpha spectrum of entering air and counts the alpha particles emitted by polonium-218 and polonium-214 (energy levels of 6.00 and 7.69 MeV). Via the pump contained in the monitor, the extracted air samples are drawn through a filter that does not allow the entry of radon progeny. Radon disintegrates inside the chamber as alpha particles are captured by the detector. The RAD7 provides the radon concentration in Bq/m^3 for each time interval that has been configured.

2.2. Diffusion coefficient calculation

The samples (tested material) are circular shaped with a diameter of $1.5 \cdot 10^{-1}$ m, equivalent to that of the chamber plus the flange, with 6 holes on the outside that match the screws for a better fit. Before the test, the thickness of each sample is measured with a micrometre (model 3006 from Baxlo Precision).

Figure 2. Radon accumulation containers

Then, the sample is placed between the source and receiver containers and the system is sealed with an O-ring joint and screws. The radon concentration in both the source and receiver container is registered at time intervals of 30 minutes during 48 hours.

Although the measurement method is based on the ISO/TS 11665-13:2017 norm, some variations have been made:

- The radon source is placed inside the source container instead of using an external radon source and recirculating air with a high radon concentration back to the source container. This results in a much faster achievement of the steady-state.
- Since a stable high radon concentration is achieved much earlier, the measurement time can be reduced to 48 h, which makes it possible to test different samples faster.
- The measurement takes place simultaneously in the source and receiver containers from the beginning, but the data from the last 24 hours (corresponding to the steady-state) are taken for the calculation of the radon diffusion coefficient because the minimum radon concentration in the source chamber required by the norm (between 300 and 500 kBq/m^3) has already been reached.

When the whole test is finished, both chambers are flushed, both RAD7 are purged and the tested material is placed after 15 minutes in a gamma spectrometry system with a NaI detector to determine if the material has adsorbed radon. The efficiency of the detector is 12.53%, the measurement time is 600 seconds and once the spectrum is displayed, two regions of interest (ROI) are selected corresponding to bismuth-214 and lead-214 peaks. The counts (cps) that are recorded under the areas of the two ROIs are registered.

With the recorded data, the diffusion coefficient is calculated following the procedure explained in Annex A.5 “Expression of results” of the ISO/TS 11665-13:2017 norm (International Standardization Organization (ISO), 2017), which is summarised in Figure 3.

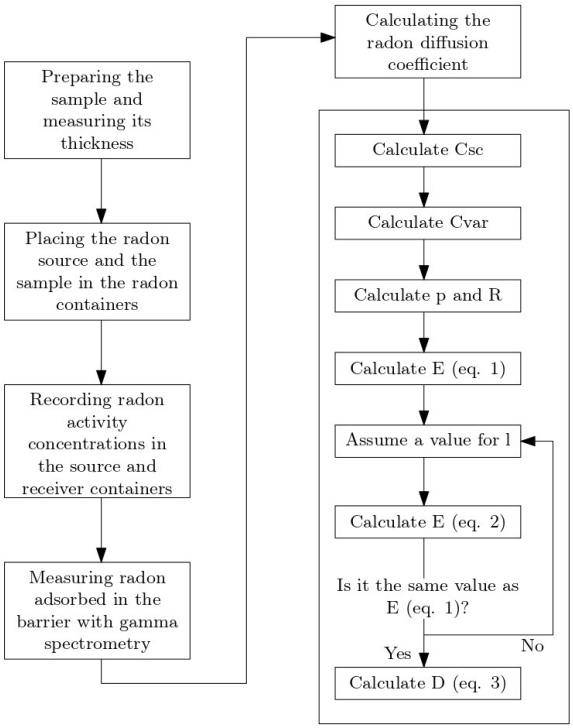

Figure 3. Flow chart of the measurement process and calculation of the radon diffusion coefficient

The average concentration, C_{SC}, is calculated from the source container data of the last 24 hours (Bq/m^3) and the coefficient of variation, C_{var}, calculated as the standard deviation of the data from the last 24 hours of the source chamber divided by C_{SC} (%). As the norm states: "the radon activity concentration in the source container shall be in the steady-state (the coefficient of variation for the data recorded during the last 24 h shall be lower than 10%), and at the same time it shall be higher than the minimum radon activity concentration". By fitting the data from the receiver container to a straight line, the slope, p, (Bq/m^{3}s) and the correlation coefficient (R^2) are calculated, considering that the slope must be significantly greater than zero and R^2 greater than 0.9. The next step is to calculate the diffusive radon exhalation (Bq/m^{2}s), E, following the equation (1),

$$E = p \cdot \frac{V}{S} \tag{1}$$

where V is the volume of the chamber (m^3) and S the test barrier area (m^2). With that calculated value and assuming a value for the radon diffusion length, l, (m), through an iterative process with the equation (2) the actual value of l is calculated.

$$E = \frac{2 \cdot C_{SC} \cdot l \cdot \lambda}{e^{d/l} - e^{-d/l}} \tag{2}$$

Where λ is the decay constant of radon ($2.11 \cdot 10^{-6}$ s^{-1}) and d is the thickness of the barrier (m). Finally, the diffusion coefficient, D, (m^2/s) is obtained with equation (3):

$$D = \lambda \cdot l^2 \tag{3}$$

2.3. Radon leakage test

A radon leakage test has been performed at the beginning of the tests to ensure that the measuring system is airtight. For this purpose, the receiver chamber is sealed underneath (where the sample would be) with a 2 mm thick stainless steel sheet after being filled with a radon concentration of 170 kBq/m^3. The decrease in radon concentration in the receiver container is recorded with the RAD7 and fits an exponential curve:

$$y = e^{(\lambda_l + \lambda_r) \cdot t} \tag{4}$$

where λ_l is the radon leakage rate (s^{-1}) and λ_r is the radon decay constant (s^{-1}). According to the ISO/TS 11665-13:2017 norm, if the radon leakage rate is lower than half the radon decay constant, the experimental set-up is appropriate for radon diffusion test measurement.

2.4. Radon barrier samples

The materials tested are polymeric (LDPE, PP, PVC, PS and PMMA) and composite (Sisalex® 871 [consisting of two layers of PE film, a polyester fibre inlay and a 0.02 mm thick aluminium layer]). Since the manufacturer indicates the value of the diffusion coefficient for the Sisalex membrane, this sample has been used as a control. LDPE, PP and PVC are polymeric materials widely studied and whose diffusion coefficient is measured in multiple works (see Section 3 – results), so these materials also help to validate the methodology comparing our results with those already published. Nevertheless, PS and PMMA are polymeric materials not studied, so their diffusion coefficient is unknown. However, this work demonstrates that these materials could also be suitable for manufacturing radon barriers.

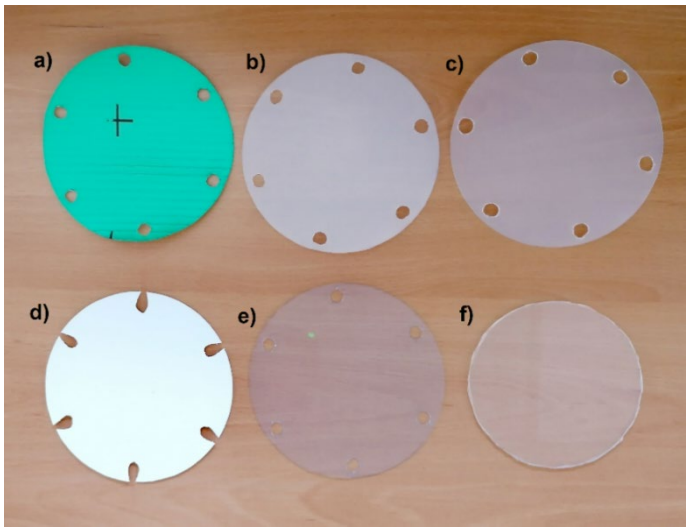

Figure 4. Radon barrier samples: a) Sisalex, b) LDPE, c) PP, d) PS, e) PVC, f) PMMA

For each material, the measurement of the diffusion coefficient has been done twice, so the graphs and calculations have been made for the average of the data from both measurements.

3. Results

3.1 Radon leakage test

Before starting the experiments, the radon leakage test was carried out as described in section 2.3., and the data is shown in Figure 5.

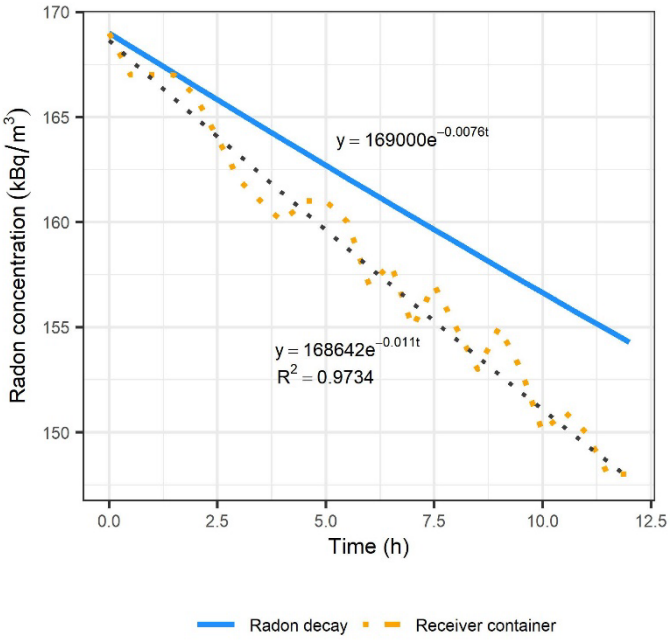

Figure 5. Radon leakage test performed in the receiver container

The solid curve represents the decrease in radon concentration if only radon decay exists, while the dotted curve shows the data collected in the receiver chamber with the RAD7. The dotted line represents the fit of the experimental data to an exponential curve. The difference between the decay curve and the one fitted from the experimental data is due to leakage, and it is possible to know its value as described in equation (4) from the time exponential (0.011 h^{-1}). Knowing that the radon decay constant is 0.0076 h^{-1}, the leakage is 0.0034 h^{-1}, which is lower than half the radon decay constant. Therefore, it can be considered that the leakage is negligible and can be assumed in the diffusion coefficient estimation tests carried out in this work.

3.2 Radon diffusion coefficient test

The RAD7 registers continuously the air temperature offering a mean airstream temperature of 17.20ºC with a variation of 1.55ºC during the tests conducted at the laboratory between December 2020 and February 2021. The mean radon concentration of two measurements registered during the tests through both RAD7 is presented in Figure 6.

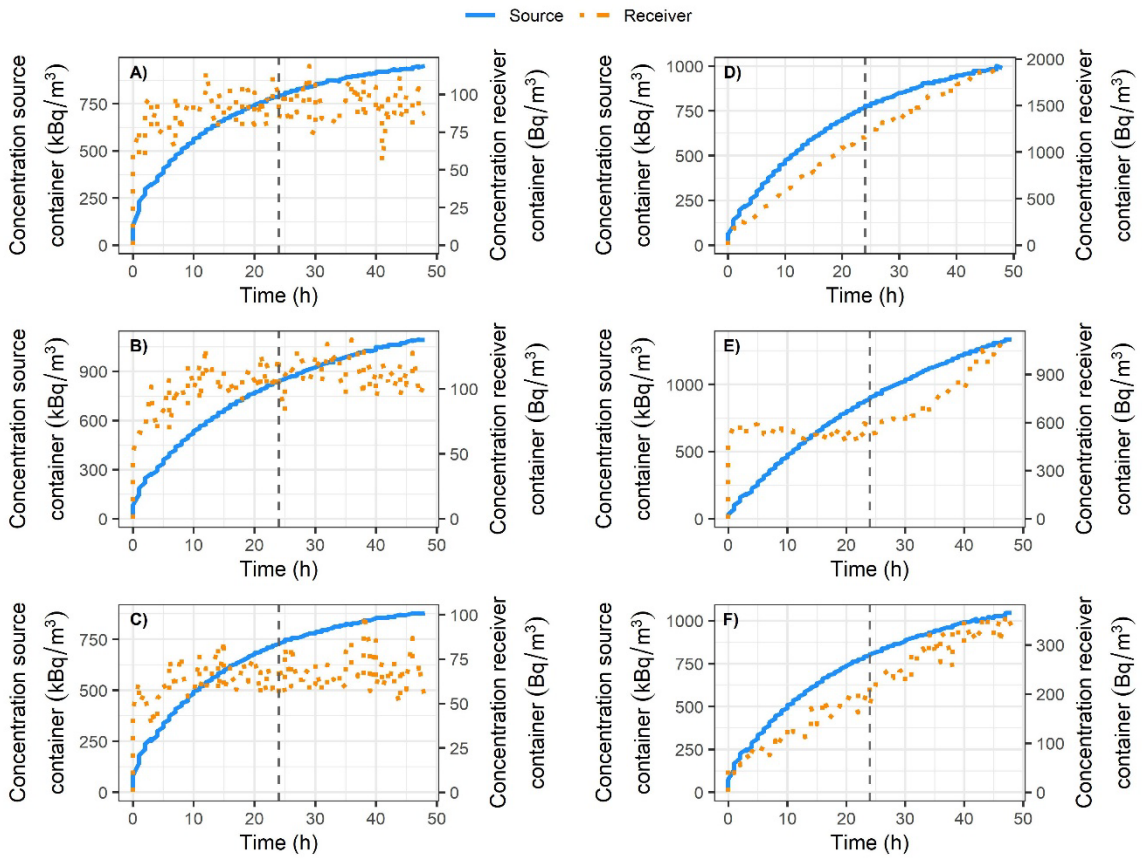

Figure 6. Radon concentration in the source chamber (solid line) and the receiver chamber (dashed line). The materials tested are A) PP, B) PVC, C) PMMA, D) Sisalex, E) LDPE and F) PS

Figure 6 shows the evolution of the radon concentration in both the source chamber and the receiver chamber over the test time. Since the concentration in the receiver chamber is much lower, the values should be read on the right y-axis. The radon concentration in the source chamber is between 800 and 1300 kBq/m^3 at the end of the experiment, while the concentration in the receiver chamber does not exceed 2 kBq/m^3 (being in most cases around 100 Bq/m^3). It is observed that for Sisalex (D), LDPE (E) and PS (F) radon continues to accumulate in the receiver chamber and the curve increases throughout the test time, while for PP (A), PVC (B) and PMMA (C) the radon concentration remains more or less constant from 50000 seconds onwards (approximately 14 hours).

Using those concentrations (C_{SC}) values and following the mathematical procedure explained in section 2.2, the radon diffusion coefficient of each material is calculated. These results are shown in Table 1.

Table 1. Values of the parameters calculated to obtain the radon diffusion coefficient of each material tested.

Material	C_{SC} (kBq/m^3)	C_{var} (%)	p (Bq/m^{3}s)	R^2	d (mm)	l (mm)	D (m^2/s)
A) PP	877.11±41.82	4.77	$2.354 \cdot 10^{-5}$	$1.651 \cdot 10^{-3}$	0.54 ± 0.005	0.144	$4.35 \pm 0.53 \cdot 10^{-14}$
B) PVC	963.03±66.45	6.90	$1.043 \cdot 10^{-5}$	$2.827 \cdot 10^{-4}$	0.50 ± 0.000	0.130	$3.55 \pm 0.43 \cdot 10^{-14}$
C) PMMA	819.72±44.34	5.41	$1.225 \cdot 10^{-5}$	$8.929 \cdot 10^{-4}$	2.15 ± 0.013	0.440	$4.07 \pm 0.49 \cdot 10^{-13}$
D) Sisalex	900.38±65.28	7.25	$8.789 \cdot 10^{-3}$	0.9787	0.53 ± 0.005	0.544	$6.21 \pm 0.75 \cdot 10^{-13}$
E) LDPE	1160.41±115.86	9.98	$7.372 \cdot 10^{-3}$	0.9436	1.18 ± 0.009	0.739	$1.15 \pm 0.14 \cdot 10^{-12}$
F) PS	940.23±70.20	7.47	$1.647 \cdot 10^{-3}$	0.7969	1.16 ± 0.029	0.486	$4.96 \pm 0.60 \cdot 10^{-13}$

The C_{SC} values are above the minimum concentration value required by the ISO norm, which for the materials and thicknesses tested would be between 300 and 500 kBq/m^3, and the C_{var} values are all below 10%, so the steady-state conditions required by the ISO norm are met.

Comparing the graphs in Figure 6 with the numerical results, it can be seen that there are two groups of materials: there is one group where *p* is higher because radon concentration increases over time and, consequently, the R^2 fit is better, having values of the diffusion coefficients medium-high (materials D, E and F); while there is another group in which the data from the receiver container do not follow a linear trend, and for them, the *p* is lower, the linear fit (R^2) is not adequate for the experimental data obtained and the diffusion coefficients are low for A and B, being medium for C. In addition, the low radon concentrations in the receiver chamber show a large oscillation, which has an impact on the low R^2 values.

Regarding the radon diffusion length (*l*), this is the distance that radon travels due to diffusion when decay occurs *e* times; ideally, this distance should not exceed the thickness of the barrier, as radon would then disintegrate inside the barrier and would not pass through it. Comparing the values of the thickness (*d*) and the radon diffusion length (*l*), it is observed that for all cases the thickness is greater, which causes the radon to decay within the material before passing through it.

The diffusion coefficient value calculated for Sisalex ($6.21 \pm 0.75 \cdot 10^{-13}$ m^2/s) can be compared with the value provided by the manufacturer ($1.4 \cdot 10^{-13}$ m^2/s) and it can be seen that they are in accordance with a variation of 77.4%.

Table 2 also shows the percentage reduction in radon concentration obtained from the application of the materials. The radon reduction is calculated as follows:

$$Reduction\ (\%) = \frac{C_{SC} - C_{RC}}{C_{SC}} \cdot 100 \quad (5)$$

where C_{SC} is the mean radon concentration in the source chamber of the last 24 hours and C_{RC} is the mean radon concentration in the receiver chamber in the last 24 hours, both measured in Bq/m^3. In all cases, a reduction of more than 99% is achieved, demonstrating the effectiveness of using radon barriers exposed to elevated concentrations of radon.

Table 2. Radon reduction for each barrier studied

Material	C_{SC} (Bq/m^3)	C_{RC} (Bq/m^3)	Reduction (%)
PP	877107.14±41818.79	92.84±12.79	99.99±0.0015
PVC	963026.32±66447.19	114.68±12.41	99.99±0.0015
PMMA	819718.75±44342.95	67.27±10.33	99.99±0.0013
Sisalex	900375.00±65278.50	1575.50±223.87	99.83±0.0127
LDPE	1160409.09±115862.60	813.49±175.48	99.93±0.0084
PS	940229.17±70203.80	291.26±46.50	99.97±0.0031

3.3 Comparison of radon diffusion coefficients

The diffusion coefficient of a wide range of polymeric materials has been measured in different studies under different experimental conditions. Table 3 shows the radon diffusion coefficient measured in other research for the most common plastics, i.e., high density (HD), low density (LD) and very low density (VLD) PE, PP and PVC as well as the values obtained in this work to compare them. In some cases, if stated, the thickness of the membrane is also shown.

Table 3. Radon diffusion coefficients for some common materials used as a radon barrier.

Material	Radon diffusion coefficient, D (m^2/s)	Thickness (mm)	Reference
HDPE	$6.2 \cdot 10^{-12}$		(Jiránek and Hůlka, 2001)
	$7.43 \cdot 10^{-12}$	0.330	(Singh et al., 2005)
	$5.8 \cdot 10^{-12}$		(Jiránek et al., 2008)
	$6.1 \cdot 10^{-12}$		(Jiránek and Kotrbatá, 2011)
	$(4.24 \pm 0.16) \cdot 10^{-12}$		(Papp and Cosma, 2015)
	$1.62 \cdot 10^{-12}$		(Georgiev et al., 2019)
LDPE	$1.64 \cdot 10^{-11}$	0.152	(Daoud and Renken, 2001)
	$1.24 \cdot 10^{-11}$		(Jiránek and Hůlka, 2001)

	$(1.8\text{-}3.3)\cdot10^{-11}$	0.025 – 0.187	(Singh et al., 2005)
	$1.9\cdot10^{-11}$		(Jiránek et al., 2008)
	$1.8\cdot10^{-11}$		(Jiránek and Kotrbatá, 2011)
	$1.93\cdot10^{-10}$		(Kumar and Chauhan, 2014)
	$(6.47 \pm 0.17)\cdot10^{-12}$		(Papp and Cosma, 2015)
	$3.07\cdot10^{-12}$		(Georgiev et al., 2019)
	$1.15 \pm 0.14 \cdot10^{-12}$	1.18	This work
VLDPE	$(7.36 \pm 0.22)\cdot10^{-12}$		(Papp and Cosma, 2015)
PP	$2.7\cdot10^{-13}$		(Jiránek and Hůlka, 2001)
	$(1.1\text{-}1.4)\cdot10^{-11}$	0.030 – 0.055	(Singh et al., 2005)
	$1.0\cdot10^{-11}$		(Jiránek et al., 2008)
	$6.4\cdot10^{-12}$		(Jiránek and Kotrbatá, 2011)
	$8.2\cdot10^{-14}$		(Georgiev et al., 2019)
	$4.35 \pm 0.53 \cdot10^{-14}$	0.54	This work
PVC	$5.0\cdot10^{-11}$		(Jha et al., 1982)
	$5.8\cdot10^{-13}$		(Hafez and Somogyi, 1986)
	$6.1\cdot10^{-13}$		
	$7.81\cdot10^{-12}$		(Jiránek and Hůlka, 2001)
	$1.5\cdot10^{-11}$	0.070	(Singh et al., 2005)
	$1.3\cdot10^{-11}$		(Jiránek et al., 2008)
	$1.8\cdot10^{-11}$		(Jiránek and Kotrbatá, 2011)
	$3.55 \pm 0.43 \cdot10^{-14}$	0.50	This work
PS	$4.96 \pm 0.60 \cdot10^{-13}$	1.160	This work
PMMA	$4.07 \pm 0.49 \cdot10^{-13}$	2.150	This work

From the literature references on radon diffusion coefficients, PE is the most studied polymeric material and a difference can be observed between the diffusion coefficient values for HDPE and LDPE, being lower for HDPE, therefore being the better of the two. Comparing all the diffusion coefficient values listed, the best materials are PP and PVC, having both radon diffusion coefficients in the order of 10^{-14} m^2/s.

The diffusion coefficient values obtained in this work are all between 10^{-12} and 10^{-14} m^2/s, which is in accordance with the range of the literature. These are very good values according to (Jiránek et al., 2008) and make the materials suitable for their use in preventing the flow of radon and being used as a barrier in buildings.

The differences between the values are due to several reasons, among others, the test conditions, the initial radon concentration and also the fact that the materials are not pure but contain different additives, which can change over time both in composition and quantity due to changes in production (Jiránek and Kotrbatá, 2011). Therefore, as the diffusion coefficient values calculated with the proposed method are comparable with those established in the literature, the modifications applied to the ISO measurement method can be validated. Thus, the radon diffusion coefficient calculated for PS and PMMA can be taken as a reference for future research, as it was a previously unknown value.

3.4 Influence of chemical composition and thickness on radon diffusion

Radon adsorbed on the barrier materials after the tests has also been measured, the net counts (cps) of gamma radon peaks obtained with the gamma spectrometer are shown in Table 4, where other parameters of the samples such as the chemical composition are also presented.

Table 4. Parameters of the samples studied

Material	Counts (cps)	Thickness (mm)	Chemical composition
Sisalex	27.6 ± 5.7	0.53	PE – glass fiber – Al – PE
LDPE	44.5 ± 6.0	1.18	$-(C_2H_4)_n-$
PP	30.4 ± 6.6	0.54	$-(C_3H_6)_n-$

PS	35.9 ± 7.3	1.16	– $(C_8H_8)_n$ –
PVC	30.2 ± 8.7	0.50	– $(C_2H_3Cl)_n$ –
PMMA	24.6 ± 8.0	2.15	– $(C_5H_8O_2)_n$ –

As can be seen in Table 4, all materials adsorb radon during exposure as the counts per second recorded vary between 24.6 and 44.5 cps. Sisalex, PP and PVC have a very similar number of counts (between 27.6 and 30.4) and also have a comparable thickness (between 0.5 and 0.54 mm), while PS and PE, which have a higher thickness (between 1.16 and 1.18 mm) and have higher counts (between 35.9 and 44.5). Therefore, the relationship between the counts adsorbed during radon exposure and the thickness of the membrane has been analysed and a positive trend is observed so the thicker the material, the more counts are adsorbed for all materials tested except for PMMA, as it is shown in Figure 7. In the case of PMMA, counts are very low (24.6 cps) even if the thickness of the material is the highest. PMMA has in its composition oxygen and according to (Gong et al., 2020), radon forms weak interactions with functional groups containing oxygen so radon tends to move away from these surfaces. This may explain why the counts are so low despite the high thickness of the material.

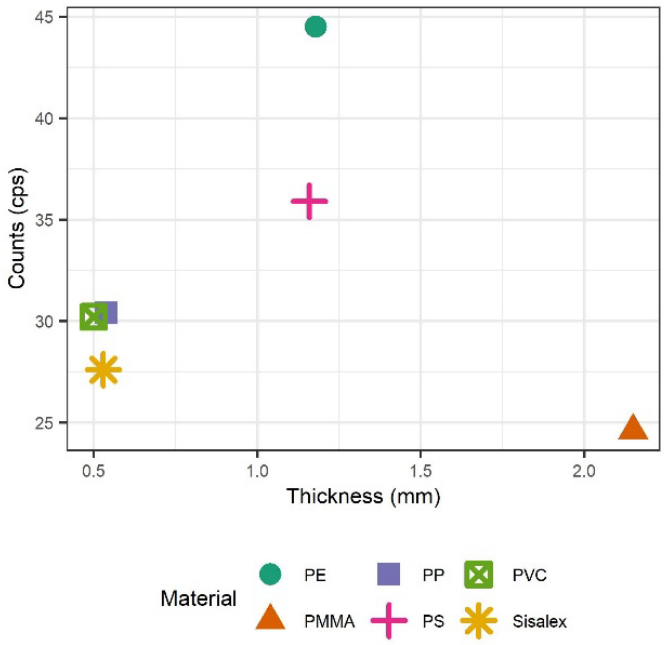

Figure 7. Radon adsorbed by each material compared to its thickness

4. Conclusions

Two main modifications have been made to the experimental methodology compared to the ISO/TS 11665-13:2017 norm: the introduction of the radon source inside the source container and the reduction of the measurement time to 48 hours. Having the radon source inside the chamber allows the radon to accumulate inside the chamber much faster and the steady-state of the radon concentration in the source chamber to be reached earlier, allowing the reduction of the measurement time mentioned above.

The measurement of the radon diffusion coefficient of different polymeric materials (LDPE, PP, and PVC), as well as a commercial anti-radon membrane, and its comparison with the values of the studies of other authors and the manufacturer allows validating the developed methodology since the diffusion coefficients provided are comparable with those of the literature.

The diffusion coefficients of PS and PMMA, non-published polymeric materials, indicate that both materials are suitable to be used as radon barriers.

Among all the materials studied, some behave as a filter reducing the radon concentration but allowing the gas to pass through the membrane (Sisalex, PE and PS), while other materials act as a barrier and stabilise the radon concentration detected on the other side of the material around very low values (PP, PVC and PMMA). In all cases, the reduction of radon concentration is above 99% for radon concentrations higher than 800 kBq/m^3.

The thickness of the barriers affects the radon adsorption. Via the counts recorded in a gamma spectrometer with a NaI detector, the amount of radon adsorbed by the material is determined and a positive relationship is found with the thickness, i.e., as the thickness increases, radon adsorption increases. A thickness of 0.5 mm is enough to ensure that the radon concentration has decreased by *e* times, that is, by a little more than half.

The chemical composition of the materials influences their ability to adsorb radon passing through them so that oxygen functional groups cause radon to create weaker bonds and try to stay away from these surfaces.

Continuing with this line of research and knowing that polymeric materials can be used as radon barriers, which in many countries are the prevention method established for new buildings, it is intended to study the mechanical and ageing resistance of these materials and, eventually, to study how the radon diffusion coefficient is affected by combining several of these materials in layers.

5. Acknowledgements

This work is financed by the Operational Programme 2014-2020 Comunitat Valenciana of the European Regional Development Fund and the Generalitat Valenciana under the projects *Bioingeniería de las Radiaciones Ionizantes. Biorad (PROMETEO/2018/035)* and *MEMO RADION (IDIFEDER/2018/038)* and also by the European Social Fund through *Ayuda Garantía Juvenil Gva-Capacidades de detección del Laboratorio de Radiaciones Ionizantes (GJIDI/2018/A/016)*.

The authors would like to thank Radiansa for providing some of the material used in the tests and Gdes for their collaboration during the research.

6. References

Brill, A.B., Becker, D. V., Donahoe, K., Goldsmith, S.J., Greenspan, B., Kase, K., Royal, H., Silberstein, E.B., Webster, E.W., 1994. Radon update: Facts concerning environmental radon: Levels, mitigation strategies, dosimetry, effects and guidelines. J. Nucl. Med. 35, 368–385.

Council of the European Union, 2013. Council Directive 2013/59/Euratom. Off. J. Eur. Union. https://doi.org/doi:10.3000/19770677.L_2013.124.eng

Daoud, W.Z., Renken, K.J., 2001. Laboratory assessment of flexible thin-film membranes as a passive barrier to radon gas diffusion. Sci. Total Environ. 272, 127–135. https://doi.org/10.1016/S0048-9697(01)00676-3

Donohoe, K., Royal, H., 1996. Importance of radon as a threat to public health. Otolaryngol. - Head Neck Surg. 114, 271–276. https://doi.org/10.1016/S0194-5998(96)70182-1

Fucic, A., Fucic, L., Katic, J., Stojković, R., Gamulin, M., Seferović, E., 2011. Radiochemical indoor environment and possible health risks in current building technology. Build. Environ. 46, 2609–2614. https://doi.org/10.1016/j.buildenv.2011.06.020

Gandolfo, G., Lepore, L., Pepperosa, A., Remetti, R., Franci, D., 2017. Radiation protection considerations on radon and building materials radioactivity in Near Zero Energy Buildings. Energy Procedia 140, 13–22. https://doi.org/10.1016/j.egypro.2017.11.119

Georgiev, S., Mitev, K., Dutsov, C., Boshkova, T., Dimitrova, I., 2019. Partition coefficients and diffusion lengths of 222Rn in some polymers at different temperatures. Int. J. Environ. Res. Public Health 16, 4523. https://doi.org/10.3390/ijerph16224523

Gong, M.S., Cha, J.R., Hong, S.M., Lee, C., Lee, D.H., Joo, S.W., 2020. Roll-to-roll graphene oxide radon barrier membranes. J. Hazard. Mater. 383, 121148.

https://doi.org/10.1016/j.jhazmat.2019.121148

Hafez, A.F., Somogyi, G., 1986. Determination of radon and thoron permeability through some plastics by track technique. Int. J. Radiat. Appl. Instrumentation. Part D. Nucl. Tracks Radiat. Meas. 12, 697–700. https://doi.org/10.1016/1359-0189(86)90682-5

Holmgren, O., Arvela, H., 2012. Assessment of current techniques used for reduction of indoor radon concentration in existing and new houses in European countries. STUK-A251. Helsinki.

International Standardization Organization (ISO), 2017. ISO/TS 11665-13:2017 - Measurement of radioactivity in the environment — Air: radon 222 — Part 13: Determination of the diffusion coefficient in waterproof materials: membrane two-side activity concentration test method.

Jha, G., Raghavayya, M., Padmanabhan, N., 1982. Radon permeability of some membranes. Health Phys. 42, 723–725.

Jiránek, M., Hůlka, J., 2001. Applicability of various insulating materials for radon barriers. Sci. Total Environ. 272, 79–84. https://doi.org/10.1016/S0048-9697(01)00668-4

Jiránek, M., Hůlka, J., 2000. Radon diffusion coefficient in radon-proof membranes - determination and applicability for the design of radon barriers. Int. J. Archit. Sci. 1, 149–155.

Jiránek, M., Kotrbatá, M., 2011. Radon diffusion coefficients in 360 waterproof materials of different chemical composition. Radiat. Prot. Dosimetry 145, 178–183. https://doi.org/10.1093/rpd/ncr043

Jiránek, M., Rovenská, K., Fronka, A., 2008. Radon diffusion coefficient - A material property determining the applicability of waterproof membranes as radon barriers, in: Proceedings of the American Association of Radon Scientists and Technologists. Las Vegas NV, p. 8.

Khan, S.M., Gomes, J., Krewski, D.R., 2019. Radon interventions around the globe: A systematic review. Heliyon 5, 1–10. https://doi.org/10.1016/j.heliyon.2019.e01737

Kumar, A., Chauhan, R.P., 2014. Active and passive measurements of radon diffusion coefficient from building construction materials. Environ. Earth Sci. 72, 251–257. https://doi.org/10.1007/s12665-013-2951-5

Pacheco-Torgal, F., 2012. Indoor radon: An overview on a perennial problem. Build. Environ. 58, 270–277. https://doi.org/10.1016/j.buildenv.2012.08.004

Papp, B., Cosma, C., 2015. Methods for measuring radon diffusion parameter of waterproof membranes. J. Radioanal. Nucl. Chem. 303, 1663–1669. https://doi.org/10.1007/s10967-014-3761-2

Singh, S., Singh, J., Singh, L., 2005. The study of some common plaster coating materials and plastic foils as a barrier to radon. Radiat. Meas. 40, 673–677. https://doi.org/10.1016/j.radmeas.2005.05.006

Spanish Ministry of Public Works and Transport, 2019. Basic document DB-HS on health and safety. Section 6 - Protection against radon, in: Technical Building Code. pp. 138–184.

Szajerski, P., Zimny, A., 2020. Numerical analysis and modeling of two-loop experimental setup for measurements of radon diffusion rate through building and insulation materials. Environ. Pollut. 256, 113393. https://doi.org/10.1016/j.envpol.2019.113393

World Health Organization, 2009. Indoor radon: A public health perspective. Int. J. Environ. Stud. 67, 108. https://doi.org/10.1080/00207230903556771