Determination of the radon diffusion coefficient of thin polyethene and aluminium foils used as single or multilayer configuration barriers

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Abstract

Radon is a radioactive noble gas exhaled from the soil that can reach high concentrations in enclosed spaces. As elevated concentrations cause serious health problems, legislation has been put in place in many countries to regulate the limit concentration and even establish mitigation techniques. One of the most effective techniques for new buildings is the installation of radon barriers. The most important parameter determining whether the barrier is adequate to protect against radon is the diffusion coefficient, whose measurement methodology is standardised in ISO/TS 11665-13:2017. This work applies a previously tested modification of this standard to calculate the radon diffusion coefficient of different materials used as single or multilayer form barriers. Given that there are wasted laminated materials, composed of polymeric materials and aluminium, which are difficult to recycle, a preliminary study of the possible effectiveness of these materials as radon barriers will be carried out using these materials separately or in combination. The materials to be tested are 10-micron sheets of polyethene (PE) and 15-micron sheets of aluminium (Al), testing in each case one, two and three layers of each material. In addition, combinations of the two materials, i.e., PE-Al-PE and PE-Al-PE-Al-PE, are also studied. The diffusion coefficients obtained vary around $2 \cdot 10^{-12}$ m^2/s for PE and around $1 \cdot 10^{-13}$ m^2/s in the case of Al. The combination of both materials improved results obtained for single-materials barriers giving a diffusion coefficient between 10^{-13} and 10^{-14} m^2/s. Radon reductions achieved range from 70-87.5% for PE to more than 98% for Al and the materials combinations. The excellent radon shielding capacity of aluminium is observed, which grows with increasing material thickness. Furthermore, it can be stated that the use of multilayer materials of different nature is very effective in reducing the radon concentration reaching an enclosed space and the use of aluminium in some of the layers is essential to achieve a more significant shielding effect.

Keywords: radon, diffusion coefficient, radon barrier, polyethene, aluminium

1. Introduction

Radon is a gas considered carcinogenic by the World Health Organization (World Health Organization, 2009). It is exhaled from soils with high radium content, like granite, sedimentary rocks or some types of sands, and enters buildings through cracks or poorly sealed joints, where it accumulates to high concentrations (Khan et al., 2019). Indoor radon concentration limits exist in many countries; in particular, the Directive 59/2013/Euratom sets a limit of 300 Bq/m^3 for closed spaces in the European Union (Council of the European Union, 2013).

To comply with this limit, the Spanish Technical Building Code requires the installation of radon barriers in new buildings to be constructed in areas where high indoor radon concentrations are expected. According to the characteristics described in the Technical Building Code, barriers must be continuous, sealed, and have durability appropriate to the useful life of the building. In addition, the minimum thickness should be 2 mm and have a radon diffusion coefficient of less than 10^{-11} m^2/s (Spanish Ministry of Public Works and Transport, 2019).

This coefficient defines the radon transport through the barrier, so it is a good indication of the adequacy of a barrier to protect against radon (Jiránek et al., 2008). ISO/TS 11665-13:2017 establishes an experimental methodology and a standard calculation process of this coefficient (International Standardization Organization (ISO), 2017). The radon diffusion coefficient varies over a wide range depending on the material from which the barrier is made. Among the lowest values (around 10^{-15} m^2/s) are barriers with some aluminium foil and ethylene vinyl acetate (EVA), while the highest values (around 10^{-8} m^2/s) are obtained for rubber membranes and polymer coatings. Common polymeric materials like polyethene (PE), polypropylene (PP) or polyvinyl chloride (PVC) have a radon diffusion coefficient between 10^{-11} and 10^{-13} m^2/s (Jiránek and Kotrbatá, 2011).

Nowadays, there are commercially available anti-radon barriers that combine different polymeric materials with aluminium, with good results (Chova, n.d.; Radiansa, n.d.). This kind of composition is also used for laminated materials in packaging. However, this kind of material represents an environmental problem because it is hard to recycle due to the different melting points between the polymeric materials of which they are composed, preventing them from being reprocessed by extrusion. Many of the wastes that present this problem are composed of PE, PP, PVC and aluminium and have a thickness between 40 and 120 μm. Therefore, this work proposes a preliminary study to verify the effectiveness as anti-radon barriers of single or combined materials similar to laminated materials, in the form of monolayer or multilayer, as this is the usual configuration of many commercial wastes that cannot be recycled at present.

2. Materials and methods

2.1. Measuring system

The measurement system is designed according to ISO/TS 11665-13:2017. It consists of two sealed chambers between which the studied barrier is placed. They are made of AISI 316L stainless steel with a thickness of 2 mm and have a volume of 0.801 l each and a test area of 78.5 cm^2. The radon source, a sealed pitchblende stone with an activity of 1.469 kBq, is placed inside the source container.

Two continuous detectors (Durridge RAD7), with a maximum standard relative uncertainty of 10%, record radon concentration in both chambers. Two desiccant units (Durridge Drystik 144-ADS-3R and drierite) maintain low relative humidity in the air inlet of the RAD7. The devices are connected with vinyl tubes 1.41 and 1.81 mm thick. The air extracted from the chamber for radon analysis is reintroduced into the chamber, thus maintaining a closed cycle. A more detailed diagram of the experimental setup can be found in (Ruvira et al., 2022).

2.2. Diffusion coefficient calculation

The samples (tested material) have the same diameter as the chambers (15 cm), with six holes on the outer perimeter to fix them with the screws. Prior to the test, the thickness of the samples is measured with a micrometre (model 3006 from Baxlo Precision, 10 μm accuracy). The characteristics of the samples tested, i. e., the type of material, the number of layers and the total thickness, are described in Table 1.

Table 1. Description of the materials tested

Material	Nº layers	Thickness (μm)
PE	1	10 ± 4.96
	2	20 ± 5.18
	3	30 ± 5.18
Al	1	15 ± 7.56
	2	30 ± 5.18
	3	45 ± 5.35
Combination	PE – Al – PE	35 ± 7.29
	PE – Al – PE – Al – PE	60 ± 6.23

The test starts when the sample is placed between both containers, and the setup is sealed with a 2 mm diameter O-ring made of rubber (SBR) and screws. The radon concentration inside the chambers is recorded at intervals of 30 min for 48 h following the methodology previously described in (Ruvira et al., 2022). After this time, the chambers are flushed, and the devices are purged. The sample is placed immediately in a gamma spectrometer with a NaI detector to determine if the material has adsorbed radon using the Pb-214 and Bi-214 peaks. For comparison, a blank consisting of virgin material that has not been exposed to the radon source has been included.

The calculation process for the steady state is explained in Annex A.5 "Expression of results" of the ISO/TS 11665-13:2017 norm (International Standardization Organization (ISO), 2017) and in (Ruvira et al., 2022).

The diffusion coefficient measurement has been done twice for each material and number of layers, so the graphs and calculations have been made for the average data from both measurements. The methodology of the radon leakage test is described in (Ruvira et al., 2022).

3. Results

Before starting the experiments, a radon leakage test was carried out. The leakage rate obtained is 0.0034 h^{-1}, which is lower than the limit value given in the ISO norm; leakage can therefore be assumed to be negligible.

The RAD7 registers the air temperature offering a mean airstream temperature of 17.95 ºC with a variation of 1.00 ºC during the tests conducted at the laboratory between February and April 2021.

3.1 Radon diffusion coefficient test of the PE foils

Figure 1 shows the evolution of the radon concentration in both the source chamber and the receiver chamber over the test time. The radon concentration in the source chamber is between 700 and 800 kBq/m^3 at the end of the experiment, and the concentration in the receiver chamber varies between 100 and 200 kBq/m^3. However, the more layers are added, the better the radon is slowed down, and the radon concentration in the receiver chamber decreases. Measurements made with the RAD7 have been compared with another detector (Radon Scout, Sarad) and the results show the same trend in the measurement range tested.

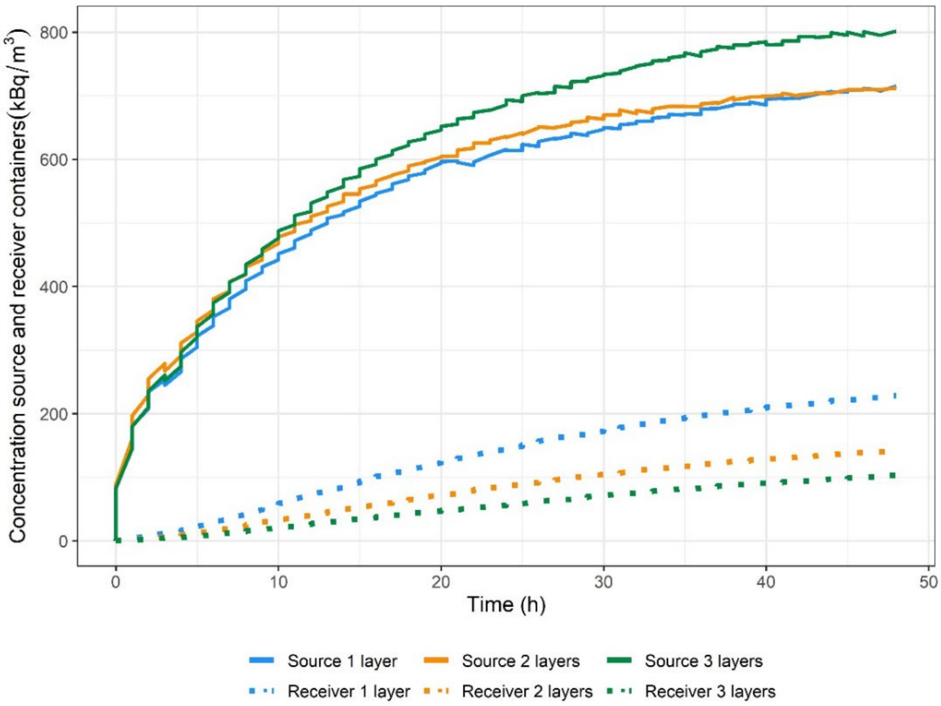

Figure 1. Radon concentration in the source chamber (solid line) and the receiver chamber (dashed line) for PE foils using 1, 2 and 3 layers

Table 2 shows the mean radon concentration in the source and receiver chamber in the last 24 h (C_{SC} and C_{RC}, respectively), the radon reduction percentage achieved, the radon diffusion length (*l*) and the radon diffusion coefficient (*D*). The radon diffusion length indicates the distance the radon travels inside the barrier until it disintegrates *e* times. The radon reduction is calculated as follows:

$$Reduction\ (\%) = \frac{C_{SC} - C_{RC}}{C_{SC}} \cdot 100 \qquad (1)$$

Table 2. Values of the parameters calculated to obtain the radon diffusion coefficient of PE foils

Nº layers (µm)	C_{SC} (kBq/m^3)	C_{RC} (kBq/m^3)	Reduction (%)	l (µm)	D (m^2/s)
1 (10 ± 4.96)	671.70 ± 29.82	193.27 ± 24.25	71.23	831	$(1.45 \pm 0.18)\cdot 10^{-12}$
2 (20 ± 5.18)	683.05 ± 22.17	117.77 ± 16.34	82.76	958	$(1.93 \pm 0.23)\cdot 10^{-12}$
3 (30 ± 5.18)	759.22 ± 33.85	82.34 ± 13.48	89.16	1010	$(2.14 \pm 0.26)\cdot 10^{-12}$

According to the results presented in Table 2, as more layers of PE are used, more radon concentrates in the source chamber, and less can reach the receiver chamber; thus, the radon reduction increases with more layers of the barrier. The radon diffusion length and the radon diffusion coefficient barely change for all the samples tested. The radon diffusion length is greater than the thickness of the foils, so much radon travels through the barrier until it disintegrates. The diffusion coefficient is in the order of 10^{-12} m^2/s, which is a typical value for PE (Jiránek et al., 2008; Jiránek and Kotrbatá, 2011).

3.2 Radon diffusion coefficient test of the Al foils

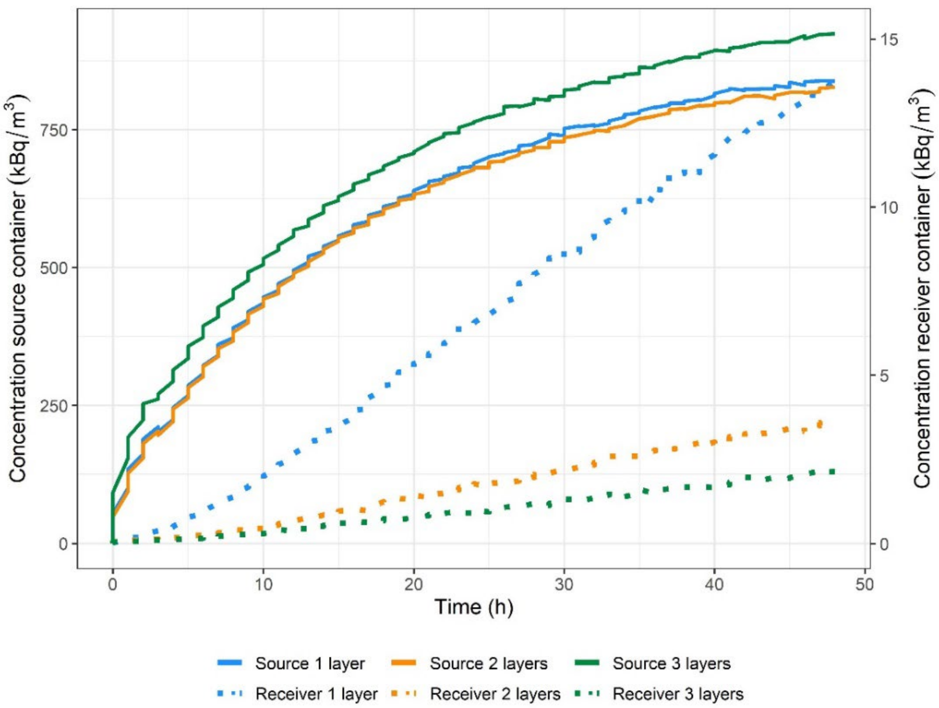

Figure 2. Radon concentration in the source chamber (solid line) and the receiver chamber (dashed line) for Al foils using 1, 2 and 3 layers

Figure 2 shows the evolution of the radon concentration in both chambers during the test. As the concentration in the receiver chamber is significantly lower than in the source chamber, the values have to be read on the right Y-axis. For Al foils, the radon concentration in the source chamber varies from 800 to 900 kBq/m^3 at the end of the test. However, the concentration in the receiver chamber ranges between 2.5 to 4 kBq/m^3 except for one Al layer, reaching up to 14 kBq/m^3 at the end of the experiment. Although the radon accumulating in the receiver chamber is much lower than in the case of PE, it is still a relatively high concentration. In addition, there is a significant reduction in concentration when going from one to two or three aluminium layers.

Table 3. Values of the parameters calculated to obtain the radon diffusion coefficient of Al foils

Nº layers (µm)	C_{SC} (kBq/m^3)	C_{RC} (kBq/m^3)	Reduction (%)	l (µm)	D (m^2/s)
1 (15 ± 7.56)	780.50 ± 45.18	10.33 ± 2.11	98.68	279	$(1.64 \pm 0.20)\cdot10^{-13}$
2 (30 ± 5.18)	766.52 ± 44.08	2.69 ± 0.57	99.65	208	$(9.09 \pm 1.10)\cdot10^{-14}$
3 (45 ± 5.35)	857.57 ± 48.14	1.55 ± 0.35	99.82	189	$(7.50 \pm 0.91)\cdot10^{-14}$

In Table 3, it can be seen that, as with PE, the more Al layers used, the higher the concentration reached in the source chamber and the less radon passes to the receiver chamber. The radon diffusion length and the diffusion coefficient hardly change with more layers. The diffusion length has a value between 189 and 279 µm, while the diffusion coefficient is around 10^{-13} m^2/s, which indicates that Al is a material with low radon permeability. The thickness increase improves the reduction in radon concentration, albeit slightly, as the difference in concentrations between the two chambers is large.

Comparing the results of PE and Al, the radon reduction achieved with a single layer of Al (98.68%) is much better than that achieved with three layers of PE (89.16%). The diffusion length for Al (around 200 µm) is much lower than the values obtained for PE (approximately 900 µm). The diffusion coefficient for Al is around an order of magnitude smaller than for PE, which means Al keeps radon from passing through better than the polymeric material.

3.3 Radon diffusion coefficient test of the PE + Al foils

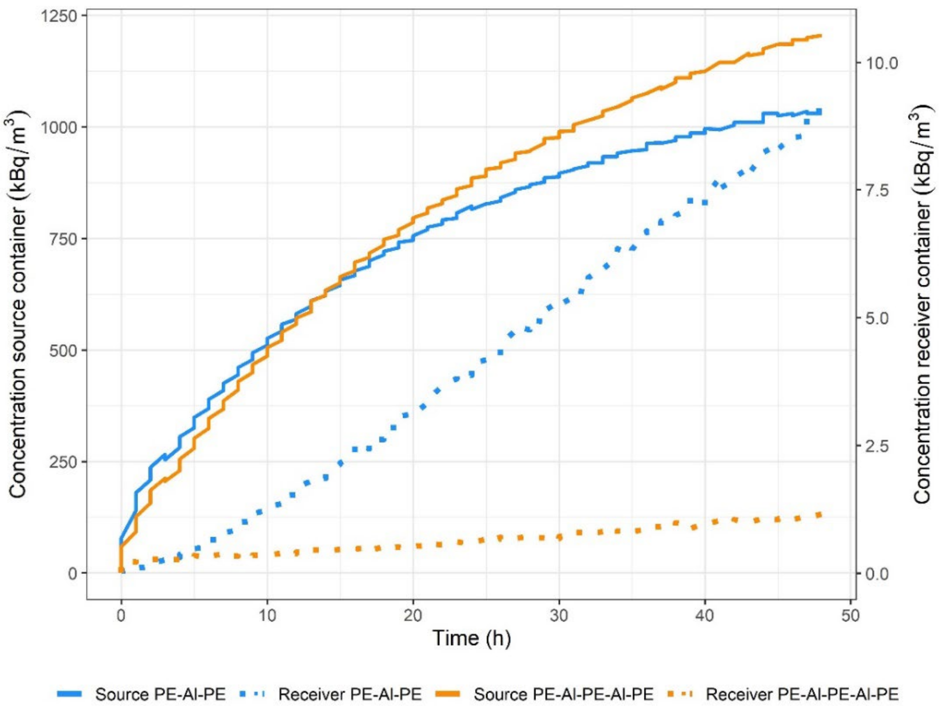

Figure 3. Radon concentration in the source chamber (solid line) and the receiver chamber (dashed line) for the combination of PE and Al foils

Figure 3 shows the evolution of the radon concentration during the experiment when various combinations of PE and Al foils are studied. In this case, the radon concentration in the source chamber surpasses 1000 kBq/m^3 at the end of the experiment. The concentration in the receiver chamber has values around 8 kBq/m^3 and 1 kBq/m^3, respectively. The lowest radon concentration in the receiver chamber among all the tests corresponds to the PE-Al-PE-Al-PE experiment.

Table 4. Values of the parameters calculated to obtain the radon diffusion coefficient of the combination of PE and Al foils

Layers (μm)	C_{SC} (kBq/m^3)	C_{RC} (kBq/m^3)	Reduction (%)	l (μm)	D (m^2/s)
PE (10) + Al (15) + PE (10)	946.13 ± 66.44	6.50 ± 1.42	99.31	316	$(2.10 \pm 0.25)\cdot10^{-13}$
PE (10) + Al (15) + PE (10) + Al (15) + PE (10)	1064.28 ± 97.19	0.87 ± 0.16	99.92	131	$(3.60 \pm 0.44)\cdot10^{-14}$

Table 4 shows the mean radon concentration for the last 24 h of the test for both chambers. With those values, the radon reduction achieved is 99.31% for PE-Al-PE and 99.92% for PE-Al-PE-Al-PE, which is higher than for monomaterial PE barrier (single or multilayer) and around the same values for monomaterial Al barrier (single or multilayer). The diffusion coefficient for PE-Al-PE is around 10^{-13} m^2/s, which is similar to the monomaterial Al barrier with one layer; however, for PE-Al-PE-Al-PE, the result of the diffusion coefficient ($3.6\cdot10^{-14}$ m^2/s) improves compared to the monomaterial Al barrier with two layers. There is a synergistic effect of the combination of both materials when the multilayer material reaches a certain thickness. In fact, the value of the radon diffusion length obtained for the latter combination is the lowest of those obtained in all the tests carried out. Therefore, it seems that the lower the diffusion coefficient, the less radon can pass through the material, and the smaller the radon diffusion length, the less barrier thickness is needed for much of the radon to disintegrate before passing through.

Therefore, the best result of all tested materials is PE-Al-PE-Al-PE because it has the highest reduction percentage, the lowest radon diffusion length and the lowest diffusion coefficient.

3.5 Radon adsorption

After the tests, the radon adsorbed on the barrier materials has also been measured; the net counts (cps) of gamma peaks of Pb-214 and Bi-214 obtained with the gamma spectrometer are shown in Table 5.

Table 5. Counts of the samples studied

Material	Thickness (μm)	Counts (cps)
PE (after diffusion coefficient test)	10 ± 4.96	4.86 ± 0.24
	20 ± 5.18	9.01 ± 0.30
	30 ± 5.18	9.56 ± 0.29
PE (before diffusion coefficient test)	10 ± 4.96	0.043 ± 0.125
Al (after diffusion coefficient test)	15 ± 7.56	7.62 ± 0.27
	30 ± 5.18	8.23 ± 0.28
	45 ± 5.35	8.22 ± 0.29
Al (before diffusion coefficient test)	15 ± 7.56	0.268 ± 0.111
PE + Al + PE (after diffusion coefficient test)	35 ± 7.29	9.77 ± 0.31
PE + Al + PE + Al + PE (after diffusion coefficient test)	60 ± 6.23	9.87 ± 0.31

Table 5 shows the radon adsorbed by the materials during exposure by the counts per second. All materials adsorb radon, and as the thickness increases, more counts are registered. However, PE adsorbs slightly more radon than Al, given that the counts are between 4 and 9 cps and around 8 cps, respectively.

Figure 4 shows the represented values of the counts. The graph shows that there seems to be a relationship between the adsorbed radon and material thickness, being higher for PE than for Al. For the combined materials, both combinations have lower cps than what would correspond to the sum of the counts of the individual materials, even though the resulting material is thicker. For example, for the PE-Al-PE-Al-PE combination, the counts are 9.87 cps, while for two layers of Al, they are 8.23 cps and for three layers of PE, 9.56 cps. Therefore, it is again observed that the combination of polymeric materials with Al in multilayer materials also has a positive effect on radon adsorption on the exposed material, being lower than that of the individual materials.

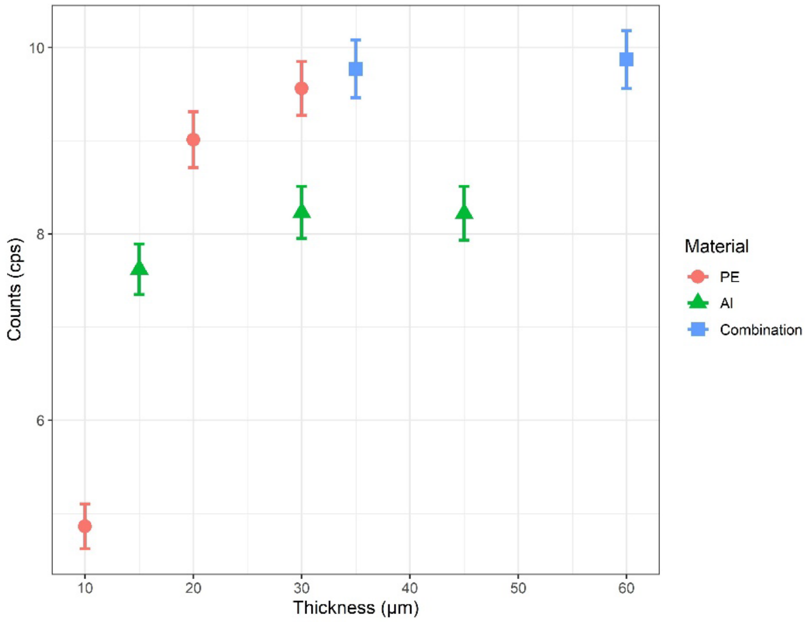

Figure 4. Radon adsorbed by each material compared to its thickness

4. Conclusions

The use of multilayered materials of different nature is very effective in reducing the radon concentration. The use of aluminium in some of the layers is essential to achieve a more significant shielding effect.

It is observed that the best material tested as radon barrier is the PE-Al-PE-Al-PE combination, with a reduction of radon concentration of 99.92%, a radon diffusion length of 131 µm and a radon diffusion coefficient of $3.60 \pm 0.44 \cdot 10^{-14}$ m^2/s, meeting the Spanish Technical Building Code conditions for this parameter.

There is a positive relationship between the thickness of the material and the radon adsorbed; however, PE adsorbs some more radon than Al. In the case of combined multilayer materials, the radon adsorption is lower than the value that would result from the sum of the individual materials.

These preliminary results are positive and show the possibility that multilayer materials resulting from the combination of polymeric materials and aluminium, similar to those laminated materials wastes, can be used as a radon barrier. This would reduce the environmental impact of these residues and contribute to sustainable development and the circular economy.

5. Acknowledgements

This work is financed by the Spanish Nuclear Safety Council under the project “Caracterización, exhalación y remediación de Radón en materiales de construcción (EXRADON)”.

The authors would like to thank GDES for their collaboration during the research.

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