Laboratorio de Radiactividad Ambiental

MEDIDAS DE RADIACION

REPORT

Radon Diffusion coefficient in FOAMGLAS® cellular glass thermal insulation

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1. Introduction

This report aims to give an overview of the project reference "Radon (diffusion resistance) & FOAMGLAS® slabs/blocks/boards – pan European test program (Esp-Sw-Cz)". On 19th November 2012, FOAMGLAS® confirmed the acceptance to start the intercomparison diffusion test on radon gas on a number of FOAMGLAS® slabs/blocks/boards of cellular glass, and the laboratory LaRUC (University of Cantabria, Spain) as scientific coordinator of this project. The test involves three laboratories, from different countries in Europe:

- LaRUC-Radon Group (University of Cantabria, Spain)
- SP Technical Research Institute of Sweden
- Czech Technical University (Faculty of Civil Engineering, Czech Republic)

The samples arrived to the offices of LaRUC on the beginning of 2013. After that, the participants received the testing material and the laboratory from Czech Republic informed about a delay in the analysis. The project involves a multidisciplinary approach based on both, theoretical and practical methods in order to analyse the behaviour of FOAMGLAS® slabs/blocks/boards suitable to be used as radon gas barriers in buildings.

2. State of the art

It is quite difficult to choose the proper material to build the radon barrier due to a lack of data on the radon diffusion coefficient in the available materials. Moreover, there is no homogeneity on the reference value for this coefficient in the European Union. Three countries have quite advance legislation on radon membranes: Republic of Ireland, Czech Republic and Germany. In the case of Ireland, they apply an upper limit for the radon diffusion coefficient. This value corresponds to 12 x 10^{-12} m^2 s^{-1} according to the Irish building regulations [1]. The case of the Czech Republic is different since they do not use the value of radon diffusion coefficient. Instead, Czech Republic uses this value for the evaluation of the minimum thickness of the membrane [2]. In Germany, in order to consider a membrane as radon proof, the thickness of the membrane must be three times the radon diffusion length calculated for the material of the membrane. If the thickness is less than this reference value, the analysis reports that the material is radon permeable [3].

There is clear evidence that most of the effective dose received by the public is due to natural sources of radiation. Within this component of the dose, more than a half is due to radon (^{222}Rn) and its daughters [4]. Radon is a noble radioactive gas, presented in the Earth crust because of radioactive decay from radium (^{226}Ra). Inhalation of radon gas is the second cause of lung cancer after cigarette smoking, being the estimation of lung cancer risk increased by 16% per 100 Bq m^{-3} [5]. Thus, there is a great concern to assess the radon gas exposures in living and working environments and keep them to the lowest possible levels. [6]. In order to establish a level of radon concentration indoors, EU has considered the value of 200 Bq m^{-3} as a reference level for new buildings and 400 Bq m^{-3} for existing buildings [7].

An effective method to prevent soil gas containing radon from entering building is the addition of a barrier between the occupied spaces and the ground. Apart from the problem of the absence of the same reference value in all European countries, the difficulty to measure the radon diffusion coefficient is quite high. To overtake this difficulty, there is an on-going ISO project to standardise the method for the determination of this parameter. There are five laboratories in Europe with the equipment for measuring the radon diffusion coefficient but applying different methodologies. Nevertheless, the application of different methodologies to determine the radon diffusion coefficient has shown a big inconsistency in the results. In 2009–2010, the Czech Technical University of Prague organized the first international intercomparison to measure the radon diffusion coefficient [8]. The test consisted in three different materials corresponding to polyethylene membranes of high and low density. These types of membranes represent the materials normally applied in the buildings as protection against water, radon and damps. The results indicated differences up to two orders of magnitude and this was one of the evidences used for the beginning of the ISO project. However, not only we can use polyethylene as protection against radon. The variety of the materials is quite high and thus the range of values of the radon diffusion coefficients goes from 10^{-15} to 10^{-8} m^2 s^{-1} as we can see in Figure 1 corresponding to one study of the parameter in 360 different materials [9].

Figure 1 show that the best values correspond to bitumen membranes with an Al carrier film, irrespective of modifications in the bitumen, and for EVA membranes. On the other hand, the highest values of the radon diffusion coefficient were for sodium bentonite membranes, rubber membranes made of EPDM, and cement coatings. The radon diffusion coefficients for waterproofing widely used to protect houses,

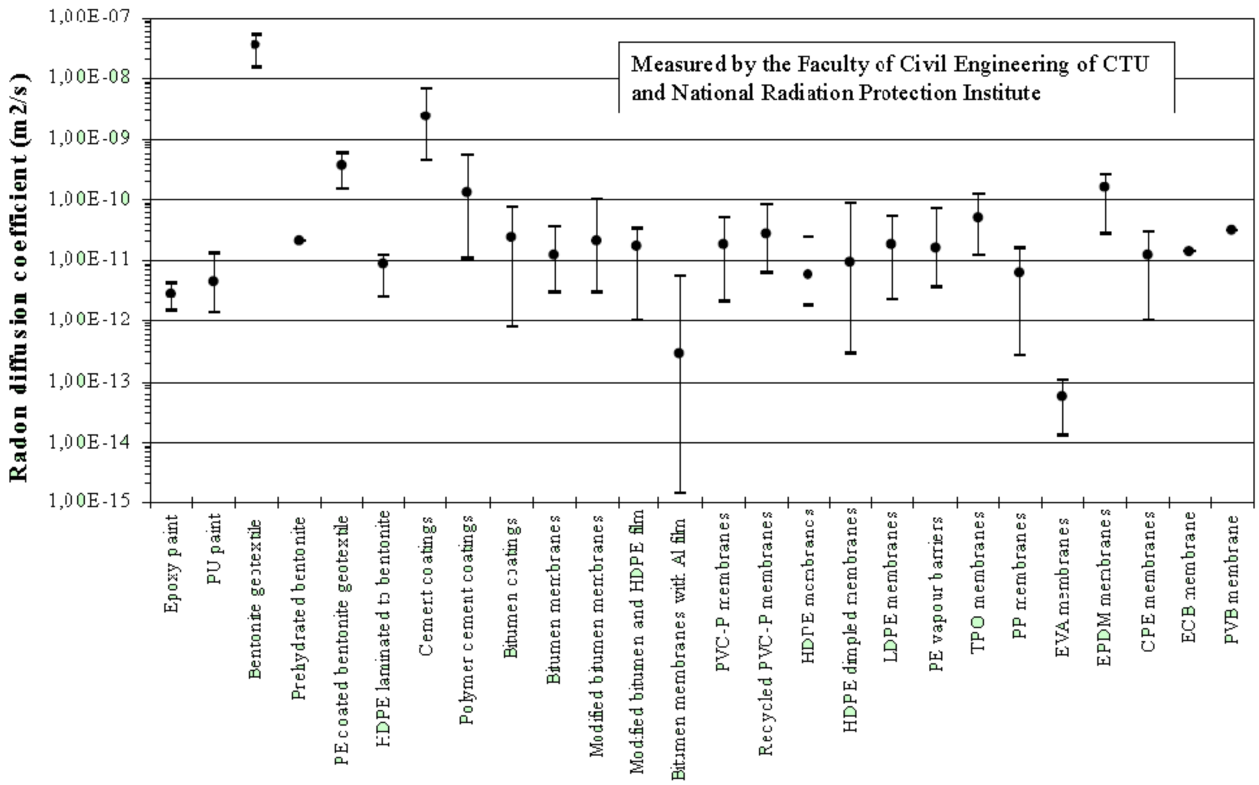

Figure 1. Results of radon diffusion coefficient measured in 360 waterproof materials of different chemical composition.

i.e. PVC, HDPE, LDPE, polypropylene and bitumen membranes vary in the range from 3.10^{-12} to 3.10^{-11} $m^2\,s^{-1}$. Relatively large differences between the minimum and maximum values (long error bars) are caused by the fact that widely-used membranes produced for the building industry are not composed of pure polymers. They contain various fillers, additives, softening agents, UV stabilisers, colours, reinforcing fabrics, etc. Even materials of the same chemical origin contain different types and different amounts of these admixtures. We do not usually know the detailed composition of the membranes, because producers consider this data as restricted. Relatively large differences between the minimum and maximum values (long error bars) are caused by the fact that widely-used membranes produced for the building industry are not composed of pure polymers. They contain various fillers, additives, softening agents, UV stabilisers, colours, reinforcing fabrics, etc. Even materials of the same chemical origin contain different types and different amounts of these admixtures.

We can describe the transport of radon trough the membrane by the radon transport equation (1),

$$\frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right)-\lambda C=\frac{\partial C}{\partial t}, \tag{1}$$

where C is the radon concentration within the membrane (Bq m^{-3}), D is the radon diffusion coefficient (m^2 s^{-1}), λ (2.1 10^{-6} s^{-1}) is the radon decay constant, x the membrane's thickness (m) and t the time (s). One attempt for the solution to this equation is by means of numerical methods such as Finite Element Methods (FEM) [10, 11]. With this methodology, it is possible to get the value of the radon diffusion coefficient assuming that the transport of radon gas through the membrane is one-dimensional. Nonetheless, to apply equation (1) it is necessary to have enough experimental data in order to operate the iterative process involved in FEM technique.

A recent study has carried out a review of the measurement techniques available in the literature for the measurement of the radon diffusion coefficient [12]. There are two basic experimental set-up's: first, we seal the sample in one container with a known radon concentration [13, 14]. Then, we measure the radon diffusion coefficient by studying the radon radioactive decay inside the container. The second method consists in two containers: source container (containing the radon source) and receiver container [10, 15-20], and the membrane is installed between both containers. The growth of radon concentration in the receiver container is measured and then we can calculate the radon diffusion coefficient. The ranges of radon concentrations can be from kilobecquerels to megabecquerels. Concerning the techniques for measuring radon gas in the receiver container, we can underlined three: Scintillation [13, 17 and 18], semiconductor detectors [21 – 23] and ionization chambers [11, 16]. In some cases, steady state conditions in the receiver container are required for the calculations [23, 24]. In most of the cases, the ambient parameters are not considered and therefore we can find some deficiencies: the use of stationary equations to analyse data from non-steady state conditions [16, 20].

The current state of the research on radon proof materials presents some inconsistencies. First, the use of the FEM method considers one-dimensional transport of radon gas in soil. The real situation includes transport in two dimensions and it is necessary to include this parameter when solving the radon transport equation in soil. Moreover, the laboratories perform most of the analyses under laboratory conditions where ambient parameters are always under control. The real situation is quite different and the changes of these parameters in a real house can be high during all seasons and affect the characteristics of the membrane.

3. Material and methods

We can see the description of the samples for the analysis in Table 1.

Table 1. Summary of the testing samples of the project organized by densities and participants.

	Density ca 115 kg/m³ FOAMGLAS® T4+	**Density ca 120 kg/m³ FOAMGLAS® HLB800**	**Density ca 130 kg/m³ FOAMGLAS® S3**	**Density ca 165 kg/m³ FOAMGLAS® F**
LABO nr 1 - ES 3 sets of 30x30x3: Nr. 1 Nr. 2 Nr. 3	A.1 A.2 A.3	B.1 B.2 B.3	C.1 C.2 C.3	D.1 D.2 D.3
LABO nr 2 - CZ 3 sets of 30x30x3: Nr. 4 Nr. 5 Nr. 6	A.4 A.5 A.6	B.4 B.5 B.6	C.4 C.5 C.6	D.4 D.5 D.6
LABO nr 3 - SE 3 sets of 45x 60x3: Nr. 7 Nr. 8 Nr. 9	A.7 A.8 A.9	B.7 B.8 B.9	C.7 C.8 C.9	D.7 D.8 D.9

Below, we provide a short description of the methods applied by participants.

- **LABO nr 1 – ES**

The method consists in inserting the simple between two containers, radon tight sealed. The volume of each container is $2.5 \cdot 10^{-2}$ m^3. The testing area is 0.08 m^2, which corresponds with the surface of the membranes. The source container comprises the radon source with a high radon production rate. Radon diffuses through the sample to the receiver container. We measure the radon concentrations by installing a radon monitor inside each container. This prevents radon leakages in the cable connections in case of measurements performed outside containers. Figure 2 offers a view of the experimental set-up in the Spanish laboratory.

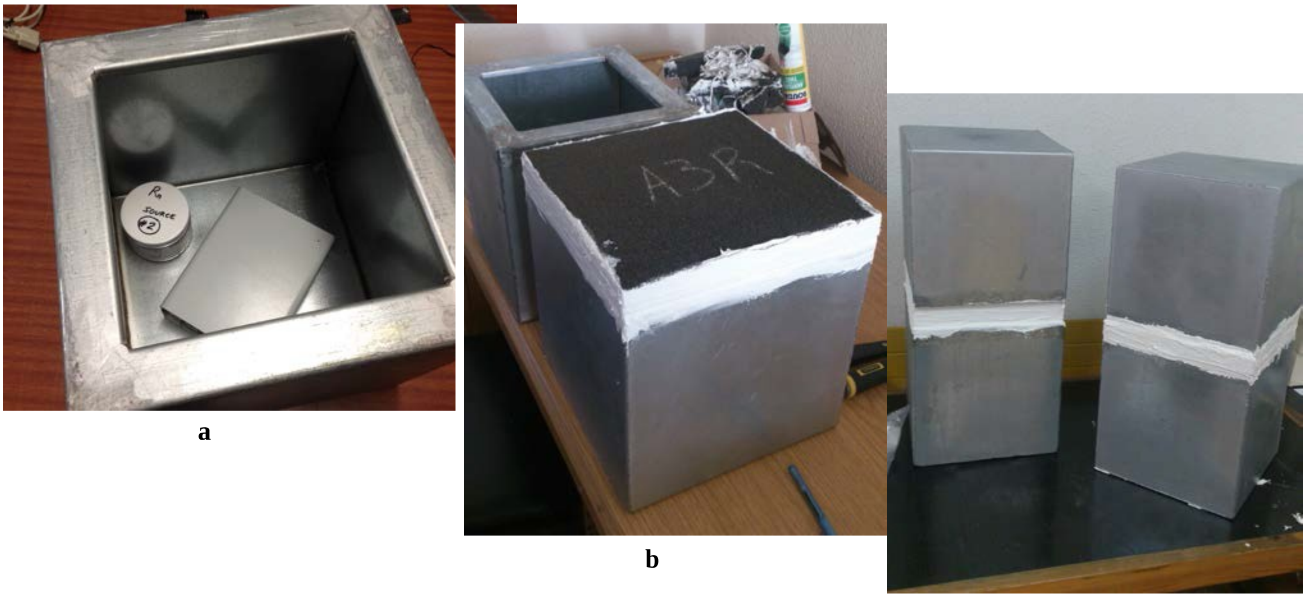

Figure 2. Experimental set-up in Spanish laboratory LaRUC, *a)* with the source container open, *b)* with the FOAMGLAS® sample covering and *c)* with the upper container (final setup).

We carry out a non-linear fit of equilibrium concentrations by using Origin software. This type of fitting procedure allows knowing the most important characteristics on this measurement, such as equilibrium values in both containers, tightness and radon production rate in the source container [24].

LaRUC has different radon sources. Firstly, there are radon sources composed by soil with a high content of ^{238}U and ^{226}Ra spread out at the bottom of the source container (see Figure 2). The radon production rate of this soil depends on the total amount of soil and its thickness. Typical values are in the range 10 ± 2 kBq m^{-3} h^{-1}. On the second hand, a radon source model PYLON RN-1025 is also available with an activity of 100 kBq made of dry ^{226}Ra. This device produces calibrated radon activities.

The equation (1) describes the transport of radon trough soil considering one dimensional transport mechanism. The available models for the solution of this equation [10, 11] consist to divide the membrane in finite elements with two nodes each. Considering that radon concentration inside each element is constant, the model evaluates the changes of radon concentration between each node and thus through the membrane. By Garlekin approach, we assume that the radon concentration between nodes changes linearly. Within the membrane, there is only transport of radon, not transmission. The transmission of radon is only in the layer of the membrane in contact with air, described by means of a coefficient used as constant in all cases. Applying a process called localization it is possible to determine the radon concentration inside the membrane. Then, the model uses an iterative process using some values of radon diffusion coefficient previously defined and fits the theoretical results to experimental data.

- **LABO nr 2 – CZ**

The laboratory in the Czech Republic follows the procedure describe in the project ISO DIS 11665-10 “Measurement of radioactivity in the environment – Air: radon 222 – Part 10: Determination of the diffusion coefficient in waterproof materials using activity concentration measurement” [27]. By the moment of the production of this interim report, the Czech laboratory is performing the tests and they did not send their interim report yet.

Radon diffusion coefficient was measured according to the accredited method in the Czech Republic (K124/01/09). Two samples of the tested material were placed between the lower container, which is connected to the radon source, and two upper containers. Schematic drawing of the measuring device is presented in Figure 3. After radon is admitted into the lower container, radon diffuses through the samples to the upper containers, in which the radon concentration starts to increase. When the steady-state radon concentration profile is set up within the tested samples, the upper containers are flushed with radon-poor ambient air and afterwards they are closed again. Since then, the increase in the radon concentration in the upper container is measured for the second time.

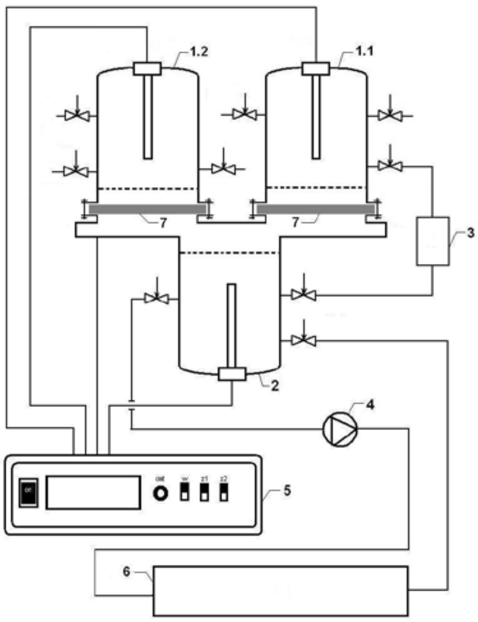

Figure 3. Schematic drawing of the measuring device (1.1, 1.2 – upper containers A21 and A22, 2 – lower container A1, 3 - pressure difference sensor, 4 – pump (during this measurement the pump was not used), 5 – control and operation unit ERM4, 6 – radon source, 7 – tested sample).

Concentrations on both sides of the tested samples (Figure 4) are measured continuously by ionisation chambers, which function at the same time as the containers.

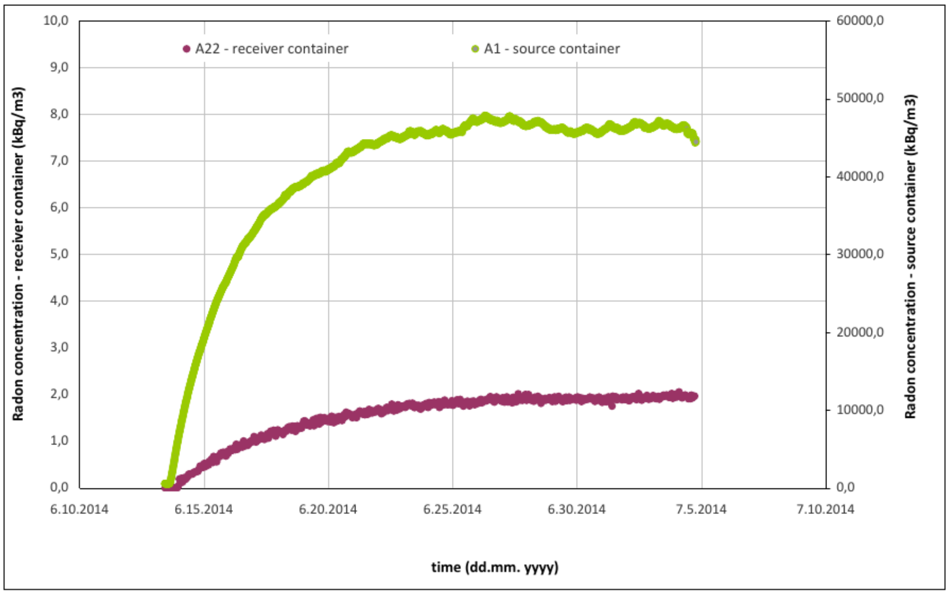

Figure 4. Radon concentrations measured in the source container A1 and in the right receiver container A22.

Diffusion coefficient was derived from the process of fitting the numerical solution to the curves of radon concentration measured in the upper containers. Numerical solution is based on the one-dimensional time-dependent diffusion equation describing radon transport through the tested material. Calculation of the diffusion coefficient was performed for non-stationary conditions using software IterRn, which description is available in [11].

- **LABO nr 3 – SE**

Testing was carried out in a test chamber comprising of two stainless steel boxes. Each box measured 351 x 500 mm. The depth of the receiver box was 154 mm and the depth of the source box was 251 mm. The test sample was placed between the boxes. The sides were then carefully tightened, to ensure an airtight connection between the boxes. The radon source was a block of aerated concrete, which contains a small amount of radium. The radioactive decay of radium will produce radon gas (^{222}Rn) which is emitted to the atmosphere in the source box. ^{222}Rn is also radioactive and its first decay product (RnD) is Polonium-218. Radon decay products (RnD) are not gases but particles, and cannot pass through the test specimen by diffusion.

The radon concentration on each side of the test specimen was determined by an instrument of type Atmos 33, SP No. 202266, produced by Gammadata in Sweden. The measuring principle used in these instruments is to determine the concentration of Polonium-218 and convert it into radon concentration assuming an invariable relationship between the Rn and Po concentrations. The instrument was calibrated at the Swedish Radiation Protection Institute on 17 th February 2014. Testing was carried out in a room with the following conditions: a relative humidity of 0 ± 5 %, and a temperature of 23 ± 2 °C. These conditions were monitored throughout the full duration of the test (7 days).

The emission of radon from the radon source will lead to an increase of the radon activity concentration in the source box and a difference in radon activity concentration between the source and receiver box. This difference will cause a flow of radon by diffusion through the test specimen. Only the radon gas (Rn) and not the radon decay products (RnD) will pass through the test specimen.

The start activity concentration (C_0) in both source and receiver boxes equals the background level at the start of the test. The regression curves for $C_1(t)$ and $C_2(t)$ are computed according to the following equations:

$$\frac{dC_1(t)}{dt} = \frac{\varnothing}{V_1} - \frac{PA}{V_1} \cdot \left(C_1(t) - C_2(t)\right) - C_1(t) \cdot \lambda_1 \qquad (2)$$

$$\frac{dC_2(t)}{dt} = \frac{PA}{V_2} \cdot \left(C_1(t) - C_2(t)\right) - C_2(t) \cdot \lambda_{Rn^{222}} \qquad (3)$$

$$\lambda_1 = \lambda_{Rn^{222}} + \lambda_B \qquad (4)$$

Ø (Bq s^{-1}) is the radon exhalation rate of the source material. The decay constants for the source box is denoted λ_1. The decay constant is composed by the radioactive decay of radon gas ($\lambda_{Rn222} = 2.1 \cdot 10^{-6}$ s^{-1}) and back diffusion (λ_B) in the source block. The radon exhalation and the total decay constant are determined to yield the best fit of $C_1(t)$ according to the measured source box concentration.

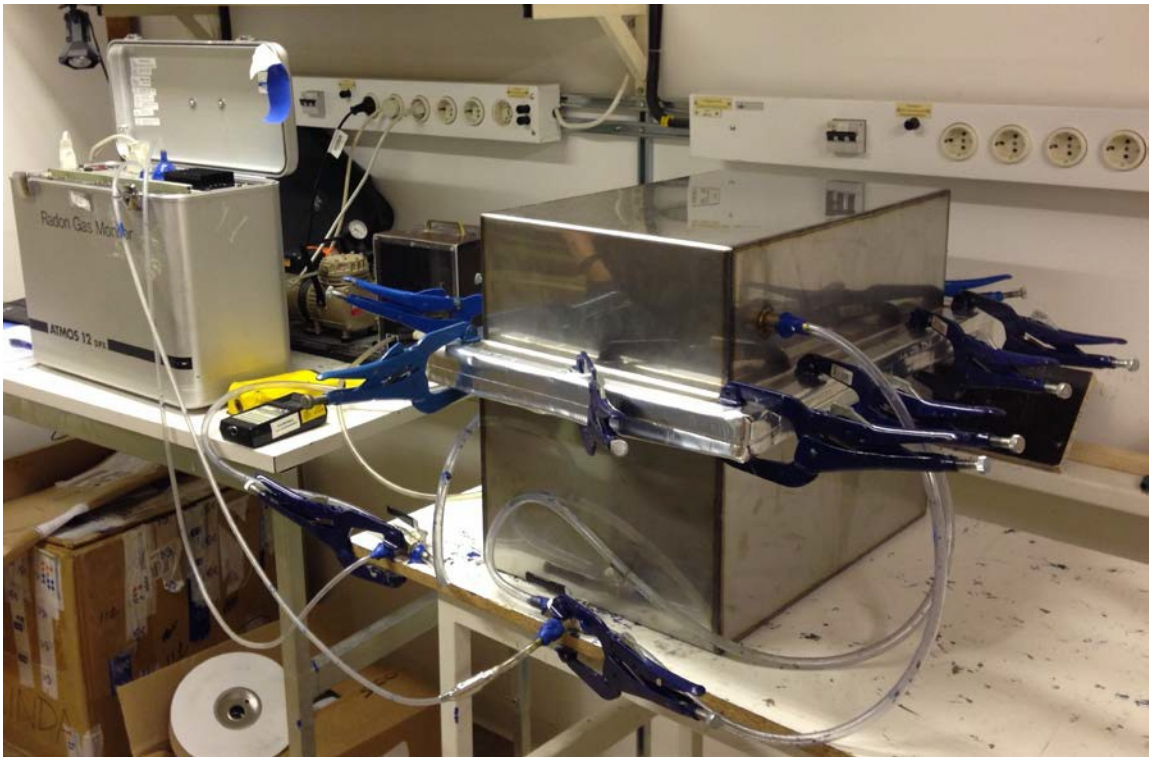

Figure 5. Experimental set-up in the Swedish laboratory.

4. Results

All participants received 4 types of samples identified as “FOAMGLAS® T4+”, “FOAMGLAS® HLB 800”, “FOAMGLAS® S3” and “FOAMGLAS® F” which they will be referred in this report as “A”, “B”, “C” and “D” respectively. Each category contained three sub-samples and the laboratories measured all in order to increase the statistics on the determination of the radon diffusion coefficient. Table 2 shows the results provided by labs and we can observe that all results are on the order of 10^{-11} m^2 s^{-1} which represents a very low value for the radon diffusion coefficient.

Table 2. Summary of the results obtained in the pan European test program. The table shows the results in terms of radon diffusion coefficient *D* (m^2 s^{-1}).

Product	Lab. 1 (Spain)	Lab. 2 (Czech Republic)	Lab. 3 (Sweden)
FOAMGLAS® “A” **T4+**	$4.8 \cdot 10^{-11}$	$2.0 \cdot 10^{-11}$	$6.3 \cdot 10^{-11}$
FOAMGLAS® “B” **HLB 800**	$2.9 \cdot 10^{-11}$	$1.3 \cdot 10^{-11}$	$2.2 \cdot 10^{-11}$
FOAMGLAS® “C” **S3**	$4.5 \cdot 10^{-11}$	$1.8 \cdot 10^{-11}$	$2.8 \cdot 10^{-11}$
FOAMGLAS® “D” **F**	$2.0 \cdot 10^{-11}$	$1.6 \cdot 10^{-11}$	$4.7 \cdot 10^{-11}$

The table above shows that the highest value for *D* corresponds to FOAMGLAS® sample type T4+ in the three participants, whereas there is not a good agreement on the lowest value of this parameter. Nevertheless, the differences in the results are negligible if we take into account the uncertainties. The determination of radon diffusion coefficient is affected by large uncertainties which are in the range of 20 – 50 % of the result.

Table 3. Results of Radon diffusion coefficient *D* (m^2 s^{-1}) for each type of sample.

Material	# tested samples	Min	Max	Mean	SD
A	9	$2.0 \cdot 10^{-11}$	$6.3 \cdot 10^{-11}$	$4.4 \cdot 10^{-11}$	$2.2 \cdot 10^{-11}$
B	9	$1.3 \cdot 10^{-11}$	$2.9 \cdot 10^{-11}$	$2.1 \cdot 10^{-11}$	$8.0 \cdot 10^{-11}$
C	9	$1.8 \cdot 10^{-11}$	$4.5 \cdot 10^{-11}$	$3.0 \cdot 10^{-11}$	$1.4 \cdot 10^{-11}$
D	9	$1.6 \cdot 10^{-11}$	$4.7 \cdot 10^{-11}$	$2.8 \cdot 10^{-11}$	$1.7 \cdot 10^{-11}$

If we look into the individual sample types more in detail, we observe the results indicated on Table 3.

First, the mean values of *D* in each FOAMGLAS® material, expressed as arithmetic mean of the results in the particular sample from the three participants, are quite similar. It would be possible to assess that there is no difference on *D*. However, to confirm this result, we would need to analyse a larger amount of samples. Figure 6 shows in terms of histogram chart the frequency of the data. It appears that the distribution of the results is far from normality. Therefore we could neglect the possibility that all samples present the same value for *D*. Anyhow, the sample size is too short to conclude such statement with statistical significance.

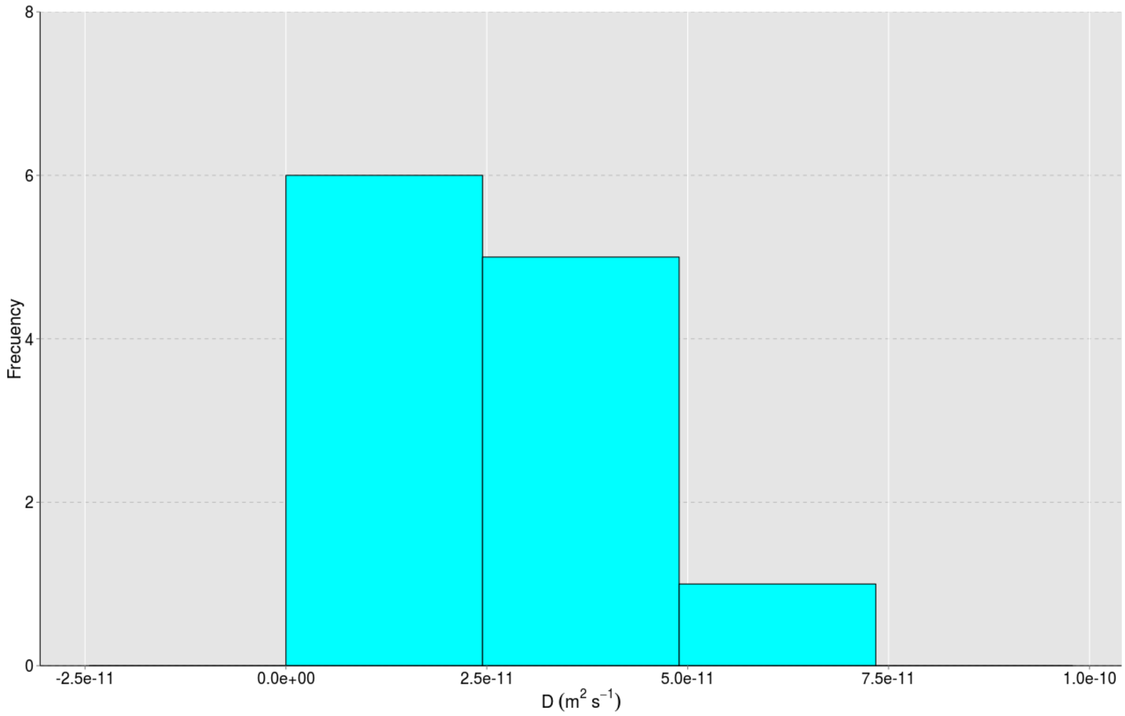

Figure 6. Histogram of the results from the pan European test on radon diffusion coefficient of FOAMGLAS® slabs/blocks/boards.

It is interesting to pay attention to the individual results in each laboratory as we can see in Figure 7. We observe that the lowest values are those from the Czech participant while Spanish and Swedish parties obtained larger results for the parameter.

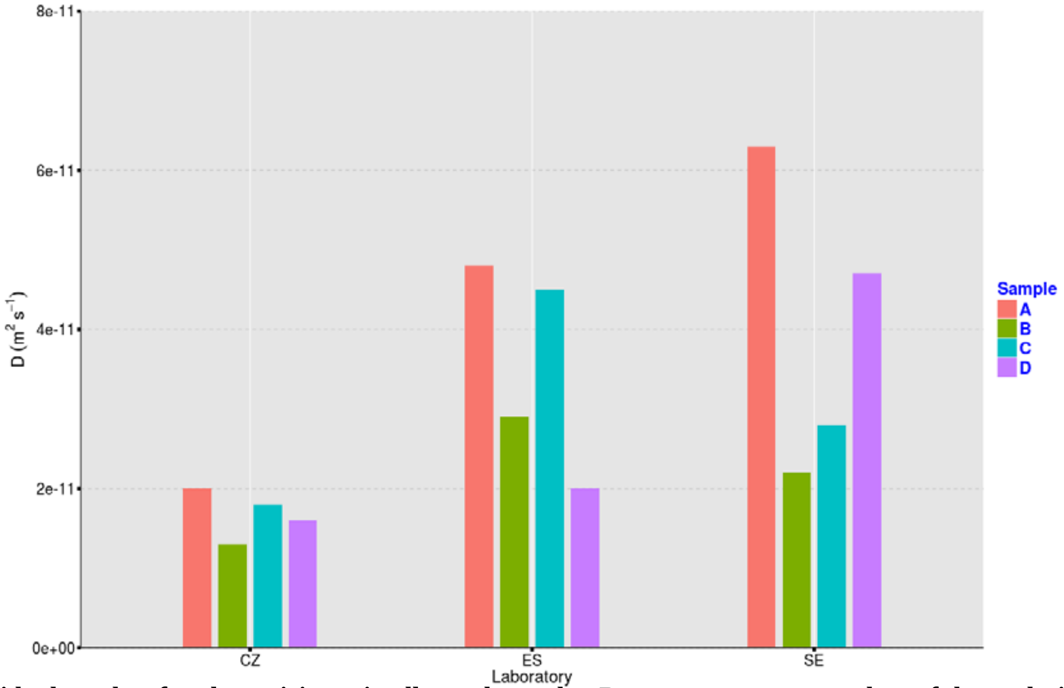

Figure 7. Individual results of each participant in all tested samples. Bars represent mean values of the analysis.

In all cases, the biggest value of *D* belongs to sample "A" or T4+, but there is not agreement on which material gives the lowest result of *D*.

Finally, we show in Figure 8 the mean values for each type of material. It is clear the difference from material T4+ and the rest. Yet we insist that the number of tested samples is very small to draw whether the different types of FOAMGLAS® materials have significantly different radon diffusion coefficients

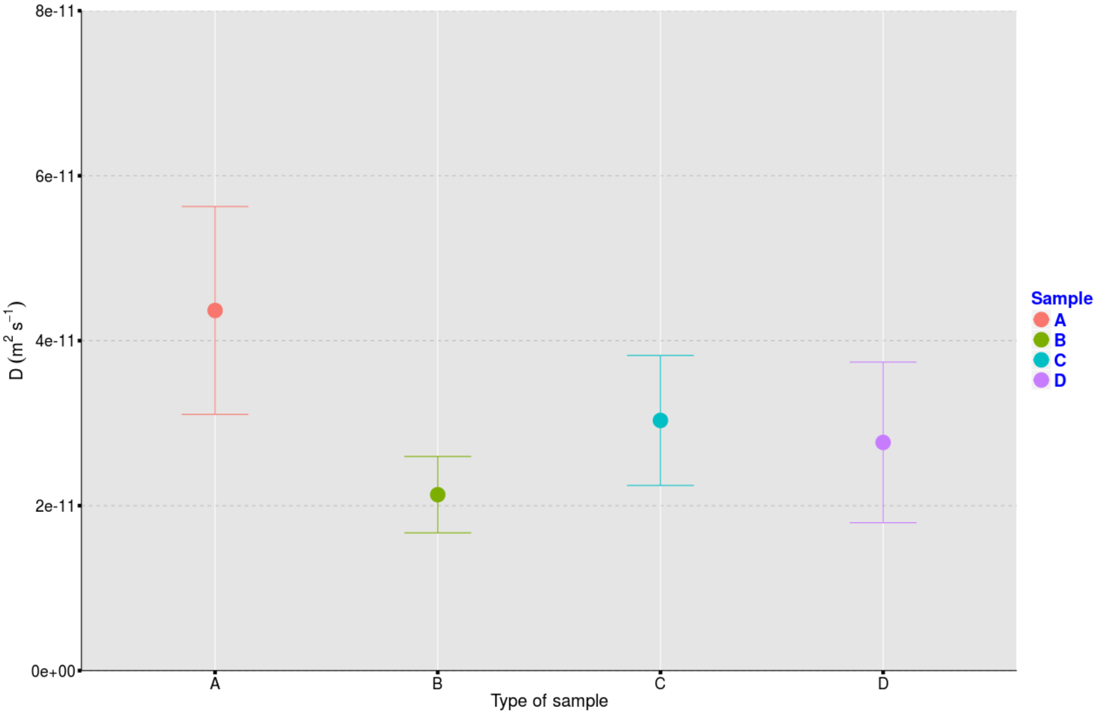

Figure 8. Mean values of each type of analysed material. Error bars represent standard error of the mean.

5. Conclusions

The results included in this report are representative of the pan European test of radon diffusion coefficient in FOAMGLAS® materials. We remark that so far there is no international standard for this type of analysis. Hence it is not possible to test the accuracy of the laboratories in terms of primary standards. Thus we have achieved the next outcomes:

1. The four materials offer a good protection against radon gas (^{222}Rn) and they can be used as radon proof barriers. The values of radon diffusion coefficient are very low (order 10^{-11}).

2. Due to the special behaviour of radon gas, it is necessary to guarantee that there are no cracks in the installation of the FOAMGLAS® barrier. So we come up with including in the installation a sealant in the joints of the barriers.

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