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Radon permeability of insulating building materials

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Abstract. The aim of the study was to determine the radon permeability coefficient of insulating building materials. Eleven insulating materials were tested. A research setup was developed and it was as follows: a tested material was tightly set on the receiver box's hole and placed into the radon chamber. The measurements showed that for various insulating materials, radon permeability coefficient varies from 1.26×10^{-10} m^2/s for film-like materials to 9.95×10^{-8} m^2/s for roofing papers. According to our calculations of all insulating materials, the foil-type insulating materials to ensure the best protection against radon flow from the ground. Comparison of different types of building materials shows that the insulating building materials ensure better radiological protection than regular building constructions materials.

Key words: insulating building materials • permeability • radon • transmittance

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Introduction

The common presence of the radioactive nuclides in the environment cause that the whole population are exposed to radiation. The dose that is annually received by Polish population from natural sources of radiation is about 2.4 mSv per year [1]. According to the UNSCEAR 2000 Report [2], radon is the greatest contributor of exposure to natural radiation for humans. Soil and the rocks on which the buildings are embedded constitute the main source of radon in the closed spaces, such as buildings. The building materials used to construct and finish the houses are the second major source. Radon penetrates from the soil into the homes through all of kinds of leaks in the foundations, primarily because of the difference in air pressure between the building and the ground. About 80% of radon in the buildings comes from the soil. About 12% of radon in building materials comes from radium contained therein [3]. The remaining portion of radon found in the homes comes from the outside ambient air, water and fuel gas. ^{222}Rn concentration in the home atmospheres is affected by various factors, including the ^{226}Ra contained in the ground, the tectonic setting and features of the subsurface layers of soil that determine the intensity of radon exhalation (porosity, moisture, permeability, temperature gradient). Also the meteorological conditions affect the concentration of radon in the lower troposphere in a specified location and have an influence on the transport of this radionuclide from remote areas.

The studies on human exposure to radon have confirmed that this gas, both in the residential and occupational setting, constitutes a real threat to human health by triggering the pathological changes in the respiratory tract that may result in the development of lung cancer [4, 5].

Radon is also included amongst the factors responsible for the development of sick building syndrome (SBS), known also as environmental illness or multiple chemical sensitivity (MCS). It is believed that SBS can have direct and indirect impact on health, workplace comfort and productivity of the employees [6].

Building-related illnesses (BRI) is defined as the illness(es) caused directly as the result of being in and around the building environment that is suspect to having SBS. BRI can be caused by a number of factors individually (biological factors, physical factors, chemical factors, organizational and management factors, psychological and psychosomatic factors) or a combination of their synergetic effects [7].

The indoor and outdoor environment plays a significant role in determining the quality of air we breathe at our workplaces or in our homes and thereby can be the cause of SBS.

Thus, the external environment is the main source of pollutants, for example, traffic pollution (CO, CO_2, NO_x, SO_x, etc.), radiation (ultraviolet exposure) and land-derived contamination (methane, radon). The indoor environment is a cocktail of diverse factors, for example, pollutants coming, amongst others, from the building materials (containing, e.g. formaldehyde, solvents, mineral fibres, radon gas, pesticides, interior furnishings and volatile organic compounds), infestation by insect, pests and other forms of biological organisms. Well-being in closed spaces also depends on humidity and mold growth, noise, odor and irritation, emission of gases and outdoor pollution, air conditioning, and control of indoor microclimate and other factors (thermal comfort, lighting, space per occupant, occupant activities, moisture and introduction of pollutants, tobacco smoking) [7].

As it can be seen, radon is mentioned amongst different kind of factors that may have impact on the occupant's health or the synergetic effect of these factors may be the cause of health-related problems in the buildings.

SBS is controversial. Although many people and some clinicians believe that there is a medical syndrome related to buildings and their internal environment (the so-called building-related illness), many other clinicians and medical organizations say there is no convincing clinical evidence that such a medical syndrome exists [8]. The controversy exists because a number of people have a mix of non-specific symptoms that have no documented medical cause, yet believe that they occur from sources inside building(s). Medical organizations such as the American Medical Association (AMA) and many experts say that without any defined symptoms and no convincing evidence of a given source or cause, there is no test to diagnose the syndrome and no treatment for the syndrome. They do not believe that such medical syndrome exist [8].

Regardless of whether the building-related illness exists or not, the harmful effect of radon (causing the lung cancer) is undoubtedly proven [9].

Unlike its mother nuclide, radon daughters are not gaseous. They are solid species that can merge with the dust particles in the air to form radioactive aerosol. Breathing the air contaminated with radon and radioactive aerosols leads to an increased incidence of lung and larynx cancer. According to EPA (United States Environmental Protection Agency), radon is a serious risk to population health [10], and so

a) If 1000 smoking people are exposed for their whole lifetime to radon concentration of 150 Bq/m^3, 62 of them may develop cancer. In this case, the risk of developing cancer is five times higher than the risk of dying in a traffic accident.
b) If 1000 non-smoking people are exposed for their whole lifetime to radon concentration of 150 Bq/m^3, seven of them may develop cancer. In this case, the risk of developing cancer is equal to the risk of dying in a traffic accident [10].

The example quoted above has not been given by chance. Recent measurements of radon concentration in homes show that the average annual radon concentrations in Polish dwellings are 170 Bq/m^3 [11]. There are regions in Poland where the average annual radon concentrations in residential houses may exceed 300 Bq/m^3. Council Directive 2013/59/EURATOM of 5 December 2013 [12] includes a statement that recent epidemiological findings from residential studies demonstrate a statistically significant increase in the risk of lung cancer from prolonged exposure to indoor radon at levels of the order of 100 Bq/m^3. The Directive recommends that "Member States shall establish national reference levels for indoor radon concentrations in workplaces. The reference level for the annual average activity concentration in air shall not be higher than 300 Bq/m^3, unless it is warranted by national prevailing circumstances" [12]. In Poland, these regulations must be made obligatory until February 2018. Implementation of the Directive will cause in many cases the need of reducing the radon diffusion from the ground into the buildings.

It is expected that in some areas of Poland, it will be necessary to reduce the radon concentration in enclosed areas by all possible means. One of the possible ways to reduce radon concentration in buildings is to use the appropriate barrier materials with low radon permeability and adequate sealing vertical surfaces, for example, painting or wallpapering rooms. Other methods, such as creating an overpressure in the building, increasing basements ventilation (if any) or reducing the radon concentration in the soil through its suction around/under the building, are technically complicated and encumbered with high costs of installation and operation. It is expected that in some areas of Poland, it will be necessary to use appropriate insulating materials in order to reduce radon concentrations. For the

Table 1. Summarized values of permeability and transmittance coefficients

Type of insulating material	Thickness d [mm]	Permeability k [m^2/s]	Transmittance P [m/s]
An ordinary thick protective film	0.1	1.26×10^{-10}	1.26×10^{-6}
Thermo-vapour barrier reinforced Al foil	0.1	2.68×10^{-10}	2.68×10^{-6}
Steam insulation film	0.1	6.36×10^{-10}	6.36×10^{-6}
Steam thermal insulation foil	0.1	6.73×10^{-10}	6.73×10^{-6}
Building waterproofing PVC film, 1 kg/m^2	0.6	3.99×10^{-9}	6.65×10^{-6}
Building waterproofing PVC film	1.0	5.24×10^{-9}	3.80×10^{-6}
	1.5	6.11×10^{-9}	4.08×10^{-6}
Insulation film under the foundations (for horizontal insulation of the foundations)	0.8	7.36×10^{-9}	7.93×10^{-7}
Roofing paper	3.1	8.29×10^{-9}	2.68×10^{-6}
Tar paper	2.7	2.75×10^{-8}	3.73×10^{-6}
Roofing paper Base	4.9	9.95×10^{-8}	1.99×10^{-6}
Absence of insulating material	–	–	4.10×10^{-4}

insulation materials, the permeability testing would be interesting for parties producing and using these materials.

The aim of the study was to determine the radon permeability coefficient of insulating building materials. Knowledge about radon permeability coefficient of insulating building materials may be very useful in the design of buildings (safe from the point of view of radiological protection) in areas with high radon concentrations in the soil. It can also useful in remediation action in cases of existing buildings with high radon concentration inside.

In the recent years, the increase in public awareness of radon and its harmful impact on human health can be observed. Single-family housing development in particular can be the reason of intensified interests in building insulation materials that reduce radon diffusion into the building.

Materials and methods

We have examined 11 pieces of materials used in architecture (summarized in Table 1), mainly roofing papers, building waterproofing polyvinyl chloride (PVC) films, steam insulation films and thermo-insulating foils for radon permeability. The size of each sample was 0.02 m^2.

The testing was carried out in the radon chamber. A constant generator of radon (Pylon radon generator) was used as the radon source. An average radon concentration used during the test and generated by the Pylon source was 270 kBq/m^3. The radioactive decay of radium-produced radon gas ^{222}Rn was emitted to the atmosphere in the radon chamber. A receiver box made of Plexiglas with a thickness of 1 cm was inserted into the radon chamber. The box measured 40 × 60 × 30 cm and was provided with a 16-cm diameter hole. The tubes were brought to the receiving box to enable radon blow out every time the new specimen were inserted. In order to avoid the underestimation of the actual radon concentration inside receiving box, radon permeability of Plexiglas receiver box was measured (with all valves and holes closed). It was found that radon permeability of Plexiglas (for entire surface of the receiver box) was equal to 1.14×10^{-10}. This is two orders of magnitude less than the values of radon permeability for all samples collected by us, so the error from the radon permeability of Plexiglas may be ignored. This was an evidence for the tightness of the receiver box, and that potential leakages inside the Plexiglas receiver box were minimum and they did not affect the results of the test.

The test sample was placed on the hole and pressed by a silk and iron rings, so the connection between the box and the testing material was airtight. The radon from the radon chamber was transmit through the testing material into the box. The measure of the radon concentration inside the box and inside the radon chamber were carried out by scintillation method using Lucas scintillation chambers.

The emission of radon from the radium source leads to a build-up of the radon concentration in the radon chamber. The difference in radon concentration between the radon chamber and the box result in radon transmission into the box through the test material. Radon transmission through the specimen can be described by two coefficients: the transmittance coefficient and the permeability coefficient. Radon transmittance P is a property of the gas and the type of material, and it represents the speed of radon flow through the specimen, expressed in [m/s]. The radon transmittance can be assessed by measuring the radon concentrations on both sides of the test specimen, as the radon is flowing through the test material.

Permeability k is the second coefficient that describes the radon transmission through the test material. The permeability k is a function of specimen thickness and the diffusing substance characteristic. It describes the rate of the radon diffusion through a specific thickness of the material (expressed in metres) and is expressed in [m^2/s].

In evaluating the radon transmission, it is assumed that the radon concentration in both radon chamber and the box is increasing linearly with time during a time interval (t_1 to t_2).

Dependence of radon permeability k on radon transmittance P is as follows:

$$k = P \cdot d \tag{1}$$

where d is the specimen thickness [m] and the unit of k is [m^2/s].

We used the equations given in the report [13] to calculate the transmittance of radon for chosen specimen material.

The density of radon flow through the specimen is as follow:

$$q = P \cdot (C_1 - C_2) \tag{2}$$

where q is the density of radon flow [Bq/m^2·s]; P is the radon transmittance [m/s]; C_1, C_2 is the radon concentration on both sides of the test specimen – C_1 in the radon chamber and C_2 in the receiver box [Bq/m^3].

Diffusion laws are responsible for radon ingrowth in the receiver container. The differential equation for the radon concentration build-up in the receiver box (C_2) is

$$\frac{dC_2}{dt} = P \cdot (C_1 - C_2) \cdot \frac{A}{V} - \lambda \cdot C_2 \tag{3}$$

where, t is the time [s]; $A = 0.02$ test specimen area [m^2]; $V = 0.072$ box volume [m^3]; $\lambda = 2.1 \times 10^{-6}$ decay constants [s^{-1}].

With $C_1 = a + b{\cdot}C_2$ (for $y = a + bx$.), Eq. (3) becomes

$$\frac{dC_2}{(a + b \cdot C_2 - C_2) \cdot \frac{P \cdot A}{V} - \lambda \cdot C_2} = dt \tag{4}$$

or

$$\frac{dC_2}{a + C_2 \cdot \left(b - 1 - \frac{\lambda \cdot V}{P \cdot A}\right)} = \frac{P \cdot A}{V} \cdot dt \tag{5}$$

Integration between t_1 and t_2 and C_2^1 and C_2^2 gives

$$\frac{1}{b - 1 - \frac{\lambda \cdot V}{P \cdot A}} \cdot \ln\left[\frac{a + \left(b - 1 - \frac{\lambda \cdot V}{P \cdot A}\right) \cdot C_2^1}{a + \left(b - 1 - \frac{\lambda \cdot V}{P \cdot A}\right) \cdot C_2^1}\right] = \frac{P \cdot A}{V} \cdot (t_1 - t_2) \tag{6}$$

where C_2^1 and C_2^2 is the radon concentration in the receiver box, respectively, in time t_1 and t_2.

The numerical methods were used to calculate P (transmittance) coefficients from Eq. (6). Afterwards, the permeability coefficient k was calculated from Eq. (1).

The uncertainty of radon concentration measurement is estimated to be ±8% (uncertainty of generator, scintillation chambers, calibration curve, statistics and the repeatability). The influence of evaluation of the box's volume, specimen surface and its thickness and uncertainty of the counts (assumed to be 1.5% for high radon concentrations) on total uncertainty of the measurements is about ±4.5%, resulting the total uncertainty of permeability coefficient to be ±12.5%.

First reading of C_1 and C_2 was taken about 4 h after the sample was installed, and further reading were taken once or twice every day.

For comparison, we measured radon concentration in the receiver box in case, when there was no material on the box's hole. We left the receiver box open for 15 min and after this time, we measured the radon concentration in it. During taking of the samples, we covered the gap with the lid made of the same material that receiver box is made (to avoid a radon suction from the radon chamber).

Results and discussion

Eleven pieces of insulating materials were examined. We can classify them into three types according to their application: roofing papers, film-like insulating material and building waterproofing PVC films (this is an insulating material with characteristics resembling those of the roofing paper).

The calculated values of the radon permeability and radon transmittance of the insulating building material listed in the ascending order of k values are presented in Table 1.

As it can be observed from Table 1, the protection from radon transmission depends primarily on the type of the specimen. The film-like insulating materials such as steam insulation films, foil thermo-vapour barrier, the insulation film under the foundations and ordinary protective film are examples of best radon-protective materials amongst all insulating building materials. We found that the permeability coefficient of different types of films to vary from $k = 1.26 \times 10^{-10}$ for ordinary protective film to 7.36×10^{-9} for the insulation material under the foundations (for horizontal insulation of the foundations). In turn, the building waterproofing PVC films have the permeability coefficient ranging from $k = 3.99 \times 10^{-9}$ to 6.11×10^{-9} m^2/s. The permeability coefficient of the roofing papers ranges from $k = 8.29 \times 10^{-9}$ to 9.95×10^{-8} m^2/s.

Obviously, for the specific material, a great role in radon permeability plays its thickness, for example, as we can observe from Table 1, the film is at least 30 times thinner than the roofing paper, and the permeability coefficient of the film is one to two orders of magnitude less than the permeability coefficient of roofing paper. It means that the radon permeability coefficient depends on the structure of the specimen material, that is, if the specimen is homogeneous, it may be thin and the diffusion coefficient will be low and the radon risk will be low, but if the specimen is heterogeneous, even thick insulating specimen poses a higher radon risk in closed area.

Similar investigations to ours were performed by Narula *et al.* [14]. He investigated the radon permeability coefficient k of construction building materials (limestone powder, sand stone, granite, soil, sand, cement, fly ash, gypsum, wall putty). According to him, for various construction building materials, the coefficients of radon permeability are in a range (0.06–6.44) × 10^{-6} m^2/s. The comparison of his results with ours shows that the insulating building materials ensure better protection against the radon transmission from the ground than regular building constructions materials. People using insulating building materials unconsciously ensure themselves better protection against the negative influence of radon.

Of course none of the insulating materials can stop radon flow from the ground, if they are not properly (i.e. in a gas-tight way) installed. For comparison of gas-tight, and non-tight installation, a case when the box was covered with no insulation material is shown under item 12 (last line) in Table 1. The calculated radon transmittance coefficient *P* for this case is two to three orders of magnitude greater than that for insulating materials. It means that when any of the insulating materials is used, the radiological protection is better than when none is applied. Of course the speed of radon flowing into the room (the radon transmittance) depends on the size of the gap. During installation, it is necessary to pay particular attention to proper insulation of all media supply systems (e.g. sewer pipes, water pipes, gas lines, power cables, telephone lines) in the erected building. Culverts must be designed and built with special care to ensure that any leaks of connections do not create a radon entry routes into the interior. Methods to eliminate pathways of incoming radon include repairing leaks in foundations, floors and walls (mainly in basements or rooms having direct contact with the ground), sealing installation of utility infrastructure, use of appropriate insulation coatings and pressure equalization inside and outside.

Conclusions

It is expected that in some areas of Poland, the use of appropriate insulating materials will be necessary in order to reduce radon concentrations. According to our calculations, of all insulating materials, the film-type insulating materials ensure the best protection against radon flow from the ground. From the comparison of various types of building materials, it seems reasonable to conclude that the insulating building materials ensure better radiological protection than the regular building construction materials. But from a radiological protection point of view, any of the insulating materials can be used; it is better than when none is applied.

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