RADON DIFFUSION COEFFICIENT IN RADON-PROOF MEMBRANES — DETERMINATION AND APPLICABILITY FOR THE DESIGN OF RADON BARRIERS

Martin Jiránek
Czech Technical University in Prague, Faculty of Civil Engineering, Thákurova 7, 166 29 Praha 6
Czech Republic

Jiří Hůlka
National Radiation Protection Institute, Šrobárova 48, 100 00 Praha 10, Czech Republic

ABSTRACT

The selection of effective radon-proof insulation from the total amount of tanking materials is very difficult due to the lack of information about radon diffusion through these materials and the absence of general guidelines for the design of radon barriers. This report describes a method for radon diffusion coefficients determination used in the Czech Republic. Results of radon diffusion coefficients measurements in more than 80 insulating materials are summarized. We have found that great differences exist in diffusion properties. The diffusion coefficients vary within four orders from 10^{-13} m^2s^{-1} to 10^{-10} m^2s^{-1}. A methodological approach is proposed in order to identify the minimal thickness of radon-proof membranes in dependence on the radon diffusion coefficient in the insulation, radon concentration in the soil and building characteristics. General guidelines for selection of radon-proof insulation and the principles of application are presented.

1. DETERMINATION OF RADON DIFFUSION COEFFICIENT

1.1 Description of the Measuring Method

Determination of radon diffusion coefficients is based on the measurement of the radon flux through the tested material placed between two cylindrical containers (volume 1 – 2 litres). The lower container is connected to the radon source commercially made in the Czech Republic (type RF-2000 containing a special highly emanating Ra-226 compound, activity of 2 MBq) that creates the radon concentration of approximately 100 $MBqm^{-3}$ within this container. Radon may diffuse through the tested material to the upper container that is permanently ventilated. The system is assumed to be in the steady state after reaching a time much longer then the "relaxation time". In this time the upper container is closed and the growth of radon concentration in it is measured by means of extraction of air samples and introducing them into the Lucas cells. Extraction is provided in the interval of 10 - 15 minutes. Lucas cells are used also for the measurement of radon concentration in the lower container. While in the lower container the radon concentration is typically 100 $MBqm^{-3}$, in the upper one the typical concentrations within the first hour are in the range 1 - 100 $kBqm^{-3}$. The whole measurement is made under room temperature. Measuring appliance is shown in Fig. 1.
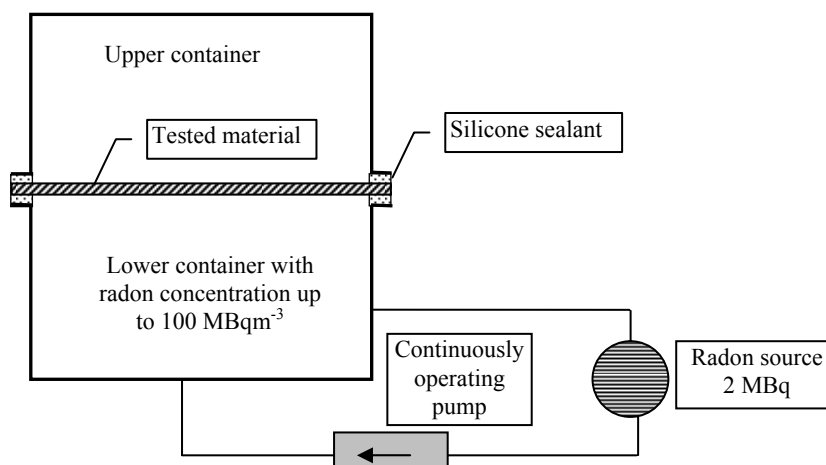

Fig. 1: Arrangement of the measuring appliance

The space-time distribution of radon concentration in the tested material can be evaluated from the partial differential equation [1], which has, in one-dimensional case, the form:

$$\frac{\partial C_{(x,t)}}{\partial t} = D \cdot \frac{\partial^2 C_{(x,t)}}{\partial x^2} - \lambda \cdot C_{(x,t)} \qquad (1)$$

where D is the radon diffusion coefficient [m^{2}s^{-1}], λ is the radon decay constant (2.1 x 10^{-6} s^{-1}) and $C_{(x,t)}$ is the radon concentration [Bqm^{-3}].

We assume simple boundary conditions: constant radon concentration in the lower container of the system $C_{(0,t)} = C_0$ and zero concentration $C_{(d,t)} = 0$ in the upper container (continually ventilated). The time development of the radon concentration profile in the tested material can be characterised by the "relaxation" time [2] evaluated as:

$$t_{rel} = \frac{1}{D.(l^{-2} + \pi^2 . d^{-2})} \qquad [s] \qquad (2)$$

where d is the thickness of the tested material [m], l is the diffusion length $l = (D/\lambda)^{1/2}$ [m] and D is the radon diffusion coefficient [m^{2}s^{-1}]. In certain case, where d is much greater than l, the relaxation time is approximately equal to the decay time (Fig. 2). After reaching a time much longer than the relaxation time, the system is assumed to be in steady state. The relaxation time is therefore important entry information for setting the time schedule of the measurement – underestimation of the relaxation time can lead to false evaluation of the diffusion coefficient.
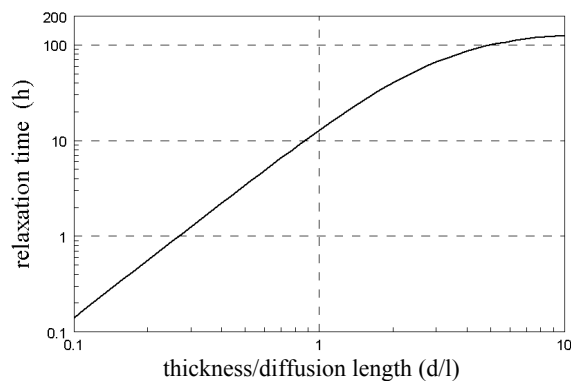

Fig. 2: Dependence of the relaxation time on the *d/l* ratio

At time of steady state the concentration profile $C_{(x)}$ in the tested material and radon flux density $J_{(d)}$ from the tested material to the upper ventilated container can be described by equations (3) and (4), which can be simply derived from equation (1) solved for one-dimensional case and boundary conditions mentioned above:

$$C_{(x)} = C_0 \cdot \frac{e^{-(x-d)/l} - e^{(x-d)/l}}{e^{d/l} - e^{-d/l}} \qquad [Bqm^{-3}] \qquad (3)$$

$$J_{(d)} = -D \cdot \frac{dC_x}{dx} = \frac{2.C_0.l.\lambda}{e^{d/l} - e^{-d/l}} \qquad [Bqm^{-2}s^{-1}] \qquad (4)$$

where C_0 is the radon concentration in the lower container [Bqm^{-3}], $C_{(x)}$ is the radon concentration in the depth x in the tested material [Bqm^{-3}] and $J_{(d)}$ is the radon flux density from the tested material to the upper container.

Immediately after closing the upper container (after reaching the relaxation time), the growth of radon concentration is measured. Because radon concentrations in the upper container during the first hour after enclosure are still low (compared with concentration when new steady state will be reached), radon flux density (emanation rate) is not significantly affected by back diffusion [3]. Therefore the growth of radon concentration in the upper container within the first hour can be described by equation (5) and in approximation by equation (6):

$$C_{(t)} = \frac{J.S}{V.k} \cdot (1 - e^{-k.t}) \qquad [Bqm^{-3}] \qquad (5)$$

$$C_{(t)} = \frac{J.S}{V} \cdot t \qquad [Bqm^{-3}] \qquad (6)$$

where J is the radon flux density [Bqm^{-2}s^{-1}], S is the area of the tested sample [m^2], V is the volume of the upper container [m^3], k is the coefficient of radon leakage from the upper container [s^{-1}] and t is the time [s]. The approximation (6) is valid if *(k.t)* << 1. During experiments we have measured k = 0.1 h^{-1} (see Fig. 3) and therefore approximation (6) can be well used within the first hour. The coefficient of radon leakage k however can differ according to the size of the container and the experiment arrangement.
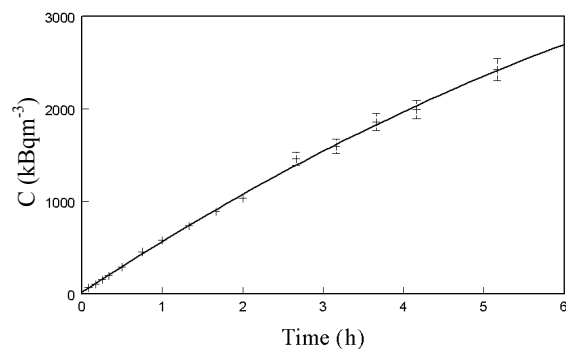

Fig. 3: The growth of radon concentration in the upper container (measured for radon concentration in the lower container 100 MBqm^{-3})

The main advantage of this method is usage of approximation (6) where radon leakage coefficient *k* does not appear. The knowledge of leakage coefficient for radon flux evaluation is hence not necessary and uncertainties in the sealing of the containers cannot play a role in the evaluation.

1.2 Evaluation of the Diffusion Coefficient

From the known time dependent curve of the radon concentration increase in the upper container (Fig. 3) and known volume *V* and area *S,* radon flux $J_{(d)}$ from the measured material can be calculated. The calculation is based on statistical fitting of measured data to the theoretical functions (5) or (6). From the evaluated radon flux, known radon concentration in the lower container and known thickness of the measured sample, the radon diffusion length *l* (equation 4) and finally the radon diffusion coefficient $D = l^2.\lambda$ can be numerically calculated.

1.3 Uncertainties of the Measurement

The total uncertainty of radon diffusion coefficient measurement depends on the following partial uncertainties:

- uncertainty in evaluation of the insulation thickness (mainly in the case of mineral or sand dressed bitumen membranes) - (10 - 30 %),

- uncertainty in evaluation of the container volume *V* and the area of the sample *S* (< 5 %),

- statistic uncertainty of radon concentration measurement in the lower container (< 10 %),

- uncertainty in evaluation of the radon flux density into the upper container, caused by the uncertainty in evaluation of the volume of air samples which are introduced into the Lucas cells (< 10 %) and by the statistic uncertainty in assessment of radon concentration in the upper container (< 10 %).

The total uncertainty of radon diffusion coefficient measurement is predicted to be 20 - 40 %. This value is mostly influenced by the uncertainty in evaluation of the insulation thickness. For that reason three different samples of insulation are used for the measurement. If the standard deviation calculated from three samples cannot be accepted, another samples must be measured.

1.4 Method Applicability

Described method of the radon diffusion coefficient measurement is convenient mainly for materials in which the radon diffusion is not dependent on material porosity and moisture content, i.e. for various types of bitumen membranes and plastic foils of different chemical composition. If porous materials are measured by this method, the obtained value of *D* is dependent on porosity and moisture content.

The minimal values of *D*, which can be measured by this method for different thickness of tested materials, are shown in Fig. 4. The curve was calculated from equations (4) and (5) for radon concentration in the lower container of 100 MBqm^{-3} and minimal measurable (by Lucas cell) radon concentration in the upper container of 50 Bqm^{-3}. It can be seen that for 1 mm thick insulation, the method can be used for measurement of diffusion coefficients up to 10^{-13} m^2.s^{-1}. Since *D* in the most frequently used insulating materials is higher than 10^{-13} m^2 .s^{-1} and the thickness of produced plastic foils and bitumen membranes varies between 1 - 4 mm, the method is applicable for all common insulating materials used as waterproof or radon-proof barriers.
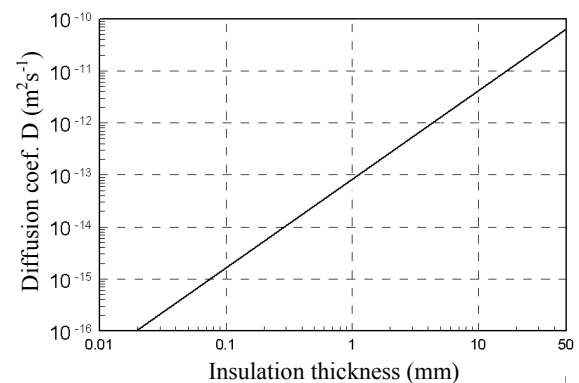

Fig. 4: Evaluated minimal measurable radon diffusion coefficient D (m^{2}s^{-1}) as a function of the thickness of the tested sample (valid for radon concentration in the lower container of 100 MBqm^{-3})

1.5 Measured Values

The results of the radon diffusion coefficient D measurement realized by the Faculty of Civil Engineering of the Czech Technical University in Prague and by the National Radiation Protection Institute in more than 80 insulating materials available throughout Europe are summarized in Fig. 5. Generalizing results obtained up to now, we have found out that for the most often used insulation, the diffusion coefficients vary between 10^{-13} m^{2}s^{-1} and 10^{-10} m^{2}s^{-1}.

The lowest values of the radon diffusion coefficient D were obtained for polypropylene foils. In HDPE foils with dimples, the coefficient varies in the range 1 x 10^{-12} and 5 x 10^{-12} m^{2}s^{-1}. Radon diffusion coefficients for HDPE and PVC

foils and plastomeric or elastomeric bitumen membranes were measured between the orders of 5 x 10^{-12} and 10 x 10^{-12} m^2s^{-1}. In the range 1 x 10^{-11} and 2.5 x 10^{-11} m^2s^{-1}, the coefficients for bitumen membranes made of oxidised bitumen, recycled PVC and LDPE or ECB membranes were found. The highest values of D were discovered for rubber foils made of EPDM, where the coefficient D increases up to the order of 10^{-10} m^2s^{-1}.

2. DIMENSIONING OF RADON-PROOF INSULATION

Under the conditions that the insulation is placed over the entire area of structures in direct contact with the soil, all joints between sheets are airtight and any penetration of utility entries through the insulation is properly sealed, we can consider the convective transport of radon to be negligible. Therefore it is possible to assume that the radon supply rate into the house with continuous tanking is created only by the diffusion through insulation. Based on this simplification, the highest permissible radon exhalation rate into the house, E_{lim}, can be expressed by equation (7):

$$E_{\lim} = \frac{C_{dif}.V.n}{A_f + A_w} \qquad [Bqm^{-2}h^{-1}] \qquad (7)$$

where V is the interior air volume [m^3], n is the air exchange rate [h^{-1}], A_f is the floor area in direct contact with the soil [m^2], A_w is the area of the basement walls in direct contact with the soil [m^2] and C_{dif} is a fraction of the highest permissible radon concentration indoors C_{lim} (in the Czech Republic C_{lim} is 250 Bqm^{-3} for new buildings and 500 Bqm^{-3} for existing buildings) caused by diffusion. The value of C_{dif} is, according to the Czech standard, estimated to be 10 %. This means that the importance of the diffusion is reduced to 10 % of C_{lim} and the remaining 90 % of C_{lim} is reserved for the accidentally occurring convection. Our estimate of C_{dif} is consistent with the range of 4 - 50 % presented by Holub and Killoran [4]. An upper limit of 50 % for the diffusion component has been found for substructures without insulation.

Detailed design of radon-proof insulation (i.e. thickness, number of layers, sort of insulation etc.), in dependence on real geological and building characteristics, is based on the condition that the radon exhalation rate E from the real insulation in a real house calculated according to equation (8) [6], must be less or equal to the highest permissible radon exhalation rate E_{lim} calculated for that house, i.e. $E \le E_{lim}$.
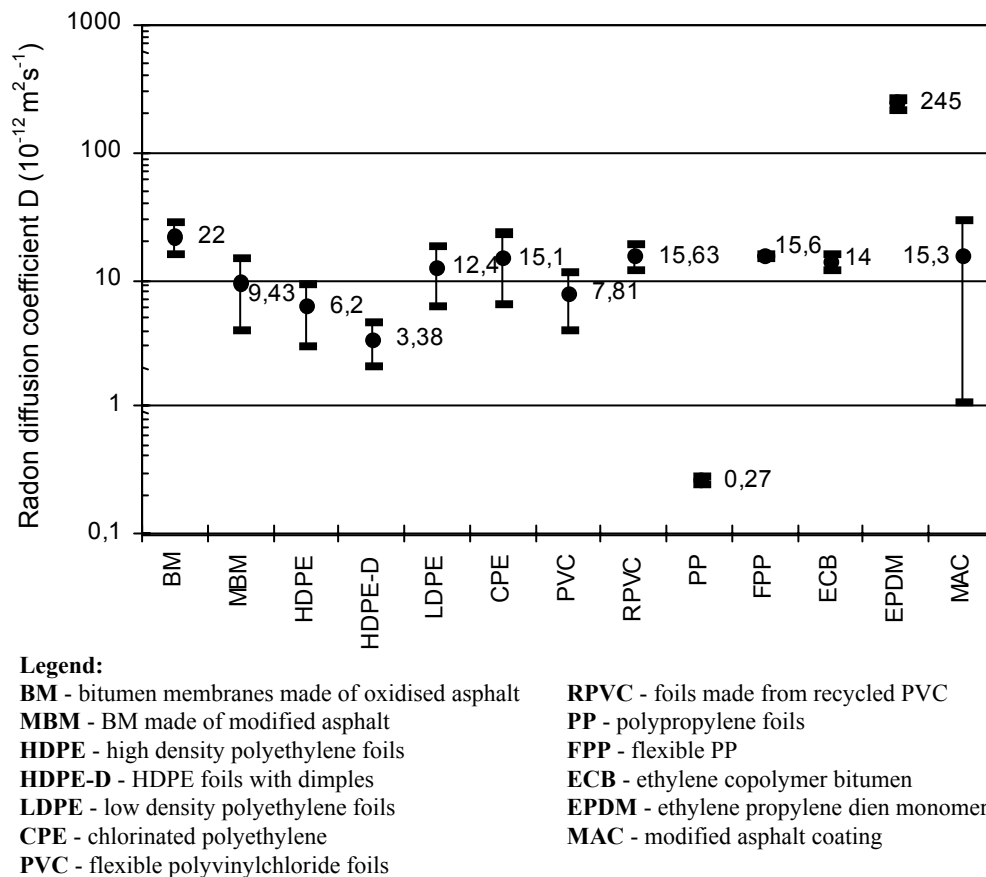

Legend:

BM - bitumen membranes made of oxidised asphalt
MBM - BM made of modified asphalt
HDPE - high density polyethylene foils
HDPE-D - HDPE foils with dimples
LDPE - low density polyethylene foils
CPE - chlorinated polyethylene
PVC - flexible polyvinylchloride foils

RPVC - foils made from recycled PVC
PP - polypropylene foils
FPP - flexible PP
ECB - ethylene copolymer bitumen
EPDM - ethylene propylene dien monomer
MAC - modified asphalt coating

Fig. 5: Radon diffusion coefficients

$$E = \alpha_1 . l . \lambda . C_S \frac{1}{\sinh(d/l)} \qquad [\text{Bqm}^{-2}\text{h}^{-1}] \qquad (8)$$

$$d \geq l . \text{arcsinh} \frac{\alpha_1 . l . \lambda . C_S}{E_{\lim}} \qquad [\text{m}] \qquad (9)$$

where C_S is the radon concentration in the soil gas [Bqm^{-3}] measured on the building site at the depth 0.8 m, λ is the radon decay constant [0.00756 h^{-1}], d is the thickness of the radon-proof insulation [m], l is the radon diffusion length in the insulation $l = (D/\lambda)^{1/2}$ [m], D is the radon diffusion coefficient in the insulation [m^{2}h^{-1}] and α_1 is the safety factor that should eliminate the inaccuracies arising during the soil gas radon concentration measurements and the possible increase of the radon concentration beneath the completed house in comparison with the radon concentration C_S measured on the unbuilt area. Values of α_1 have been estimated according to the soil permeability (for highly permeable soils $\alpha_1 = 7$, for soils with medium permeability $\alpha_1 = 3$ and for low permeable soils $\alpha_1 = 2,1$). Equation (8) can be derived from equation (1) by the same way as equation (4).

On the assumption that the insulation is homogeneous, its minimal thickness can be calculated from equation (9) obtained after the replacement of E in the equation (8) by E_{lim} from equation (7).

3. DISCUSSION

The principle of designing radon barriers according to the above mentioned method, which is also incorporated into the Czech Standard ČSN 730601 "Protection of buildings against radon from the soil" [5,6], can be illustrated in Fig. 6 in which the thickness of the insulation with D = 1 x 10^{-11} m^{2}s^{-1} is plotted against the soil gas radon concentration for different soil permeabilities. It can be seen that the thickness increases proportionally to the radon concentration in the soil and soil permeability.

The thickness of the insulation against the radon diffusion coefficient D for different soil permeabilities and house types is plotted in Fig. 7. From this figure, it is clear that the thickness of the insulation with D of order 10^{-12} m^{2}s^{-1} can be only several tenths of one millimetre, even in the areas with high radon concentration in the soil. Such small thickness is hardly produceable and applicable due to risk of puncturing and thus, in the practical sense, thicker insulation must be used. This may lead to uneconomic and inefficient over-dimensioning.
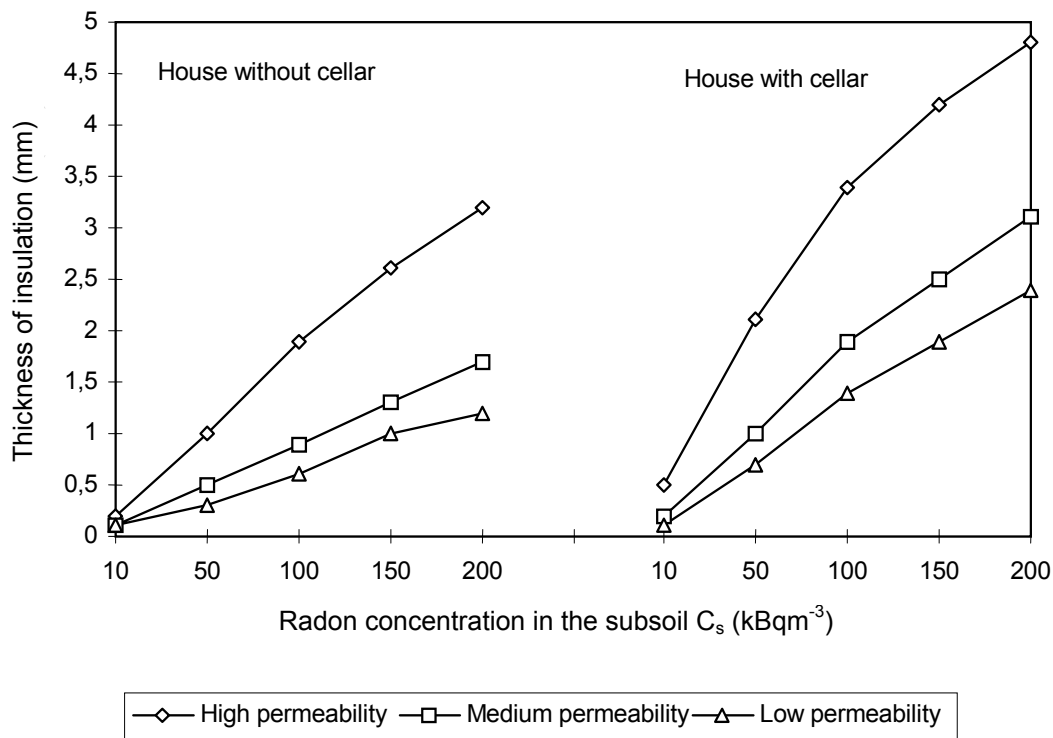

Fig. 6: The thickness of the insulation with D = 1 x 10^{-11} m^{2}s^{-1} plotted against the soil gas radon concentration, soil permeability and the type of a house
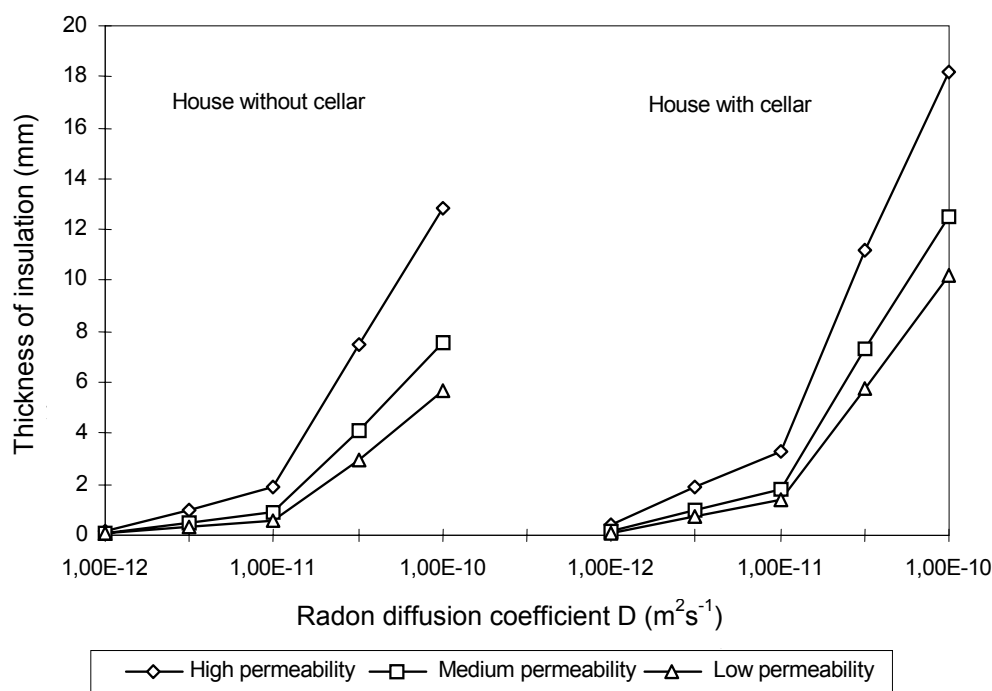
Fig. 7: The thickness of the insulation in dependence on the radon diffusion coefficient D, soil permeability and the house type (for C_s = 100 kBqm^{-3})

On the other hand, the applicability of the insulation with D of order 10^{-10} m^{2}s^{-1} will be very strongly dependent on building characteristics and the radon concentration in the soil. Since radon barriers made of such insulation must be thicker (several millimetres), the insulation must be placed in two or three layers. It can be seen that rubber foils made of EPDM with D around 2.45 x 10^{-10} m^{2}s^{-1} are too permeable to be used for radon-proof insulation.

This clearly leads to the conclusion that the optimal value of D lies in the interval 5 x 10^{-12} to 1 x 10^{-11} m^{2}s^{-1}. This interval corresponds with the production thickness of most frequently used insulating materials, that is 1 or 2 mm for plastic foils and 3 or 4 mm for bitumen membranes (which in addition can be applied in two or three layers).

4. CONCLUSION

Experience from the Czech republic shows that the presented method for the evaluation of the minimal thickness of radon-proof membranes, which depends on the building and soil characteristics and the radon diffusion coefficient in the insulation, is useful in examining the effectiveness of the insulation and in optimising its design. The possibility of over or under-dimensioning is thus very strongly reduced. However it is important to stress that insulating materials must function effectively over their required service time. From this point of view, the preference should be given to insulating materials with longer durability and higher resistance to ageing, i.e. membranes based on oxidised bitumen should be replaced by membranes based on plastomeric (APP modification) or elastomeric (SBS modification) bitumen, as far as the reinforcing fabrics concerns, only membranes with moisture resisting fabrics made of mineral, glass or synthetic fibres should be used. Concerning application aspects, bonded sheets (fully adhered to the substrate) represent higher degree of protection compared to unbonded sheets, which rely on perfect joints to a far greater extent than do bonded membranes, because radon can travel in the air gap beneath the unbonded membranes to the untight places in the insulation.

ACKNOWLEDGEMENT

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