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RADON DIFFUSION COPEFFICIENT – A MATERIAL PROPERTY DETERMINING THE APPLICABILITY OF WATERPROOF MEMBRANES AS RADON BARRIERS

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ABSTRACT

Barrier properties of various waterproofing materials against radon were studied by means of the radon diffusion coefficient. Method for the determination of this material property used in the Czech Republic is presented. Results of radon diffusion coefficients measurements in more than 300 insulating materials are summarized. We have found out that great differences exist in diffusion properties because the diffusion coefficients vary within eight orders from 10^{-15} m^2/s to 10^{-8} m^2/s. Various possibilities of application of the radon diffusion coefficient for the design of radon barrier materials are discussed. Setting strict limits for maximal radon diffusion coefficient or minimal thickness of membranes results in significant reduction of the amount of materials that can be used for protection against radon. Calculation of the membrane thickness based on the radon diffusion coefficient and particular soil conditions and building characteristics seems to be the most effective and convenient approach.

INTRODUCTION

Some damp-proof or waterproof membranes placed over the entire surface of the house substructure can prevent radon from entering buildings from the soil. However the selection of effective radon barriers from the total amount of tanking materials is very difficult due to the lack of information about radon diffusion through these materials. Radon diffusion coefficient is a material property that determines this transport and therefore it can be used for the proper selection of radon-proof membranes based on a quantitative parameter assessment.

Testing of barrier properties of tanking materials against radon by means of the radon diffusion coefficient started in the Czech Republic in 1995 according to the method introduced by the Faculty of Civil Engineering of the Czech Technical University in Prague in cooperation with the National Radiation Protection Institute in Prague. Up to now a quite excellent database of results is available (more than 300 materials obtained throughout Europe and Canada, have been measured). This enables to make some general requirements considering the applicability of the radon diffusion coefficient for the design of radon-proof membranes.

DETERMINATION OF THE RADON DIFFUSION COEFFICIENT

The Czech test method, which is accredited by the Czech Accreditation Institute, is based on the determination of the radon flux through the tested material placed between two cylindrical containers. Radon diffuses from the lower container, which is connected to the radon source, through the sample to the upper container. From the known time dependent curves of the radon concentration in both containers the radon diffusion coefficient can be calculated. Radon concentration in both containers that serve as ionisation chambers operating in current mode is measured continuously by fully automatic measuring device enabling monitoring in very short time intervals (from 1 minute). Detailed description of the measuring technique, which schematic drawing is presented in Fig. 1, can be found in (Jiránek, 2008).

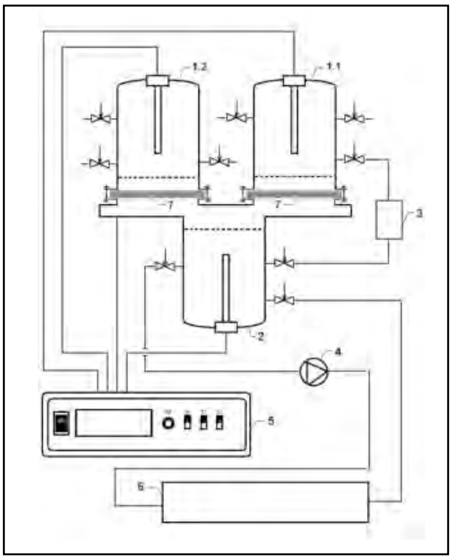

Figure 1: Schematic drawing of the new device (1.1, 1.2 – upper (receiver) containers, 2 – lower (source) container, 3 - pressure difference sensor, 4 - pump, 5 – control and operation unit, 6 – radon source, 7 – tested sample).

In order to obtain reliable values of the radon diffusion coefficient from the measured data, the calculation must be based on an appropriate mathematical model reflecting the time dependent radon concentration curves not only in the source and receiver containers, but also in the membrane itself. For the analysis of the radon concentration curve measured in the upper container we use the time dependent numerical modelling of the non-steady state radon diffusion through the membrane. Applied numerical model simulating the whole measuring process solves the one-dimensional diffusion equation:

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

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

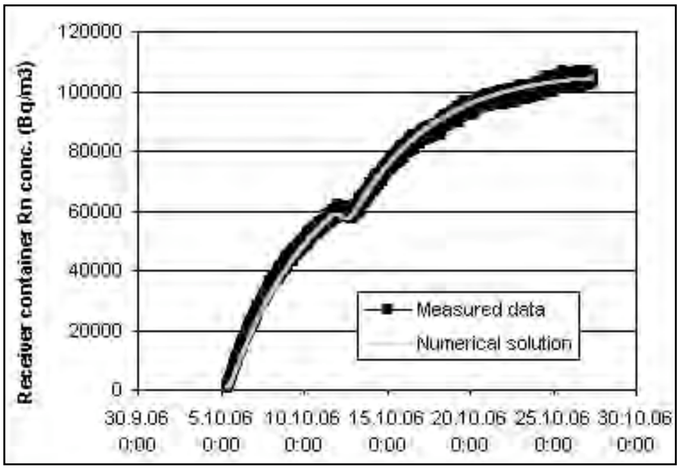

Figure 2: Fitting the numerical solution to the measured concentration in the receiver container.

Radon diffusion coefficient is derived from the process of fitting the numerical solution to the measured curve of radon concentration in the receiver container (Fig. 2). The great advantage of the applied mathematical solution is that it enables to determine the radon diffusion coefficient from data obtained by all known measuring modes used throughout Europe. This is very helpful, especially in the situation when no uniform measuring method exists within Europe.

VALUES OF THE RADON DIFFUSION COEFFICIENT

Results of the radon diffusion coefficient measurements are summarized in Fig. 3. On x-axis materials are grouped into categories according to the chemical composition.

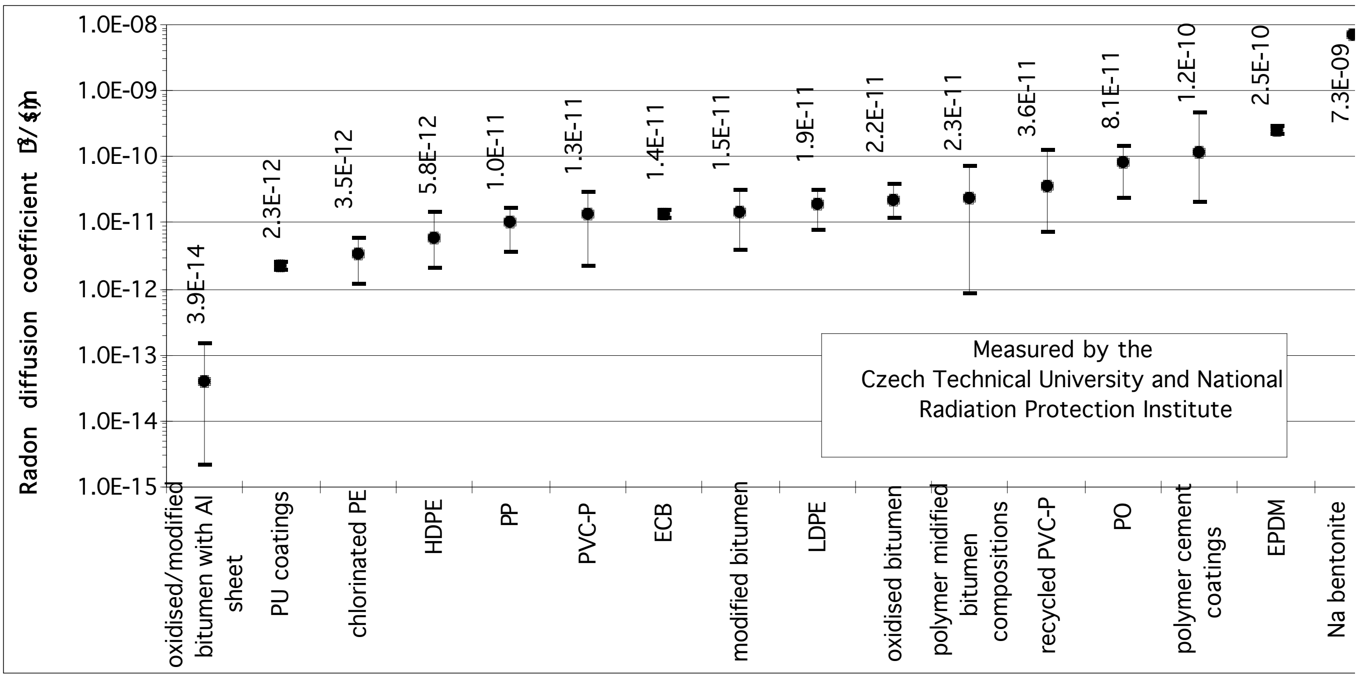

Fig. 3: Summary of the radon diffusion coefficient measurements realized in various waterproofing materials. On x-axis materials are registered with respect to rising order of their diffusion coefficients (HDPE - high density polyethylene, LDPE - low density polyethylene, PVC-P - flexible polyvinyl chloride, PP - polypropylene, PO – polyolefin, PU – polyurethane, ECB - ethylene copolymer bitumen, EPDM - ethylene propylene dien monomer).

Fig. 3 shows very clearly that in common insulating materials used for protection of houses against radon the diffusion coefficients vary within eight orders from 10^{-15} m^2/s to 10^{-8} m^2/s. The lowest values were obtained for bitumen membranes with Al foils no matter whether the bitumen was modified or not. On the other hand the highest values of the radon diffusion coefficient were discovered for sodium bentonite placed between paper or geotextile sheets, rubber membranes made of EPDM, polymer cement coatings and polyolefin membranes. Radon diffusion coefficient for the majority of materials varies in the range 3 X 10^{-12} and 3 X 10^{-11} m^2/s.

From Fig.3 it is also evident that relatively long scatter lines were obtained for two material categories – for bitumen membranes with Al foils and for polymer modified bitumen compositions. In case of membranes with Al foils it is caused by different thickness of Al foils. However in the category of bitumen compositions it is a result of different chemical composition of each material. It means that on the lower end of the scatter line materials with very good barrier properties can be found, but on the upper end there are materials, which could be hardly considered as radon-proof. Since the mean value lies close to the upper end, we can assume that majority of these materials will not work satisfactorily.

DESIGN OF RADON-PROOF MEMBRANES

Radon diffusion coefficient has been adopted in several countries (Czech Republic, Germany, Spain, Netherlands, Ireland, etc.) as a suitable parameter for the design of radon-proof membranes. However the application of this parameter differs from country to country. In general we can find three different approaches how to use the diffusion coefficient for the design of membranes:

1. the radon diffusion coefficient D of the radon-proof membrane must be below the strict limit value,
2. the thickness d of the radon-proof membrane must be at least three times greater than the radon diffusion length l (Keller) calculated as $l = (D/\lambda)^{1/2}$,
3. the thickness of the membrane is calculated for each house (Jiránek, Hůlka, 2000, Jiránek, Hůlka, 2001, Jiránek, 2004, CTS, 2006) according to the radon diffusion coefficient in the membrane, radon concentration in the soil on the building site and house parameters (ventilation rate, area in contact with the soil).

Limit for the maximal value of the radon diffusion coefficient

The main problem connected with this approach is how to choose correctly the limit value. To be safe and reliable under all circumstances (for all types of houses and radon concentrations in the soil) it should be rather lower than higher. However the lower the limit will be, the more materials will be of no use. For a typical single-family house, typical soil gas radon concentration and typical thickness of the membrane the maximum value of the radon diffusion coefficient should be 1 X 10^{-11} m^2/s. As a consequence of this the protection against radon will be solved preferably by materials with Al foils, which is from the technical point of view meaningless, because membranes with Al foils feature very low elongation and therefore they can very easily loose their barrier properties by destroying of the Al foil.

Limit for the minimal thickness of the membrane

Limits for the minimal thickness of membranes are derived from the assumption that most radon atoms will decay before they pass through the insulation, if the thickness of the insulation is greater than the diffusion length. However the condition that the insulation thickness should be at least three times greater than the diffusion length that is applied in some countries (Keller) leads to the enormous thickness of membranes. The required thickness exceeds the production thickness, if the radon diffusion coefficient is higher than 1 X 10^{-12} m^2/s in case of plastic membranes or 4 X10^{-12} m^2/s in case of bitumen membranes. This simply leads to the conclusion that the requirement $d \geq 3l$ is stricter than the previously described limit for D and will be met by a considerably smaller group of materials.

CALCULATION OF THE MEMBRANE THICKNESS

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 the insulation. Based on this simplification the highest permissible radon exhalation rate into the house, E_{lim}, can be expressed by equation (2):

$$E_{\lim} = \frac{C_{dif}.V.n}{A_f + A_w} \qquad (\mathrm{Bq/m^2h}) \qquad (2)$$

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 reference level for indoor radon concentration C_{ref} caused by diffusion. The value of C_{dif} can be estimated, for example according to the Czech standard ČSN 730601 as 10% (CTS, 2006). This means that the importance of the diffusion is reduced to 10 % of C_{ref} and the remaining 90 % of C_{ref} is reserved for the accidentally occurring convection.

In this context, the thickness of the radon-proof insulation can be derived with respect to real geological and building characteristics from the condition that the radon exhalation rate E from the real insulation in a real house calculated according to equation (3) must be less or equal to the highest permissible radon exhalation rate E_{lim} calculated for that house, i.e. $E \leq E_{lim}$.

$$E = \alpha_1.l.\lambda.C_S \frac{1}{\sinh(d/l)} \qquad (\mathrm{Bq/m^2h}) \qquad (3)$$

where C_s is the radon concentration in the soil gas (Bq/m^3) measured on the building site, 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) and α_1 is the safety factor that should eliminate the inaccuracies arising during the soil gas radon concentration measurements. Values of α_1 can be estimated according to the soil permeability (for highly permeable soils α_1 = 7, for soils with medium permeability α_1 = 3 and for low permeable soils α_1 = 2,1).

On the assumption that the insulation is homogeneous, its minimal thickness can be calculated from equation (4) obtained after the replacement of E in the equation (3) by E_{lim} from equation (2).

$$d \geq l.\mathrm{arcsinh}\frac{\alpha_1.l.\lambda.C_s.(A_f + A_w)}{C_{dif}.n.V} \qquad (\mathrm{m}) \qquad (4)$$

The great advantage of this approach is that the design of the radon-proof membrane can be fitted according to particular conditions (soil and building characteristics). The possibility of under- or over-dimensioning is thus strongly reduced. This method of the radon-proof insulation design was thoroughly verified in practice because it has been used in the Czech Republic since 1996.

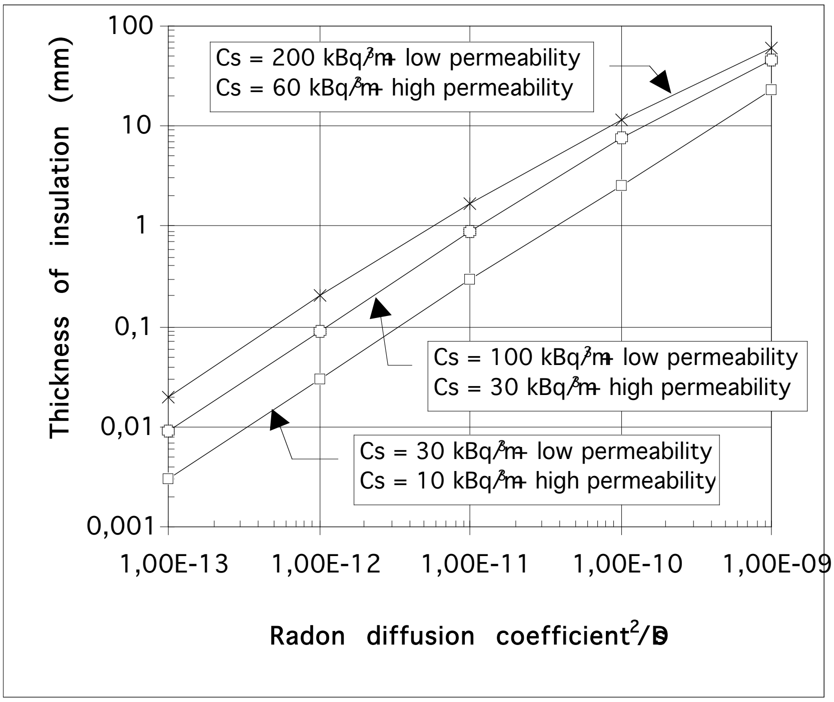

Fig. 4; Thickness of the insulation calculated according to the equation (6) for different values of D and various combinations of soil gas radon concentration and soil permeability. The influence of the soil permeability is introduced by the safety factor α_1 that increases proportionally with the permeability. Chart is valid for the house with habitable rooms in the basement.

The principle of designing according to this method can be identified from Fig. 4 in which the thickness of the insulation is plotted as a function of the radon diffusion coefficient and various combinations of soil gas radon concentration and soil permeability. It is clear that the thickness of the insulation with *D* lower than 10^{-12} m^2/s can be only several tenths of one millimetre, even in the areas with high radon concentration in the soil. Such small thickness is hardly producible and applicable due to sensitivity to puncturing and thus thicker insulation must be in practice used. On the other hand, the applicability of the insulation with *D* of order of 10^{-10} m^2/s will be very strongly dependent on building characteristics and the radon concentration in the soil. Membranes with *D* above 1.10^{-10} m^2/s are too permeable to be used for radon-proof insulation.

This clearly leads to the conclusion that the optimal value of the diffusion coefficient can be found within the interval 5.10^{-12} to 5.10^{-11} m^2/s. This interval corresponds with the production thickness of the 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).

CONCLUSIONS

Radon diffusion coefficient seems to be a convenient parameter for testing of radon-proof membranes and thus measurement of this parameter should be required for all insulating materials designated as a radon barrier.

Time dependent numerical modelling of the radon diffusion through membranes under the non-steady state conditions that simulates the whole measuring process provides evaluation of the radon diffusion coefficient with the highest accuracy.

Based on the experience from the Czech Republic controlling applicability of membranes by setting strict limits for the maximal value of the radon diffusion coefficient or the minimal thickness of the membrane is not a convenient approach. It seems to be reasonable to replace strict limits by the real design of the insulation in dependence on particular building and soil characteristics. Radon diffusion coefficient plays a crucial role in this design, because it enables to calculate the membrane thickness and the radon exhalation rate from the membrane.

In countries, where radon prone areas are classified according to indoor radon data and where measurements of the radon concentration in the soil gas are not so common, evaluation of C_s can be a problem. However it can be overcome, if we realize that in fact radon prone areas substitute real soil gas concentrations. Therefore an appropriate value of the radon concentration in the soil can be added to the each type of the radon prone area. The uncertainty of this procedure can be covered by the safety factor α_1.

Design of radon resisting membranes should be more complex. It should be stressed that barrier properties of membranes should be in balance with other very important properties such as durability, flexibility, buildability, chemical resistance, etc.

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REFERENCES

1. Jiránek M., Hůlka J.: Radon Diffusion Coefficient in Radon-proof Membranes – Determination and Applicability for the Design of Radon Barriers. In: International Journal on Architectural Science, Vol. 1, No. 4, p. 149 – 155, 2000
2. Jiránek M., Hůlka J.: Applicability of various insulating materials for radon barriers. In: The Science of the Total Environment 272 (2001), pp. 79-84
3. Jiránek M.: Testing and Design of Radon Resisting Membranes Based on the Experience from the Czech Republic. In: Proceedings from the 4th European Conference on Protection against Radon at Home and at Work. Praha 28.6.-2.7.2004
4. Jiránek M., Froňka A.: New Technique for the Determination of Radon Diffusion Coefficient in Radon-proof Membranes. In: Radiation Protection Dosimetry, 2008, doi:10.1093/rpd/ncn121
5. Fernández P.L., Quindós L.S., Sainz C., Gómez J.: A Theoretical Approach to the Measurement of Radon Diffusion and Adsorption Coefficients in Radonproof Membranes. In: Nucl. Instr. and Meth. in Phys. Res. B, 2003
6. Keller G.: "Radonisolierte Häuser" – Bautechnische Vorschläge für den Neubau und für Sanierungsmaßnahmen. Universität des Saarlandes, Homburg
7. Labed V., Rannou A., Tymen G.: Study of ^{222}Rn Permeation through Polymer membranes: Application to Continuous Measurement of ^{222}Rn in Water. In: Health Physics 1992, Vol. 63, N.2, pp.172-178
8. Spoel W.H.: Radon Transport in Sand. A Laboratory Study. Technische Universiteit Eindhoven, 1998
9. Czech Technical Standard ČSN 73 0601 "Protection of Buildings against Radon from the Soil". Czech Standards Institute, Praha, 2006