The Radon Diffusion Length as a Criterion for the Radon Tightness

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INTRODUCTION

The rising public wish for natural residing, new rules for energy and resource saving construction and the interest of the industry on inexpensive raw materials lead to new developments in the field of building materials. In this connection, the isolation of buildings against radon, especially in radon prone areas, shows an inclining interest. A recent German case control study on indoor radon (1) as well as extrapolation of data from uranium underground miners (2) suggest a relevant contribution of indoor radon to the lung cancer risk in the general population. Higher radon concentrations indoors usually depend on the possibility, that radon can penetrate from the surrounding soil. In most cases, the intrinsic radon exhalation of the materials can be neglected (3).

We measure routinely building and isolation materials to determine the diffusion coefficient and decide, whether the materials are radon tight or not. Naturally, the tightness of the material depends on its, so we use the diffusion length, respectively the dimensionless quotient between the diffusion length and the thickness as a criterion.

EXPERIMENTAL SETUP

The arrangement of the diffusion measurement is schematically shown in figure 1. One metallic hemisphere is used as reservoir and the other one as measurement volume. Between these hemispheres the sample is placed and tightened to avoid leakage. The lower reservoir chamber is continuously filled with radon from a dry radium-226 source to get a high and constant concentration.

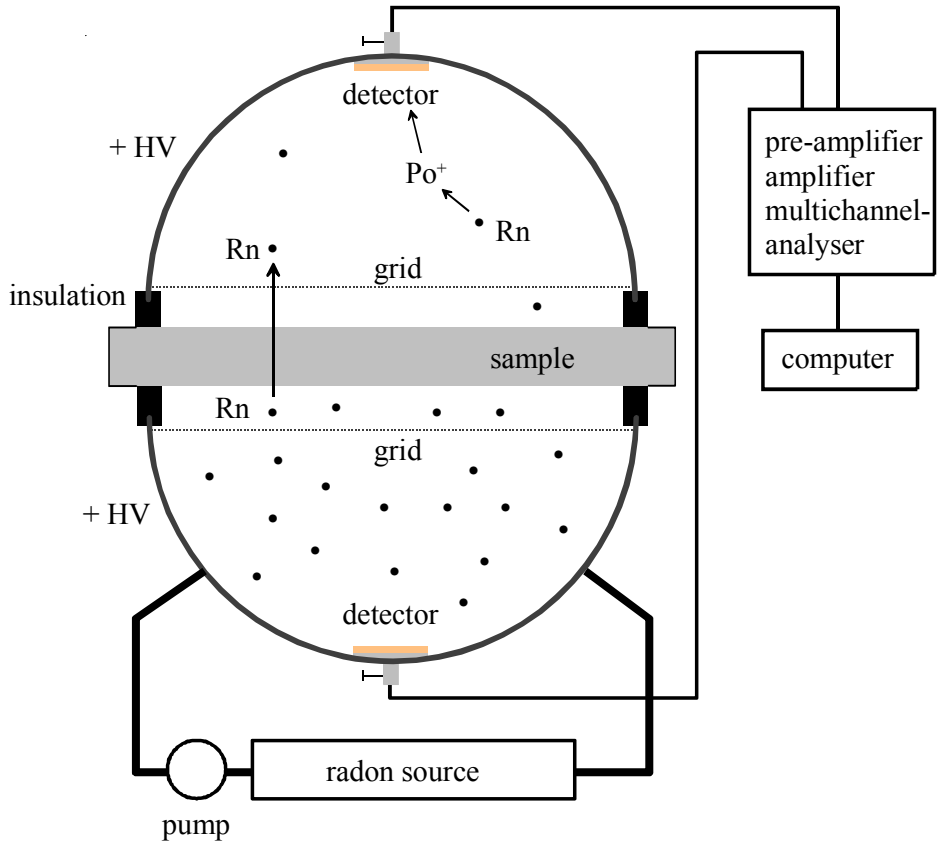

Figure 1. Principle of the measuring method for determining the radon diffusion coefficient

The radon concentrations in both chambers are determined alpha-spectroscopically using surface barrier detectors. The radon progeny Polonium-218 is positively charged and can be electrostatic deposited onto the

detector by an electrostatic field between the hemispheres and the grids on the one hand and the grounded detectors on the other hand. Alpha spectrums in several successive time intervals deliver the constant concentration inside the reservoir and the increasing concentration inside the measuring chamber.

Before the measurement, the sample is placed on the reservoir and the air inside the reservoir is mixed with radon. After a constant concentration gradient between the air inside the reservoir and the free side of the sample is reached the measuring chamber is placed on top of the sample.

The measurements were made under laboratory conditions. Generally, the samples were stored for minimal two weeks under normal conditions (temperature about 20 °C, relative humidity about 40-60 %). Especially the content of pore water can influence the diffusion, therefore the reactions (binding of water) inside the samples had to be stopped and an equilibrium between the humidity inside and outside had to be reached.

THEORETICAL BACKGROUND OF THE MEASUREMENT

The principle of the measurement is to observe the radon flow from a chamber with high radon concentration through the medium to a second chamber with a much lower radon concentration. The diffusive flow of radon through porous media is described by the Fick's law. In the case, where the transportation of gas is considered only in one direction and the production of radon inside the media can be neglected, the differential equation for the diffusion is

$$\frac{\partial c(x,t)}{\partial t} = D\frac{\partial^2 c(x,t)}{\partial x^2} - \lambda c(x,t) \tag{1}$$

$c\,(x,t)$ = concentration of radon inside the sample (Bq/m^3),
D = constant diffusion coefficient for radon (m^2/s),
λ = decay constant of radon (s^{-1}).

In the approximation of a time-independent steady-state condition (4) with an unbound exhalation, the solution of (1) is

$$c(x) = \frac{1}{2\sinh(d/l)}\left[\left(c_d - c_0 e^{-d/l}\right)e^{x/l} + \left(c_0 e^{d/l} - c_d\right)e^{-x/l}\right] \tag{2}$$

$c_0 = c\,(x = 0)$ concentration in the radon reservoir (Bq/m^3),
$c_d = c\,(x = d)$ concentration in the measurement chamber (Bq/m^3),
d = thickness of the material (m),
$l = (D/\lambda)^{1/2}$ diffusion length of radon (m).

The radon flux at the boundary to the measuring chamber is given by

$$\Phi = -Dp_{eff}\left.\frac{\partial c(x)}{\partial x}\right|_{x=d} \tag{3}$$

Φ = exhalation rate, the radon concentration flux (Bq/m^{2}s),
p_{eff} = effective porosity, the fraction of the sample volume, occupied by open pores.

Together with equation (2), the diffusion coefficient is

$$D = \frac{\Phi l \sinh(d/l)}{p_{eff}\left[c_0 - c_d \cosh(d/l)\right]} \tag{4}$$

The exhalation rate is determined by measuring the growing concentration in the measuring chamber, the time intervall between two succesive measurements and the area of the observed radon flux, which is here identical to the open area of the chambers (5). Due to the fact, that the diffusion length l is a function of D, the diffusion coefficient has to be calculated numerical with an iteration method.
Certainly, the diffusion coefficient is independent of the thickness. In contrast, the flux through a sample

decreases with the thickness. To get a thickness-independent parameter, which describes the tightness, we use the quotient of the sample thickness and the diffusion length. If the thickness of the sample is three times the diffusion length, less than 5% of the initial radon passes the sample and the material is called radon-tight.

RESULTS AND DISCUSSION

The results of our measurements of some building and insulating materials are shown in table 1. The results are rounded mean values of several specimen. In general, the diffusion coefficient of the investigated building materials are in the order of magnitude of about 10^{-9} to 10^{-6} m^2/s, corresponding to a diffusion length of about 6 to 100 cm. Therefore, the material had to be processed in an unrealistic thickness of up to 300 cm, to get a certain level of protection. Only some concrete, especially with polymer admixture, can hinder the diffusion. The polymer fills most of the pore system and closes the diffusion path through the material.

Insulating materials show a different behaviour. The diffusion coefficients are smaller than 10^{-11} m^2/s, thus the diffusion lengths are less than 2 mm. Surprisingly, the black butyl rubber was not radon tight, the high content of soot or graphite leads to an open structure.

Material	***Thickness*** $10^{-3}m$	***Diff. coeff.*** $10^{-6}\ m^2/s$	***Diff. length*** $10^{-3}m$	***Valuation***
Gypsum	100	2.35	1100	Permeable
Pumice	150	1.50	850	Permeable
Limestone	150	0.34	400	Permeable
Brick	150	0.35	400	Permeable
Sandstone	100	2.20	1000	Permeable
Aerated concrete	100	1.30	800	permeable
Heavy concrete	100	0.007	60	permeable
Polymer concrete PCC	40	$< 10^{-6}$	7	tight
Granite	30	0.053	160	permeable
Glass foam plate	70	$< 10^{-6}$	< 0.7	tight
Asphalt-asbestos	3	10^{-6}	0.7	tight
Bitumen	3	$< 10^{-6}$	< 0.7	tight
PEHD foil	1	$< 10^{-6}$	< 0.7	tight
Silicone rubber	3	$< 10^{-6}$	< 0.7	tight
Lead foil	0.1	$< 10^{-6}$	< 0.7	tight
Butyl rubber	1.5	10^{-5}	2	permeable
Polyurethane coating	5	$< 10^{-6}$	< 0.7	tight
Plastic foil	3	$< 10^{-6}$	< 0.7	tight
Epoxy resign	3	$< 10^{-6}$	< 0.7	tight

Table 1: Experimental mean values of the diffusion coefficient and the diffusion length of ^{222}Rn in some materials.

Up to now, the materials were investigated as samples, partially prepared only for these measurements. Therefore, no statements can be made about the sealing efficiencies after the use at the building site. For example, the examined plates, made from glass foam, were tight as well as the bitumen clued joints between them. In the case of a different adhesive or insufficient contingent of adhesive, the joints were permeable. And naturally, small pinholes in insulating foils can ruin the radon hindering behavior. The preparation of concrete and cement samples have to be done carefully and comparable to the situation on the building site. For example, core drilling

on selected samples showed, that inside the small plates (about 400 x 400 x 30 mm^3) the material was not always sufficient consolidate.

To get an assessment of the effect of a building renovation with a "radon-tight" insulation, the contribution of the radon flux from the surrounding soil to the concentration inside the building in a radon prone area was estimated. If the radon concentration inside the soil is in the order of magnitude of 100 kBq/m^3 and the whole cellar is "radon-tight", i.e. for all walls and for the floor, the criterium $d/l \geq 3$ is fulfilled, the calculated exhalation of radon Φ is less than 0,001 Bq/m^2s = 3,6 Bq/m^2h. The contribution of this exhalation to the radon concentration in the cellar can be estimated to

$$C_{Rn} \approx \Phi \cdot S \cdot \nu^{-1}, \quad (5)$$

S = ratio between the surface and the volume of the cellar (m^{-1}) = $2m^{-1}$,
ν = ventilation rate (h^{-1}) = $0{,}4h^{-1}$.
In this case, the radon flux results to a concentration of 18 Bq/m^3. Therefore, the criterium is usefull and can fulfill the desired quality of the radon mitigation.

SUMMARY

Most of the materials used to isolate a building against humidity and thermal flow, are radon tight. But a critical point of interest is the processing in general and especially the junctions between the building elements. Welding seams and adhesive sealings must be proper processed to keep the radon hindering capability. Unfortunately, standard concrete as well as other construction materials do not hinder the migration of radon in a sufficient way. It is also necessary to consolidate the concrete carefully to avoid a strongly connected pore system and therefore a diffusion path. Up to now, only polymer concretes or a combination of concrete and a foil respecively a polyurethane coating shows a behaviour, suitable for restoration of radon burdened houses. The investigated materials point out, that it is not possible to declare a group of material generally as „radon-tight" without measuring. Even small changes in the composition, for example the admixture of graphite to color a rubber, can alter the diffusion properties.

REFERENCES

1. H.E.Wichmann, L.Kreienbrock, M.Kreuzer, M.Gerken, D.Gingerkus, J.Wellmann, G.Keller, *Lungenkrebsrisiko durch Radon in der Bundesrepublik Deutschland (West)*, Fortschritte in der Umweltmedizin, ecomed Verlagsgesellschaft, Landsberg (1998).
2. J.H.Lubin, J.D. Boice, C.H.Edling, R.Hornung, G.Howe, E.Kunz, A.Kusiak, H.I.Morrison, E.P.Radford, J.M.Samet, M.Tirmarche, A.Woodward, Y.S.Xiang, D.A.Pierce, *Radon and Lung Cancer Risk: A Joint Analysis of 11 Underground Miners Studies*, US National Institutes of Health, NIH publication No. 94-3644 (1994).
3. G.Keller, H.Muth*, Radiation Exposure in German Dwellings, Some Results and a Proposed Formula for Dose Limitation*, The Science of the Total Environment 45, 299-306 (1985).
4. K.H.Folkerts, G.Keller, H.Muth, *Experimental Investigations on Diffusion and Exhalation of ^{222}Rn and ^{220}Rn from Building Materials*, Rad. Prot. Dosim. 7(1-4), 41-44 (1984).
5. G.Keller, K.H.Folkerts, H.Muth, *Method for Determination of ^{222}Rn (Radon) and ^{220}Rn (Thoron) Exhalation Rates using Alpha-Spectroscopy*, Rad. Prot. Dosim. 3(1/2), 83-89 (1982).