

NEW TECHNIQUE FOR THE DETERMINATION OF RADON DIFFUSION COEFFICIENT IN RADON-PROOF MEMBRANES

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This paper describes a new device and a method to determine the radon diffusion coefficient in damp-proof membranes developed in the Czech Republic. The main advantage of the device is that it enables tests to be carried out in all the known measuring modes used throughout Europe. Two recently developed computer programs are presented for the numerical modeling of the time-dependent radon transport through damp-proof membranes. According to this method, the radon diffusion coefficient is derived from the process of fitting the numerical solution to the measured curve of radon concentration in a receiver container. Numerical simulation and measured data are also compared. Reasons for disagreements between different methods and specific configurations of the measuring device are also discussed.

INTRODUCTION

Testing of barrier properties of waterproofing membranes against radon is usually based on the measurement of the proportionality coefficient D in the equation describing one-dimensional radon transport through the material tested. As this coefficient describes the diffusive properties of radon, it is mostly known as the radon diffusion coefficient^(3,4). However, some authors refer to this parameter as the permeability⁽⁶⁾ or the permeation coefficient⁽⁵⁾.

Measurement of the radon diffusion coefficient on a commercial basis started in the Czech Republic in 1995 according to the method K124/02/95 accredited by the Czech Accreditation Institute⁽¹⁾. Up to now, a quite excellent database is available (more than 200 materials obtained throughout Europe have been measured)^(2,7). The Czech method is based on measuring the radon flux of 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 curve of the radon concentration increase in the upper container, the radon diffusion coefficient can be calculated.

ELEMENTARY FEATURES AND DETECTION PROPERTIES OF THE NEW MEASURING DEVICE

The growth of radon concentration in the upper container was formerly measured discontinuously by extracting of air samples and transferring them into Lucas cells. Since 2006, when a new fully automatic measuring device was developed, radon

concentrations on both sides of the membrane have been measured continuously by ionisation chambers, which function at the same time as the containers. The schematic diagram of the new device is presented in Figure 1.

The detection principle is based on an ionisation chamber operating in the current mode with a sensitive detection volume of 2.2 l. The operating voltage of the detector is 150 V and the dynamical measuring range is from 100 Bq m⁻³ to 10 MBq.m⁻³. The ionisation current is amplified and then electronically and statistically processed. The processed results are subsequently displayed and stored in the memory. The electrometer device, the control and the operation unit, enables the current signal record each 2 s, and thus the radon concentrations can be monitored at 1 min intervals. The presented device, therefore, enables a very detailed and precise studying of time variations of radon concentrations. This is the biggest advantage of this system compared with the former one.

NEW METHOD FOR THE DETERMINATION OF THE RADON DIFFUSION COEFFICIENT

Method of measurement

The definite increase in the radon concentration in the upper container is measured under the steady-state conditions. After radon is admitted into the lower container, radon concentration starts to increase in the upper container however, under non-steady state conditions. When the steady-state radon concentration profile is set up within the membrane (assuming that the system is in the steady state after reaching a time much longer than the relaxation time¹), the upper container is flushed with radon-poor ambient air and the container is closed again. Since then, the

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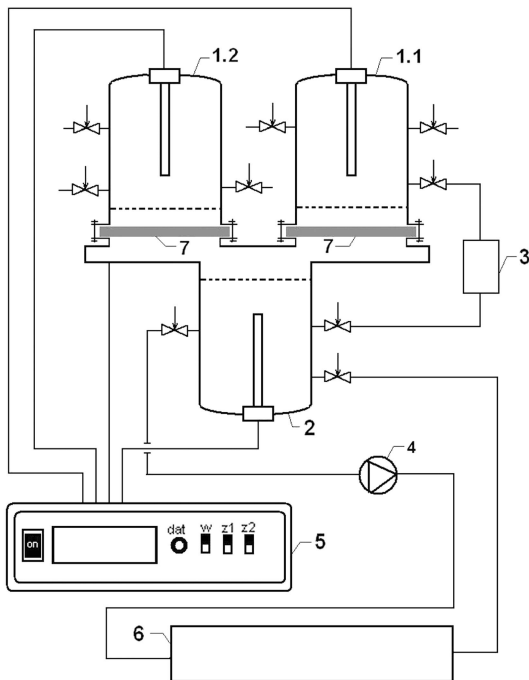


Figure 1. Schematic drawing of the new device [1.1, 1.2—upper (receiver) containers, 2—lower (source) container, 3—pressure difference sensor, 4—pump, 5—control and operation unit, 6—radon source, 7—tested sample].

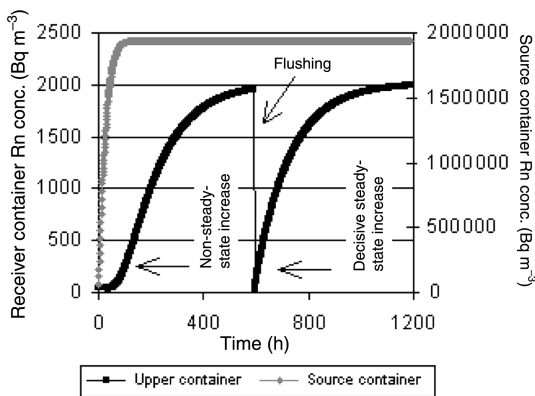


Figure 2. Phases of the measurement.

definite increase in the radon concentration in the upper container can be measured (Figure 2).

Evaluation of the diffusion coefficient from measured data

A numerical computer program has been developed to analyse the radon concentration measured in the

upper container. The program is able to find the most appropriate value of the radon diffusion coefficient of the tested material. Diffusion coefficient is derived from the process of fitting the numerical solution to the measured curve of radon concentration in the receiver container, using the inserted radon concentration curve. The program had been successfully verified by comparing the results with the analytical solution of simple transport cases. Furthermore, model predictions compare favourably with the measurements.

The program solves the one-dimensional diffusion equation by describing time-dependent radon transport through the tested material:

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

where D is the radon diffusion coefficient ($\text{m}^2 \text{s}^{-1}$), λ the radon decay constant $2.1 \cdot 10^{-6} \text{s}^{-1}$, C the radon concentration within the membrane (Bq m^{-3}) and t the time (s).

The solution of equation (1) is derived using the finite-element method. Boundary conditions on both sides of the membrane are defined according to the equation:

$$-D \frac{\partial C}{\partial x} = h \cdot (C_s - C_n) \quad (2)$$

where h is the radon transfer coefficient (m s^{-1}), C_s the radon concentration on the membrane surface (Bq m^{-3}) and C_n the radon concentration in either lower or upper container (Bq m^{-3}) depending on which surface the condition is applied. The calculation proceeds in a number of time steps. At the end of each step, the radon exhalation rate from membrane (3) and the radon concentration in the upper container are calculated (4).

$$E_{h,i-1} = h \cdot (C_{s,i-1} - C_{h,i-1}) \quad (3)$$

where $E_{h,i-1}$ is the radon exhalation rate from the membrane to the upper container in time t_{i-1} ($\text{Bq m}^{-2} \text{s}^{-1}$), $C_{s,i-1}$ the radon concentration on the membrane surface in time t_{i-1} (Bq m^{-3}) and $C_{h,i-1}$ the radon concentration in the upper container in time t_{i-1} (Bq m^{-3}).

$$C_{h,i} = C_{h,i-1} \cdot e^{-(\lambda+n_h)\Delta t} + \frac{E_{h,i-1} \cdot A}{V_h \cdot (\lambda + n_h)} \cdot (1 - e^{-(\lambda+n_h)\Delta t}) \quad (4)$$

where $C_{h,i}$ is the radon concentration in the upper container in time t_i (Bq m^{-3}), n_h the ventilation rate of the upper container (s^{-1}), Δt the time difference between t_{i-1} and t_i (s), A the area of the membrane sample (m^2) and V_h the volume of the upper container (m^3).

RADON DIFFUSION COEFFICIENT IN RADON-PROOF MEMBRANES

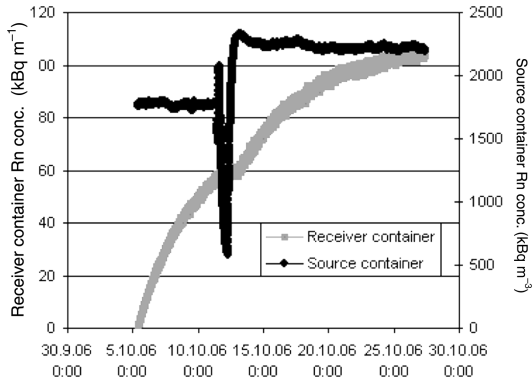


Figure 3. Radon concentrations in the source and receiver containers measured for the membrane 1.5 mm thick after reaching the steady-state concentration profile within the membrane.

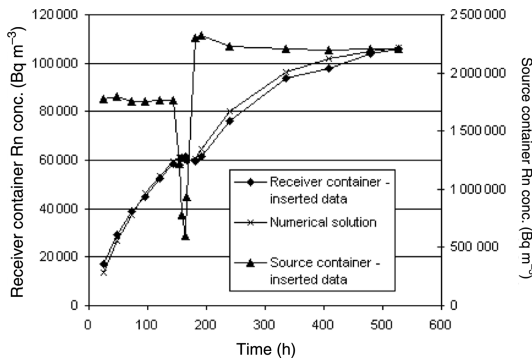


Figure 4. Measured curves from Figure 3 broken into the straight lines data of which is given to the program. The numerical solution results from the least-squares fit of the numerical model to the measured data.

The whole process of the radon diffusion coefficient determination is further illustrated by means of measurements carried out on a PVC membrane 1.5 mm thick. Figure 3 shows time-dependent curves of radon concentration measured for this membrane in both containers. It can be seen that changes in the radon concentration in the source container have direct influence on the concentration in the upper container. For simplification the curves were broken into straight lines (Figure 4). These data, together with the membrane thickness, its surface area and volume of the receiver container, are given to the program. For different diffusion coefficients, the program performs calculations of the radon concentration curves in the upper container. Radon diffusion coefficient corresponding to the curve with the least-squares fit to the measured data is identified as the right coefficient (in this case $D = 2,6 \cdot 10^{-11} \text{ m}^2 \text{ s}^{-1}$).

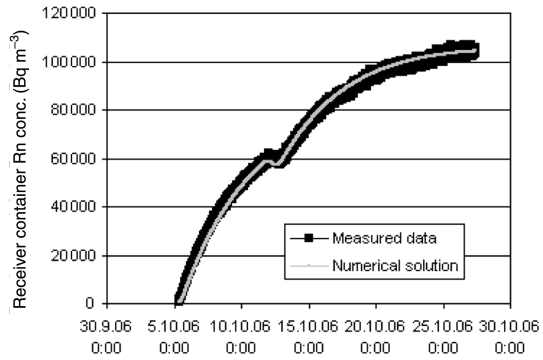


Figure 5. Correlation between the numerical solution obtained from the program TransRn and the measured concentration in the receiver container.

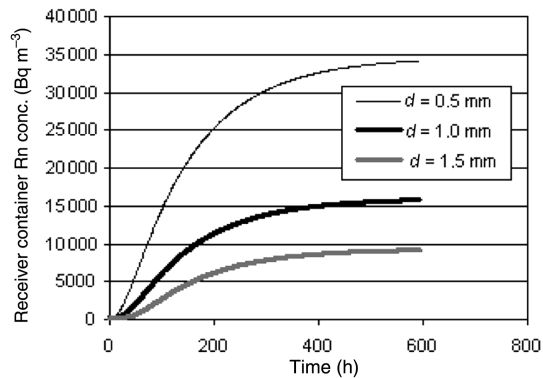


Figure 6. Radon concentrations increase in the receiver container in accordance with the membrane thickness.

The final step includes the verification of the numerical solution by comparison with measured curve of radon concentration in the upper container (Figure 5).

COMMENTS ON MEASURING METHODS

At present times, different measuring methods are available in Europe. According to those methods, the build-up of radon concentration in the receiver container is measured immediately after the test is commenced, i.e. under the non-steady-state conditions; furthermore, it is assumed that the build-up is linear with time and the test duration is only 2–6 days.

From the time-dependent numerical simulation, it can be concluded that such conditions cannot generate the correct value of the radon diffusion coefficient. From Figures 2, 6 and 7, it is obvious that the initial part of the concentration curve is not linear, because at the beginning of the test, the membrane

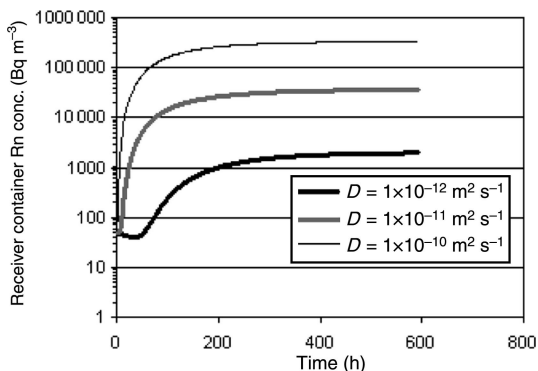


Figure 7. Radon concentration increase in the receiver container in accordance with the diffusion coefficient.

is not fully saturated with radon. The significance of this non-linear shape increases as the thickness of the membrane increases and radon diffusion coefficient decreases. The initial non-linear part can be observed within the first 4 days after the test has commenced. A linear approximation of measured data in this phase, especially if the duration of the test is shorter than 4 days, can lead to serious mistakes and to underestimation of the diffusion coefficient (Figure 8). Such mistakes can be minimized by prolongation of the measuring time at least for 8 days and by better mathematical processing of measured data. Linear approximation should be replaced by the fitting of the numerical solution to the measured curve.

CONCLUSION

Thus, the new technique developed by the National Radiation Protection Institute and the Faculty of Civil Engineering of the Czech Technical University to measure the radon diffusion coefficient in radon-proof membranes is unique, due to the following features—very fast response to changes in radon concentrations, measurement of two samples and control of radon concentration in the lower container. The computer program developed evaluates the radon diffusion coefficient with the highest accuracy. Time-dependent numerical modelling of the radon diffusion through membranes can also be used for the clarification of possible effects that can influence the accuracy of any test method used for the determination of the radon diffusion coefficient. This is very helpful, especially in the situation in which no uniform measuring method exists within Europe and when different values of the diffusion coefficients can be found for the same material. Disagreements are due to different durations of tests,

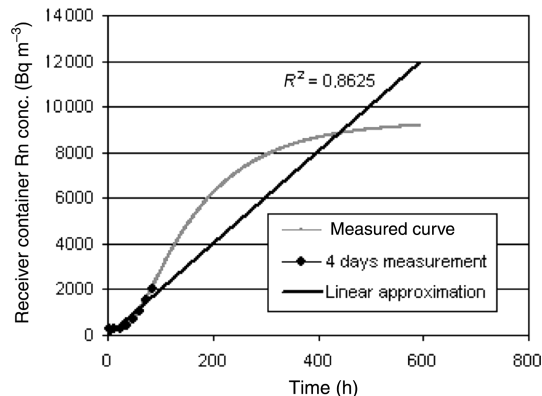


Figure 8. Underestimation of the radon diffusion coefficient caused by linear approximation of data measured within the first 4 days.

different boundary conditions under which the tests are carried out and different mathematical processing of measured data.

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