THE PROBLEM WITH TEMPERATURE DEPENDENCE OF RADON DIFFUSION CHAMBERS WITH ANTI-THORON BARRIER

DOBROMIR PRESSYANOV[1], DIMITAR DIMITROV[2]

[1] Faculty of Physics, Sofia University "St. Kliment Ohridski", Sofia, Bulgaria
[2] Mining and Geology University "St. Ivan Rilski", Sofia, Bulgaria
E-mail: pressyan@phys.uni-sofia.bg

Abstract. The response of radon diffusion chambers that use polymer membranes or small gaps/holes as anti-thoron barriers at different temperatures is studied. A theoretical model of radon diffusion through polymer foils is presented. Experiments were made at 2°C, 21.5°C and 45°C. Results show that polymer foils introduce significant temperature dependence in radon response. The chambers with small gaps/holes do not show significant temperature dependence but their response may be affected by humidity and air turbulence. A novel approach to overcome these problems and design chambers with efficient anti-thoron and anti-humidity barrier as well as compensated temperature dependence is discussed.

Key words: radon, diffusion chambers, radon permeability, compensated temperature bias.

1. INTRODUCTION

The implementation of the EC directive 2013/59/EURATOM [1] needs metrological/quality assurance of ^{222}Rn measurements at low concentrations. One problem addressed in the ongoing European MetroRADON project is to investigate and reduce the influence of thoron (^{220}Rn) on radon end-user measurements [2]. Widely used anti-thoron barrier for radon detectors is thin polymer foil that stops ^{220}Rn while allowing ^{222}Rn to diffuse through it in the detector volume [3–5]. As noted elsewhere [6, 7], albeit effective against thoron and humidity, such barrier may introduce significant temperature bias in the radon response due to the temperature dependence of the diffusion properties of polymer foils. Other anti-thoron barriers are based on small gaps/pin-holes, usually around the chamber cap, through which radon diffuses in their volume [8]. If sufficiently small, the gaps/holes can serve as diffusion barriers against thoron, but their performance at high humidity or under turbulent air conditions might be problematic [8, 9].

In this report we present an experimental and theoretical study of the influence of the temperature on the response of two kinds of diffusion chambers with track detectors for ^{222}Rn measurement. Both kind of chambers were metallic cylinders. In the first kind of chambers radon diffused through polyethylene foils into the volume. The second kind of diffusion chambers were where radon penetrated into the volume

by diffusion through small gaps/holes around the chamber cap. The alpha track detectors used in both kind of chambers were Kodak-Pathe LR-115 type II. Exposures to controlled ^{222}Rn concentrations were made at temperatures of 2°C, 21.5°C and 45°C using a dedicated laboratory facility [10]. A generalized theoretical model of radon diffusion through polymer foils was developed and applied to model radon and thoron penetration through polymer foils into the chamber volume. A novel concept to minimize the thoron influence on the passive radon detectors, not introducing in the same time strong temperature bias in the radon response is proposed and pilot experimental results that demonstrate its feasibility are shown.

2. THEORETICAL MODEL

Consider a closed volume V, where at least one of the sides is a foil of material through which radon can penetrate by diffusion, while the other sides are not permeable for radon. The total area of all permeable sides is S, the thickness of the polymer foil h, the diffusion coefficient of radon in the polymer material is D. Assume that radon can penetrate into the volume V only by diffusion through the permeable sides. The radon diffusion through the foil is described by the diffusion equation:

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

(1)

where c is radon concentration in the foil material and λ is the decay constant of the isotope of interest (^{222}Rn or ^{220}Rn). Equation (1) is considered with the initial condition $c(t = 0, x) = 0$ and border conditions $c(t, x = 0) = Kc_{out}(t)$ and $c(t, x = h) = Kc_{in}(t)$, where $x = h$ is the coordinate of the internal surface of the foil, $x = 0$ is that of its external surface. The quantities $c_{out}(t)$ and $c_{in}(t)$ are the ambient concentration outside the chamber and the concentration inside the chamber volume, respectively, and K is the partition coefficient of the foil material (the partition coefficient is the dimensionless solubility of the material that is the ratio on the border of the concentration in the material to that in air). The time evolution of the concentration inside the chamber volume is described by the equation:

$$\frac{dc_{in}}{dt} = \frac{SD}{V}\frac{\partial c}{\partial x}\bigg|_{x=h} - \lambda c_{in}.$$

(2)

The first term in the right-side of equation (2) describes the flux by diffusion (Fick's law) from the internal surface of the foil, while the second term is for the radioactive decay. Further passive (integrated) mode of measurement is modeled, in which the determined quantity is the time-integrated radon concentration for exposure time t_{exp}. The integrated quantities will be denoted by capital symbols, e.g.:

$$C = \int_0^T c(t)\,\mathrm{d}t, \tag{3}$$

where the integration time T is the time for which the detector in the chamber gathers signal. In our experiments, after exposure to radon the chamber is left at radon-free air (or at concentration that is of lower orders of magnitude than that during the exposure) until all ^{222}Rn (or ^{220}Rn) and its progeny atoms degas or decay, before analyzing the detectors. Under such circumstances the upper limit of integration can be replaced by ∞. Let's transform the equations (1,2) by integrating both sides:

$$\int_0^\infty \frac{\partial c}{\partial t}\,\mathrm{d}t = D\frac{\partial^2}{\partial x^2}\int_0^\infty c(x,t)\,\mathrm{d}t - \lambda\int_0^\infty c(x,t)\,\mathrm{d}t \tag{4}$$

$$\int_0^\infty \frac{\mathrm{d}c_{in}}{\mathrm{d}t}\,\mathrm{d}t = -\frac{SD}{V}\frac{\partial}{\partial x}\int_0^\infty c(x,t)\,\mathrm{d}t\Big|_{x=h} - \lambda\int_0^\infty c_{in}(t)\,\mathrm{d}t. \tag{5}$$

The left side of equations (4) and (5) is zero, as $c(t=0) = c(t=\infty) = 0$ and the same goes for $c_{in}:c_{in}(t=0) = c_{in}(t=\infty) = 0$. Then, the equations for the integrated quantities become:

$$D\frac{d^2C}{dx^2} - \lambda C = 0, \tag{6}$$

$$-SD\frac{\mathrm{d}C}{\mathrm{d}x}\Big|_{x=h} - \lambda C_{in}V = 0, \tag{7}$$

considered with border conditions:

$$C(0) = KC_{out}; \tag{8}$$

$$C(h) = KC_{in}, \tag{9}$$

where C_{out} is the integrated for the exposure time ambient ^{222}Rn concentration outside the chamber. By combining (7) and (9) one obtains:

$$C(h) = -\frac{KSD}{\lambda V}\frac{\mathrm{d}C}{\mathrm{d}x}\Big|_{x=h}. \tag{10}$$

It will be convenient to introduce the diffusion length of the particular isotope (^{222}Rn or ^{220}Rn) in the material: $L_D = \sqrt{D/\lambda}$. By standard methods the solution of (6) is obtained in the form:

$$C(x) = Ae^{x/L_D} + Be^{-x/L_D} \qquad (11)$$

At the given border conditions the expressions for the constants A and B are:

$$A = \frac{KC_{out}e^{-2h/L_D}}{1 + e^{-2h/L_D}\left(\dfrac{SP - \lambda VL_D}{SP + \lambda VL_D}\right)}\left(\frac{SP - \lambda VL_D}{SP + \lambda VL_D}\right); \qquad (12)$$

$$B = \frac{KC_{out}}{1 + e^{-2h/L_D}\left(\dfrac{SP - \lambda VL_D}{SP + \lambda VL_D}\right)}, \qquad (13)$$

where $P = KD$ is the "permeability" of the material through which radon diffuses.

This way, using also eqn. (7), for the integrated concentration in the volume one obtains:

$$C_{in} = C_{out}\frac{2SP}{SP + \lambda VL_D + (SP - \lambda VL_D)e^{-2h/L_D}}e^{\frac{h}{L_D}}. \qquad (14)$$

Actually, the signal of the detector is proportional to C_{in}, while "the calibration factor of the chamber" is the ratio "signal/C_{out}". To distinguish between the temperature dependence of radon penetration through the membrane, from eventual temperature dependence of the detector response, we will describe the first one by the "penetration ratio" R, which is the ratio of the integrated concentrations inside and outside the volume:

$$R = \frac{C_{in}}{C_{out}} = e^{-\frac{h}{L_D}}\frac{2SP}{SP + \lambda VL_D + (SP - \lambda VL_D)e^{-2h/L_D}}. \qquad (15)$$

For thin foils, when $h \ll L_D$ the expression for R simplifies to:

$$R \xrightarrow{\quad h \ll L_D \quad} R = \frac{1}{1 + \lambda\dfrac{hV}{PS}}. \qquad (16)$$

The last term of eqn. (16) is what one usually finds in the literature for the penetration ratio R [3, 4]. Notably, this expression is valid for thin membranes – whose thickness is less than the diffusion length of the considered radon isotope.

Because of the short half-life of [220]Rn its L_D is way smaller than that of [222]Rn. One membrane that is thin regarding [222]Rn can be thick regarding [220]Rn. For instance,

in the described experiments foils of *low density polyethylene* (LDPE) of thickness 75 μm and *high density polyethylene* (HDPE) of thickness 120 μm were used. At room temperature the diffusion length of ^{222}Rn in LDPE is about 1500 μm and 700 μm for HDPE [11]. In the same materials the diffusion length of ^{220}Rn in these materials are about 19 μm and 9 μm, correspondingly. Therefore, the used foils can be considered as "thin" regarding ^{222}Rn and "thick", regarding ^{220}Rn.

The difference between the diffusion length and half-life of radon and thoron makes it possible to use diffusion barriers, like polymer membranes, to discriminate between these isotopes. For instance, the penetration ratio in chambers of $V/S = 7.5$ cm (the same used in our experiments), and covered with 10 μm thick LDPE will be $R(^{222}\text{Rn}) = 0.86$ and $R(^{220}\text{Rn}) = 0.001$, *i.e.* the thoron influence on the signal of the detector placed inside the chamber in this case will be reduced by three orders of magnitude. With 75 μm thick LDPE the reduction will be almost five orders of magnitude. Such close to the absolute anti-thoron protection can hardly be achieved by other anti-thoron barriers. Another benefit of the use of polymer foils as a barrier is that they also provide an effective protection of the chamber volume against moisture/humidity.

The major problem in the use of membrane-based anti-thoron barriers arises from the temperature dependence of the penetration ratio R which makes their response to radon temperature dependent. Actually, the temperature dependence of R is due to the temperature dependence of the foil permeability P.

3. EXPERIMENTAL

Experiments at different temperatures were carried-out at the Laboratory of Dosimetry and Radiation Protection at the Sofia University "St. Kliment Ohridski". Diffusion chambers covered by foils were cylinders of diameter 80 mm and height 75 mm. At the bottom of the chamber a piece of detector Kodak-Pathe LR-115 type II was fixed. Chambers' openings were covered by different materials: filter paper, LDPE of thickness 75 μm or HDPE of thickness 120 μm. Special care was taken to check that the fixing of the foils (by hot and cold silicone, applied consecutively) was hermetic. For that purpose other chambers were covered by metal foil, not permeable for radon, which was fixed in the same manner as plastic foils. A set of chambers that contain at least one chamber of any kind (covered by filter paper, metal foil, foil of LDPE, foil of HDPE) were loaded for exposure in a 50 L exposure vessel. In the same vessel a second set of diffusion chambers of "gaps/holes" type were also loaded. Those chambers were of size $\varnothing 75 \times 75$ mm with SSNTDs of Kodak-Pathe LR-115/II and were traditionally used for radon monitoring at the Laboratory of Dosimetry and Radiation Protection (see the photo Fig. 1 in [12]). Details about their calibration and traceability are provided elsewhere [13]. Three different exposure to reference ^{222}Rn concentrations at three different temperatures: 2°C, 21.5°C and 45°C were made, using the dedicated calibration

facility [10]. During exposure the [222]Rn concentration was followed by a reference radon monitor AlphaGUARD PQ2000 Pro (Bertin/Saphymo GmbH). After exposure the set of chambers was left at the same temperature but at low radon levels ($\sim$ 20 Bq m^{-3}) for two weeks to allow practically full [222]Rn decay and out-gazing (from the materials where it can be absorbed – *e.g.* the polymers). After that the detectors were etched with 10% NaOH at 60°C for 100 min, followed by 30 min washing in agitating water and 2 min wash in still solution of 50% ethanol. After the detectors dried the tracks were counted visually, by a microscope.

As the track-density of the chambers covered by metal foils was equal to the background track density (21 ± 4 cm^{-2}), we consider the technique used to fix hermetically the foil to the chamber to be efficient and to guarantee that radon penetrates in the chamber volume only by diffusion through the polymer membrane. For the cans covered with filter paper $R = 1$ was assumed. This way, the penetration ratio for the other chambers was determined as the ratio of the detector signal (net track density) of the detector placed in foil-covered chamber to that from a chamber covered with a filter paper.

4. RESULTS AND DISCUSSION

Experimentally determined values of R at the three different exposure temperatures are shown in Fig. 1. Clearly, the response of the diffusion chambers with polymer foil strongly depends on the temperature. The results for the calibration factor (CF = net track density/integrated [222]Rn concentration (C_{out})) of the "pin-hole" diffusion chambers are shown in Fig. 2. For these chambers there is no statistically significant indication for temperature dependence of their response.
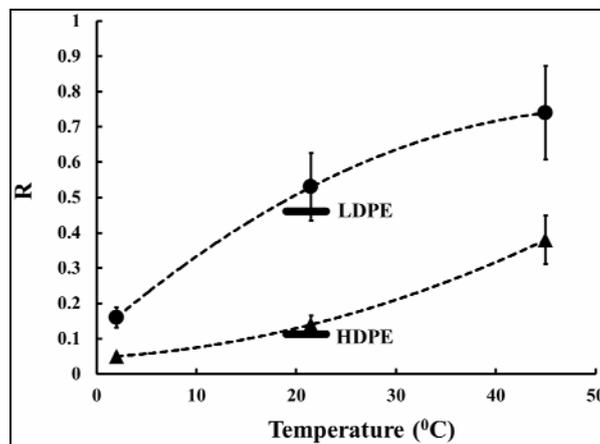

Fig. 1 – The penetration ratio at different temperatures of the chambers covered by LDPE (●) and HDPE (▲) with interpolation curves shown. The horizontal section-lines represent the estimates based on LDPE and HDPE data reported in [11]. As experiments [11] were made at a room temperature, that is typically within 19–23°C, the section-lines cover this temperature range.
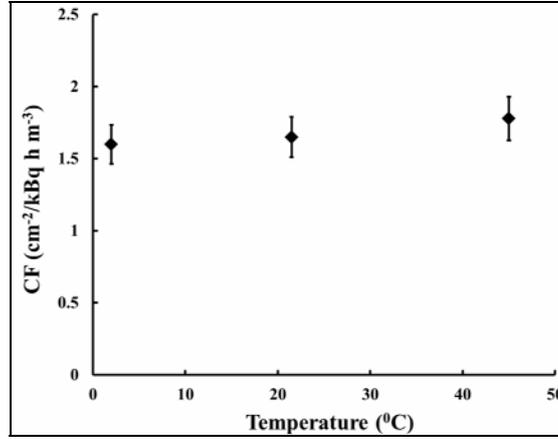
Fig. 2 – The CF of diffusion chambers in which radon diffuses through small gaps/holes.
No significant difference between the results at different temperatures are visible.

Despite the temperature bias, the polymer foils can provide an efficient anti-thoron and anti-humidity barrier. Therefore, we focused efforts on whether this dependence can be reduced or even eliminated. Using the expression for R for a specified material and the experimental value R_0 at given h_0, V_0, S_0 one can model R for the same material at the same temperature for different h, V, S using the expression that can be derived from eqn. (16):

$$R = \frac{1}{1 + \left(\dfrac{1 - R_0}{R_0}\right)\dfrac{hVS_0}{h_0V_0S}}.$$ (17)

The modeling of R for temperature different from 2°C, 21.5°C and 45°C were made by polynomial interpolation between the experimental points at 2°C, 21.5°C and 45°C.

By varying parameters of the foil-coverage and V/S ratio, the temperature dependence can be somewhat reduced, but not eliminated, as seen in Fig. 3. In addition when using thin polymers (e.g. $h \leq 10$ μm) care about their integrity during exposure may be needed. It appears that measurements with foil-covered diffusion chambers over wide range of ambient temperatures need to account for the temperature bias that can be substantial.

However, the identified problem of temperature dependence of radon permeability through polymer foils offers a surprising opportunity to compensate the temperature dependence of many kinds of radon detectors. There are many radon detectors whose sensitivity decreases with increasing the temperature. Those include e.g. detectors based on activated charcoal [14], detectors that use radon absorbers/adsorbers [15], the most widely used alpha track detectors of CR-39

which show fading that is greater at higher temperature [16]. The key concept is to combine the increase of the penetration ratio with the reciprocal decrease of the sensitivity of the radon detectors placed in the chamber.
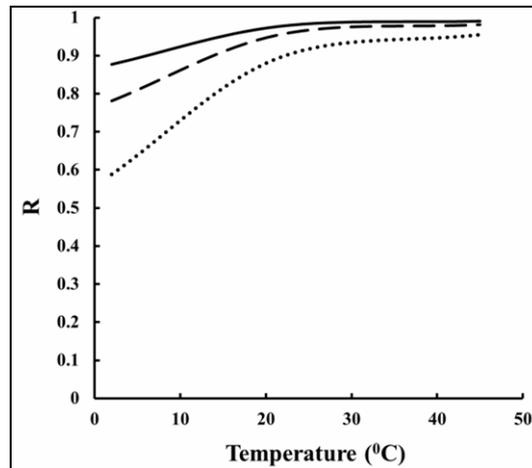

Fig. 3 – Modeled dependence of the penetration ratio (R) on the temperature for thin foils of LDPE and different volume to surface ratios. The solid line is for h = 10 μm and V/S = 1.5 cm, the dashed line is for h = 10 μm and V/S = 3.0 cm and the dotted line is for h = 50 μm and V/S = 1.5 cm.

The technical approach is to construct modules in which the increased permeability of radon inside with increasing temperature compensates the decreasing sensitivity to radon of detectors placed inside. The design of such "compensating modules" is described in the patent application [17]. Its potential can be demonstrated by a dedicated experiment carried out by the authors. One kind of detectors for which our experience indicated significant temperature dependence is those based on Kodak Pathe LR-115/II detectors covered with solid absorbers as radiators [15]. The best response with such detectors is obtained when the foil is of Makrofol N. This foil is of uniquely high radon absorption ability (K = 112 ± 12 at 20°C) [18]. In present experiments detector of Kodak-Pathe LR-115/II covered with 2 foils of 43 μm Makrofol N was used. However, when the foils of Makrofol N are used a strong temperature dependence of the response is observed, the signal decreases about 2.7 times when the temperature increases from 5 to 35°C. By technical concept described in [17] and modeling, using the present experimental data and equations (16, 17) it was found that if a module is designed with a covering foil of 120 μm thick HDPE and V/S = 1.5 cm, and the detector is placed inside, the temperature dependence should be compensated. The experimental results are shown in Fig. 4. As seen, such construction provides very good temperature compensation. Notably, we are at the very beginning of studies and the practical application of this approach for different kind of detectors is a matter for the future. However, the pilot experimental results suggest for its promising potential.
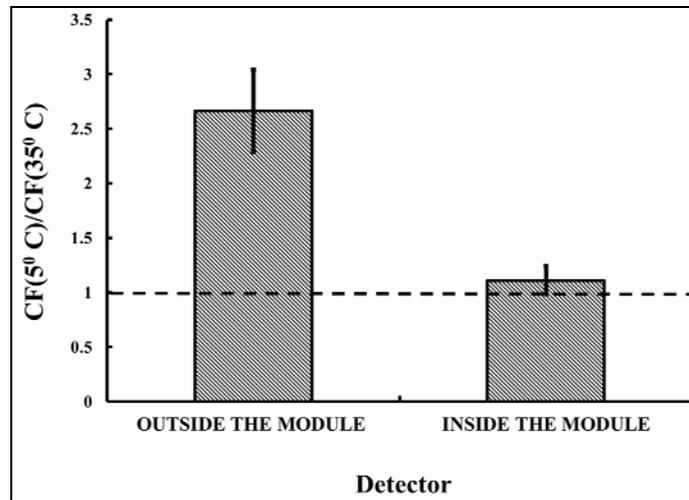
Fig. 4 – The CF ratio of 5°C to 35°C for detectors of Kodak Pathe LR-115/II covered
with Makrofol N. Outside the compensation module the temperature bias
is about 270% while inside it is within about 10%.

5. CONCLUSION

This work was focused on the temperature dependence of the response of
radon diffusion chambers with anti-thoron barrier. One kind of such chambers was
with polymer foils used to cover their volume. The foils used were of low density
polyethylene with thickness 75 μm or high density polyethylene of thickness 120 μm.
The second kind was "gap/ holes" kind of chambers. The experimental results
revealed strong temperature dependence of the chambers with polymer foils. The
temperature influence on the response of the other kind of chambers was not
significant. A theoretical model of radon penetration through polymer foils was
developed and used the radon penetration by diffusion through foils. The modeling
and the experimental data suggest that the temperature influence on the penetration
ratio of ^{222}Rn in chambers covered by polymer membranes can hardly be eliminated.

However, a surprising opportunity was found and explored. By combining
the temperature dependence of radon penetration ratio through polymer membranes
with the reciprocal temperature dependence of some widely used radon sensors/
detectors, a novel design of radon monitors with highly efficient anti-thoron/anti-
humidity barriers and compensated temperature dependence becomes possible. The
key-concept is to place the detector with decreasing with the temperature response
in a module in which the radon penetration ratio increases reciprocally thus the
temperature bias being compensated. The pilot experimental results with detectors
of Kodak-Pathe LR-115/II covered by absorber of Makrofol N showed that the
temperature bias between 5°C and 35°C is reduced from about 270% to within 10%.

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