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# Approaches for reduction of the temperature bias on radon detectors packed in anti-thoron polymer membranes

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## Abstract

Passive and active detectors used for radon ( $^{222}\text{Rn}$ ) measurements can be influenced by thoron ( $^{220}\text{Rn}$ ). Polyethylene membranes are very appropriate diffusion barriers for anti-thoron protection of such detectors. However, if not properly chosen, these membranes may not reduce efficiently the thoron influence or could introduce temperature bias in the radon sensitivity of the detectors. In this work three approaches are proposed dealing with the thoron influence reduction and the temperature bias introduced by packing in polymer membranes: An approach that allows to optimize the membrane properties to efficiently reduce the thoron influence while introducing small temperature bias; A differential approach that uses two passive devices packed in membranes of different diffusion properties, that allows to perform good thoron suppression and to estimate and correct for the temperature bias in  $^{222}\text{Rn}$  readings; And an approach for temperature correction applicable for monitors with anti-thoron membrane that uses the temperature data of the monitor to correct for the temperature bias.

*Keywords:* Radon, Thoron, Radon detectors, Polymer membranes, Temperature bias, Thoron influence

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## 1. Introduction

Radon ( $^{222}\text{Rn}$ , half-life  $T_{1/2}=3.8232(8)$  d (Bé et al., 2016)) is widely spread near the earth surface, as it is a part of the natural radioactive chain of  $^{238}\text{U}$ . As an inert gas with a relatively long half-life, radon could migrate far from the spot  
5 of its generation and could accumulate indoors. The indoor radon is responsible for more than 50% of the annual effective dose of the general population from natural sources (UNSCEAR, 2000) and is the leading cause for lung cancer for non-smokers (WHO, 2009). Thoron ( $^{220}\text{Rn}$ ,  $T_{1/2}=55.8(3)$  s (Bé et al., 2016)) is also a part of a natural radioactive chain ( $^{232}\text{Th}$  series) and could also be  
10 a health hazard (UNSCEAR, 2000; Tokonami et al., 2001; UNSCEAR, 2008; Chen and Moir, 2012). However, due to its much shorter half-life, it could only migrate a short distance. Therefore, thoron reaches hazardous concentrations indoors only at specific conditions (Tokonami et al., 2001; UNSCEAR, 2008; Meisenberg et al., 2017).

15 Dealing with the indoor radon is a complex task with several aspects: identifying buildings with high radon, finding/locating the sources of radon, performing risk assessment, mitigation, etc. To address these, various methods were developed, which use passive devices or monitors with passive and/or active sampling (Ward III et al., 1977; Fleischer et al., 1980; Tommasino, 1990, 1998; Nikolaev and Ilić, 1999; Papastefanou, 2002; Mark Baskaran, 2016; Galli et al.,  
20 2019). The passive devices, such as diffusion chambers with Solid-State Nuclear Track Detector (SSNTD) or electret detector or charcoal canisters, use passive detectors and passive radon sampling. The monitors use active detectors such as PIPS-detectors, Lucas cell, ionization chambers, etc. and passive or active sam-  
25 pling (e.g. passive diffusion through a diffusion barrier or active air-sampling with a pump).

Most radon detectors employ the secular equilibrium between radon and its Short-lived Decay Products (SDPs) in the detector's volume and measure radon by detecting the alpha-particles of radon ( $^{222}\text{Rn}$ ) and its SDPs –  $^{218}\text{Po}$   
30 and  $^{214}\text{Po}$ . Thoron has similar decay scheme with several short-lived alpha-

emitters –  $^{220}\text{Rn}$ ,  $^{216}\text{Po}$ ,  $^{212}\text{Bi}$  and  $^{212}\text{Po}$ . That makes all these radon detectors (passive devices and monitors) potentially sensitive to both radon and thoron (Ward III et al., 1977; Fleischer et al., 1980; Tommasino, 1990, 1998; Nikolaev and Ilić, 1999; Ggriffith and Tommasino, 1990; Michielsen and Bondiguel,  
35 2015; Dwivedi et al., 2001; Bochicchio et al., 2009). There are three commonly used approaches to discriminate between radon and thoron:

The *alpha-spectrometry* is probably the best way to discriminate between radon and thoron. For example, the semiconductor detectors allow good enough energy resolution to discriminate between the alpha-peaks of radon, thoron and  
40 their SDPs. However, this approach is applicable only to monitors.

The idea of the *"delayed-measurements"* is to delay the measurement of the sampled air for a few minutes and let thoron ( $T_{1/2}=55.8(3)$  s (Bé et al., 2016)) and  $^{216}\text{Po}$  ( $T_{1/2}=0.148(4)$  s (Bé et al., 2016)) decay. The other two alpha-emitters among the thoron's SDPs are preceded by  $^{212}\text{Pb}$ , which has a  
45 long enough half-life of 10.64(1) h (Bé et al., 2016), so that they would not contribute to the signal in a short-term radon measurement. Similar concept is used in AlphaGUARD PQ2000 PRO (Rn/Tn) to measure radon and thoron simultaneously. The sampled air is pumped for a few minutes in the pulsed ionization chamber of the detector and is measured during the pumping, then  
50 the pumping is stopped and the air is measured again. These two measurements are used to estimate both radon and thoron. For long-term measurements the thoron concentration estimated in previous intervals is used to calculate the build-up of  $^{212}\text{Pb}$  and the two alpha-emitters that follow after it and to correct for their influence. This approach is more complicated and is applicable only  
55 when active sampling is used.

The *diffusion barriers* such as polymer membranes, spongy materials or thin air-gaps are used in almost all passive devices and monitors with passive sampling. Their purpose is to keep out the SDPs present in the ambient air and to let radon enter inside the detector's volume (Ward III et al.,  
60 1977; Fleischer et al., 1980; Tommasino, 1990, 1998; Nikolaev and Ilić, 1999; Ggriffith and Tommasino, 1990; Dwivedi et al., 2001; Bochicchio et al., 2009).

As the SDPs are attached to dust particles or grouped in clusters, they are stopped by the diffusion barrier, while the inert-gas atoms could diffuse through it. Additionally, as the thoron atoms decay much faster, their diffusion length  
65 (mean path traveled by the atoms before decay) in the diffusion barrier is much shorter than the diffusion length of radon atoms. Thus, if the barrier thickness and diffusion properties are carefully chosen, it could reduce significantly the thoron penetration through it, while it remains almost transparent to radon (Ward III et al., 1977; Fleischer et al., 1980; Tommasino, 1990, 1998;  
70 Ggriffith and Tommasino, 1990; Dwivedi et al., 2001; Bochicchio et al., 2009; Hafez and Somogyi, 1986; Miles et al., 2009). However, the diffusion properties of the diffusion barriers are temperature dependent and that could lead to two types of bias: 1. A thoron influence: With the increase of the temperature, the diffusion length also increases and the barrier could become partially  
75 transparent to thoron and at high temperature could introduce non-negligible, temperature-dependent thoron contribution to the radon measurement; 2. A temperature bias of the radon response: The temperature variations also influence the radon diffusion length which could change the transparency of the barrier to radon and thus to bias the radon signal of the detector (Tommasino,  
80 1990, 1998; Ggriffith and Tommasino, 1990; Fleischer et al., 2000).

Although the air-gaps and the spongy materials are successfully used as diffusion barriers in some detectors, the polyethylene (PE) membranes are probably the best choice, because their diffusion properties allow good radon/thoron discrimination at common indoor and outdoor temperatures. Therefore, the  
85 purpose of this work is to address the thoron influence and the temperature bias introduced by the diffusion barrier, considering PE membranes.

For that purpose a previously proposed model (Mosley, 1996) is adopted, modified and further developed to describe the transport of radon and thoron from the ambient media to the volume of the detector through a PE mem-  
90 brane with known diffusion properties. The modified model is experimentally validated. Based on that model three approaches for dealing with the thoron influence and the temperature bias are proposed. Additionally, a novel device

used in the presented studies is shown to be very appropriate for estimation of the permeability of membranes.

## 95 **2. Theoretical modeling**

Radon detectors, such as passive devices and monitors with passive sampling, consist of a passive or active detector for alpha-particles (in most of the cases), placed in a well-defined volume isolated from the ambient air by a diffusion barrier. The polymer barriers, particularly the low density polyethylene (LDPE) 100 membranes, seem to be very appropriate choice for a diffusion barrier due to several advantages that they offer:

- The temperature dependence of their diffusion properties could be estimated precisely and the radon transport through them could be modeled (Mosley, 1996; Georgiev et al., 2019);
- 105 • They are hydrophobic – their diffusion properties are not influenced by the humidity and they ensure water resistance for the detectors (Tommasino, 1990, 1998; Ggriffith and Tommasino, 1990; Miles et al., 2009; Azimi-Garakami et al., 1988) (some detectors are sensitive to humidity/moisture (Hopper et al., 1999; De Simone et al., 2016));
- 110 • They are airtight. If the detector volume is not airtight, that could lead to active transport through the diffusion barrier and cause change of the thoron influence and the radon sensitivity of the detector;
- The LDPE foils are durable, flexible and they are produced in various thicknesses, so their radon/thoron diffusion properties could be precisely 115 chosen and they could be easily manipulated to fit almost everywhere;
- Because of the above mentioned mechanical properties, they could be applied to already existing detectors with other types of diffusion barriers.

Therefore, in the present work LDPE membranes are considered, however the model could be easily applied to other types of diffusion barriers.

120 *2.1. Diffusion model*

In (Mosley, 1996) a method for measuring the diffusion coefficient  $D$  of radon in thin films is presented. The experimental setup is shown in Figure 1. It consist of two chambers separated by the studied film and an alpha-detector inside the accumulation chamber (detector's volume). To study the diffusion  
 125 coefficient, a model of radon transport through the membrane and the build-up of radon in the accumulation chamber is developed. It is considered that the diffusion of radon through the thin film is described by one-dimensional diffusion equation with additional term that accounts for the radioactive decay:

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

where  $c = c(x, t)$  is the radon atomic concentration inside the film,  $\lambda$  is the  
 130 decay constant of radon,  $t$  is a time variable and  $x$  is a space coordinate in direction perpendicular to the surface of the film. It is also stated that steady state diffusion in the thin film is reached for a characteristic time (Mosley, 1996):

$$\tau_r = \frac{1}{\lambda + \frac{\pi^2 D}{d^2}} = \frac{1}{\lambda \left(1 + \left(\frac{\pi L_D}{d}\right)^2\right)}, \quad (2)$$

where  $d$  is the thickness of the film and  $L_D = (D/\lambda)^{1/2}$  is the diffusion length of radon in the film's material. If that characteristic time<sup>1</sup> is much smaller  
 135 than the life-time of radon  $\tau_r \ll 1/\lambda$  or equivalently  $d \ll \pi L_D$  (see Eq. 2), it could be assumed that the transport of radon through the film is at steady-state condition (Mosley, 1996), which leads to  $\partial c/\partial t = 0$ . On the other hand, the higher the ratio  $L_D/d$  is, the more permeable the film is and as the film should be permeable to radon, the steady-state condition should be satisfied.  
 140 In that case, Eq. 1 is solved with boundary conditions  $c(x = 0) = C_{out}$  and

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<sup>1</sup>For instance, for membranes made of LDPE ( $L_D \approx 1$  mm (Georgiev et al., 2019)) and of polycarbonate (PC) ( $L_D \approx 0.05$  mm (Georgiev et al., 2019)) with thickness  $d \approx 0.1$  mm, these times are  $\tau_{r,LDPE} \approx 8$  min and  $\tau_{r,PC} \approx 40$  h.

$c(x = d) = C_{in}$ :

$$c(x) = \frac{C_{out} \sinh((d-x)/L_D) + C_{in} \sinh(x/L_D)}{\sinh(d/L_D)}, \quad (3)$$

where  $C_{out}$  is the radon concentrations in the ambient media (the source chamber) and  $C_{in}$  is that in the accumulation (detector's) volume. At this point, the first modification to the model described in [3] is made, because this model does not take into account the partition coefficient of radon between the air and the polymer. In this work the partition coefficient  $K = C_P/C_M$  is defined as the ratio of the concentration at the two sides of the border between the polymer ( $C_P$ ) and the ambient media ( $C_M$ ) and  $K$  depends on the temperature. Another modification is introduced to account for exponentially decreasing radon concentration in the source chamber opposed to the constant radon concentration used in (Mosley, 1996): Prompt introduction of radon in the exposure system at the beginning of the experiment, followed by radon decrease during the experiment due to radioactive decay and leakage (if the source chamber is not fully hermetic to  $^{222}\text{Rn}$ ) described as  $C_{out} = C_{0,out}e^{-\lambda_{out}t}$ , where  $\lambda_{out}$  accounts for both leakage and radioactive decay. If the leakage is small ( $\tau_r \ll 1/\lambda_{out}$ ), then Eq. 3 is modified to:

$$c(x) = K \frac{C_{0,out} \sinh((d-x)/L_D)e^{-\lambda_{out}t} + C_{in} \sinh(x/L_D)}{\sinh(d/L_D)}. \quad (4)$$

Including the partition coefficient in the model is important as the partition coefficient of radon in some polymers could be much greater than 1. This would increase significantly the radon concentration inside the membrane which would enhance the radon transport through it. If  $\lambda_{out}$  is set to zero (i.e. no decrease in the radon concentration), then  $C_{out}(t) = \text{constant}$  and Eq. 4 takes the form of the corresponding equation obtained by the original model (Mosley, 1996).

In the model of Mosley (1996), the build-up rate of radon in the detector's volume is described as a build-up due to radon inflow through the membrane and radon loss due to radioactive decay:

$$\begin{aligned}\frac{dC_{in}(t)}{dt} &= J_c(t, x = d) \frac{S}{V} - \lambda C_{in}(t) \\ &= -D \frac{S}{V} \frac{\partial c(t, x)}{\partial x} \Big|_{x=d} - \lambda C_{in}(t),\end{aligned}\quad (5)$$

where  $J_c(t, x = d) = -D(\partial c/\partial x)|_{x=d}$  is the radon flux through the membrane at the detector's side. Considering  $c(x)$  given by Eq. 4, the radon flux is calculated:

$$J_c(t, x = d) = -D \frac{\partial c(t, x)}{\partial x} \Big|_{x=d} = \frac{P}{L_D} \frac{C_{0,out} e^{-\lambda_{out} t} - C_{in} \cosh(d/L_D)}{\sinh(d/L_D)}, \quad (6)$$

where the quantity  $P = KD$  is the permeability of radon through the membrane. Combining Eqs 5 and 6 leads to:

$$\frac{dC_{in}(t)}{dt} + \left( \lambda + \frac{PS}{VL_D \tanh(d/L_D)} \right) C_{in}(t) = \frac{PS}{VL_D \sinh(d/L_D)} C_{0,out} e^{-\lambda_{out} t}. \quad (7)$$

170 Considering an initial condition  $C_{in}(t = 0) = 0$ , the solution of Eq. 7 is:

$$C_{in}(t) = \frac{C_{0,out} e^{-\lambda_{out} t}}{\left(1 + \frac{\lambda}{\lambda_d}\right) \cosh(d/L_D)} \left(1 - e^{-(\lambda + \lambda_d)t}\right) \quad (8a)$$

$$C_{in}(t \gg \tau_{eq}) = \frac{C_{0,out} e^{-\lambda_{out} t}}{\left(1 + \frac{\lambda}{\lambda_d}\right) \cosh(d/L_D)} \quad (8b)$$

where  $\tau_{eq} = (\lambda + \lambda_d)^{-1}$  is the characteristic time for reaching equilibrium radon concentration in the detectors volume and  $\lambda_d$  accounts for radon transport through the membrane:

$$\lambda_d = \frac{PS}{VL_D \tanh(d/L_D)}. \quad (9)$$

It should be noted that solving the partial differential equation (7) yields to the solution (8) with a term ( $C_{0,out} e^{-\lambda_{out} t}$ ) which is the same as the inhomogeneous (right-hand side) part of (7) and actually represents the time dependence of the outside (source) concentration. It is also seen from Eq. 8b that after certain period of time  $t \gg \tau_{eq}$  the inner radon activity concentration is proportional

to the outside activity concentration  $C_{in}(t) \propto C_{out}(t)$ . Additionally, as the  
 180 membrane should be permeable to radon, then the condition  $L_D \gg d$  should be  
 fulfilled. Taking into account that  $\cosh(d/L_D) \rightarrow 1$  and  $\tanh(d/L_D) \rightarrow d/L_D$   
 when  $(d/L_D) \rightarrow 0$ , the Eqs. 8 and 9 could be simplified to:

$$C_{in}(t) = \frac{C_{out}(t)}{(1 + \frac{\lambda}{\lambda_d})} (1 - e^{-(\lambda + \lambda_d)t}) \quad (10a)$$

$$C_{in}(t \gg \tau_{eq}) = \frac{C_{out}(t)}{(1 + \frac{\lambda}{\lambda_d})} \quad (10b)$$

with:

$$\lambda_d = \frac{PS}{Vd}, \quad (11)$$

If equilibrium is reached much faster than the change in the outside activity con-  
 185 centration, then the inner concentration  $C_{in}$  will follow the outer concentration  
 $C_{out}$  and the ratio  $R = C_{in}/C_{out}$  should be constant at the given temperature  
 and should only depend on  $\lambda_d$ . Thus, if the ratio  $R$  is estimated (e.g. exper-  
 imentally) then the permeability could be determined, or, if the permeability  
 of the membrane is known, then the bias in the response of the detector could  
 190 be predicted and its readings could be corrected, which is the basic idea in the  
 current manuscript.

It is also of interest to consider the model with an initial activity concentra-  
 tion inside the detector's volume  $C_{in}(t = 0) = C_{0,in}$  and a zero concentration  
 outside  $C_{out}(t) = 0$  – e.g. a packed passive device exposed to radon and then  
 195 left to degas in radon-free air. The changes in the model that these conditions  
 yield, lead just to setting  $C_{0,out} = 0$  in Eqs 4, 6 and 7. Thus, Eq. 7 becomes  
 homogeneous and its solution is:

$$C_{in}(t) = C_{0,in} e^{-(\lambda + \lambda_d)t} \quad (12)$$

In the case of passive device exposed for time  $t_{exp}$  the activity concentration  
 $C_{0,in}$  in the device volume at the end of the exposure could be obtained, if the

200 time  $t$  in Eq. 10 is set to  $t_{exp}$ . Thus, Eq. 12 takes the form:

$$C_{in}(t_d) = \left[ \frac{C_{out}(t)}{\left(1 + \frac{\lambda}{\lambda_d}\right)} (1 - e^{-(\lambda+\lambda_d)t_{exp}}) \right] e^{-(\lambda+\lambda_d)t_d} \quad (13)$$

where  $t_d$  is the degassing time and the initial moment  $t_d = 0$  is at the end the exposure. It could be shown by Fleischer et al. (1980) that for a passive device exposed in a packing, if it is left to degas in its packing for time  $t_d \gg (\lambda+\lambda_d)^{-1}$ , its signal is proportional to the average ambient activity concentration during  
 205 the exposure:

$$n_0 = CF \frac{\overline{C}_{out} t_{exp}}{1 + \frac{\lambda}{\lambda_d}} = CF \frac{\overline{C}_{out} t_{exp}}{1 + \lambda \frac{V_d}{PS}} \quad (14)$$

where  $n_0$  is the net signal of the passive device (e.g. the net track density of a SSNTD) and  $CF$  is the calibration factor of the device.

#### *Application of the diffusion model to thoron*

Due to the relatively short half-life of thoron, all transitional processes (dif-  
 210 fusion through the membrane and build-up in the detector's volume) terminate within less than 10 minutes. This time is much shorter than the typical times of practical interest, e.g. calibration exposures, field measurements with passive detectors or dynamics follow up with monitors. Therefore, in the case of thoron Eq. 1 is considered with steady-state condition  $\partial c / \partial t = 0$ . Actually, this yields  
 215 the same solutions for the thoron distribution  $c(x)$  in the diffusion barrier (membrane) given by Eq. 4 and for the thoron flux  $J_c(t, x = d) = -D(\partial c / \partial x) |_{x=d}$  through the membrane at the detector's side given by Eq. 6. Then, Eq. 5 is also considered with steady-state condition  $dC_{in}(t)/dt = 0$  and solution for the flux given by Eq. 6 is substituted in it. This leads to a linear (not differential)  
 220 equation with solution:

$$C_{in, Tn}(t) = \frac{C_{out, Tn}(t)}{\left(1 + \frac{\lambda_{Tn}}{\lambda_{d, Tn}}\right) \cosh(d/L_{D, Tn})}, \quad (15)$$

and

$$\lambda_{d,Tn} = \frac{PS}{VL_{D,Tn} \tanh(d/L_{D,Tn})}, \quad (16)$$

where  $C_{in,Tn}(t)$  and  $C_{out,Tn}(t)$  are the thoron concentrations in the detector's volume and the ambient media,  $\lambda_{Tn}$  is the thoron decay constant and  $L_{D,Tn} = (D/\lambda_{Tn})^{1/2}$  is the diffusion length of thoron in the membrane's material. Equations 15 and 16 are very similar to those for radon (see Eqs 8b and 9): the permeability of the membrane  $P = KD$  is considered the same for radon and thoron as their atoms are chemically the same and, essentially, the differences are due to the different decay constants of radon and thoron. Following from the definition the diffusion lengths of radon and thoron are related as:  $L_D/L_{D,Tn} = (\lambda_{Tn}/\lambda)^{1/2}$ .

## 2.2. Application of the diffusion model to passive devices

In the present work two approaches based on the diffusion model are proposed that account for the thoron and the temperature influence on the radon signal of passive devices packed in a polymer foil.

The **optimization approach** allows an estimation of the effect of a given packaging and an optimization of the parameters of the packaging in order to minimize the thoron and the temperature influence. An example for such an optimization is shown in Figure 2 considering a LDPE membrane with known diffusion properties (Georgiev et al., 2019). It is also considered that the passive device is calibrated in the packaging at temperature  $T=20^\circ\text{C}$  and during the field measurements the temperature variations are in the interval  $10\text{-}30^\circ\text{C}$  (which is an overestimated interval of the typical indoor temperature variations). In Figure 2 the thoron influence is shown as the part of the ambient thoron activity concentration that penetrates the packed volume:  $R_{Tn} = C_{in,Tn}/C_{out,Tn}$ , estimated by Eq. 15. The temperature influence on the radon signal is shown as the relative difference  $R_{in} = \frac{C_{in}(T) - C_{in}(20^\circ\text{C})}{C_{in}(20^\circ\text{C})}$ , where  $C_{in}(T)$  and  $C_{in}(20^\circ\text{C})$  are estimated by Eq. 10b assuming the same ambient activity concentration

for both temperatures. Because both  $P(T)$  and  $L_D(T)$  increase with the temperature, the temperature bias is positive at higher temperatures (compared to the temperature during the calibration) and negative at lower temperatures (Fig. 2). Thus, this is an estimation of the maximum bias – if the temperature is systematically either lower or higher than the temperature during the calibration. In the case of random variation, this bias should be smaller. The thoron and the temperature influence are given as functions of  $(Vd/S)$  and once the  $V/S$  ratio is known the optimum value of  $d$  could be chosen (see Eqs 11 and 16). The range of the  $(Vd/S)$ -axis in Figure 2 is chosen so it could match real detectors' dimensions and reasonable LDPE membrane thicknesses. It is seen that if, for example,  $(Vd/S)=0.001 \text{ cm}^2$ , then the maximum thoron influence could be less than 2% and the maximum temperature influence could also be about 2%. The optimization approach would in many cases allow a reasonable reduction of the thoron influence without introducing a significant temperature bias. The temperature bias could be estimated and added to the uncertainty budget of the measurement.

**The differential method** (the second approach) allows to estimate the temperature influence and to correct the signal of the passive device. It requires two passive devices to be used in the radon measurement. These two devices, called a "device couple", should be packed in packages with different diffusion properties. The most simple way to do that is to use the same geometry ( $V/S$ ) and the same material ( $P$  and  $L_D$ ) of the packings with difference only in the membrane thicknesses. The idea of this approach is to use the ratio  $R_s = n_1/n_2$  of net signals  $n_1$  and  $n_2$  of the two devices in the couple and the temperature dependence of this ratio  $R_s(T)$  in order to estimate the temperature and then to correct the signal of the device couple.

This could be done by using identical devices for the device couple calibrated at a single temperature. As the devices are identical, they should have one and the same sensitivity (or calibration factor  $CF$ ) to radon. Thus, using Eq. 14

the signal ratio could be expressed as:

$$R_s(d_1, d_2; T) = \frac{n_1}{n_2} = \frac{1 + \frac{\lambda}{\lambda_{d,2}}}{1 + \frac{\lambda}{\lambda_{d,1}}} = \frac{1 + \lambda \frac{Vd_2}{SP(T)}}{1 + \lambda \frac{Vd_1}{SP(T)}}, \quad (17)$$

The rational function  $R_s(P)$  is monotonous and it increases with the increase of  $P$  if  $d_1 < d_2$  or it decreases with the increase of  $P$  if  $d_1 > d_2$ . It could be  
 280 solved in respect to the permeability:

$$P = \frac{\lambda V}{S} \frac{d_2 - d_1 R_s}{R_s - 1}. \quad (18)$$

This allows the device couple to be used to estimate the permeability of the membrane. On the other hand, as the temperature dependence of the permeability is already known, it could be used to estimate the exposure temperature. Further, if the devices are calibrated with no packaging (bare), the correction  
 285 coefficient  $\kappa$  to be applied to the calibration factor of a packed device is given by the ratio of the activity concentrations inside and outside the packaging:  $\kappa = C_{in}/C_{out}$  and could be estimated by the diffusion model (Eq. 10b). The permeability from Eq. 18 could be substituted in Eq. 10b to obtain a direct relation  $\kappa(R_s)$ :

$$\kappa_1 = \frac{1}{1 + \lambda \frac{Vd_1}{SP}} = \frac{d_2 - R_s d_1}{d_2 - d_1} \quad (19a)$$

$$\kappa_2 = \frac{1}{1 + \lambda \frac{Vd_2}{SP}} = \frac{d_2 - R_s d_1}{R_s(d_2 - d_1)}. \quad (19b)$$

290 Then, the experimental value of  $R_s = n_1/n_2$  could be used to estimate the correction coefficients  $\kappa_i$  (Eq. 19) and to correct the calibration factors of both devices  $CF_i = \kappa_i CF$ .

The differential method could also be applied without using the theoretical model to estimate the correction. This is a great advantage if the two devices  
 295 in the couple are different or if the permeabilities of the packings are different: It is required that the devices in the couple are calibrated (in their packings) at several different temperatures, in order to obtain the functions  $R_s(T)$  and

$CF_i(T)$ , where  $CF_i$  are the calibration factors of the packed devices. Then, in the field radon measurement, the ratio  $R_s = n_1/n_2$  could be estimated by the device couple signals and the already known function  $R_s(T)$  to be used to deduce the exposure temperature and through it the calibration factors  $CF_i = CF_i(T)$ . Moreover, the device couple could be calibrated directly versus the  $R_s$ -ratio –  $CF_i(R_s)$  and then the experimentally estimated  $R_s = n_1/n_2$  to be used to estimate directly  $CF_i = CF_i(R_s)$  without the intermediate estimation of the temperature.

The diffusion model allows some general considerations to be drawn that should be followed with both the optimization approach and the differential method:

- The ratios  $R_s$  and  $C_{in}/C_{out}$  depend on the  $S/V$ -ratio of the packaging. The higher the  $S/V$ -ratio is, the smaller the temperature bias ( $C_{in}/C_{out}$ ) and the temperature sensitivity of  $R_s$  are. Varying the  $S/V$ -ratio allows modification of the temperature sensitivity of the device couple;
- The temperature sensitivity of  $R_s$  could be increased if the packings of the two devices differ significantly in their thicknesses  $d_1$  and  $d_2$ ;
- One of the devices should be packed with a membrane with a small thickness, so that the temperature correction would be small (i.e.  $\kappa$  to be close to 1);
- If the packaging is very thick (so that  $\lambda_d$  becomes comparable to or smaller than  $\lambda$ ), it could introduce a significant decrease of the radon sensitivity of the packed detector.

### *2.3. Application of the diffusion model to a monitor with passive sampling*

The radon monitors with passive sampling already have diffusion barriers (incl. polymer membranes) that aim to prevent the diffusion of radon SDPs from the ambient media in the detector's volume. These barriers also impede the thoron diffusion in the monitor at different level. The diffusion model allows to choose suitable polymer membrane as an anti-thoron barrier in order

to reduce the thoron influence under a desired level. Nevertheless, the membrane (or any other diffusion barrier) will introduce temperature dependence of the readings of the monitor as the diffusion properties of the materials are temperature dependent. Currently, most of the radon monitors have an in-built  
330 temperature sensor. The idea of this approach is to use the temperature data from the monitor, the known temperature dependence of the permeability  $P(T)$  of the membrane and the diffusion model (Eq. 10) to perform correction of the monitor's measurement. As noted, the diffusion barriers slow down the diffusion  
335 of thoron in the monitor's volume in order to reduce its influence. Inevitably this also slows down the diffusion of radon and introduces a delay in the response of the monitor. Therefore, this approach is not suitable for devices that need fast response (e.g. devices that are supposed to follow fast changes in the radon concentration).

### 340 **3. Experimental**

The experimental setup used in this work is shown in Figure 3. It consists of two parts. The "Exposure setup" (the left part in Figure 3) used to create radon/thoron atmosphere and to perform exposure at different temperatures, is described in details by Pressyanov et al. (2017). It consists of a 50 L hermetic  
345 vessel (50.4 L Emanation Calibration Container, Saphymo GmbH, Germany), peristaltic pump with variable flow-rate, radon/thoron monitor AlphaGuard PQ2000 RnTn Pro with AlphaPump (Saphymo GmbH, Germany), a certified flow-through thoron source (Pylon Electronics Inc., Canada), a certified flow-through radon source (Czech Metrological Institute, Czech Republic), a specially  
350 designed thermostat (the 50 L vessel could fit in it) capable to sustain constant temperature or to reproduce predefined temperature variations in the temperature interval  $[-10^{\circ}\text{C}; +60^{\circ}\text{C}]$ , additional accessories such as: valves, connecting hoses, filters, smaller glass vessel with volumes from 0.2 L to 10 L.

The other part of the experimental setup is the "PIPS-system" (the right  
355 part in Figure 3). It is a modified version of the French reference thoron detector

developed at Laboratoire National Henri Becquerel (CEA/LNHB). This primary thoron detector is described in detail in (Sabot, 2015; Sabot et al., 2016). The PIPS-system is based on a PIPS-detector coupled to an electric field in order to catch the decay products of radon or thoron at the surface of the detector.

360 To allow the precise study (including the dynamics) of the transport of radon and thoron through polymer membranes in a well-defined volume, the primary thoron detector (Sabot, 2015; Sabot et al., 2016) was modified and now it possesses two chambers as shown in Figure 3: The lower chamber with circulation of air containing radon and/or thoron; and the upper chamber, which is separated by a stainless steel grid and has metalized surface in front of the silicon

365 detector, in order to perform direct measurement of the gas and the decay products. The grid between the two chambers allows any kind of thin membrane to be placed and then to perform measurement of the radon and/or thoron passing through the membrane. The PIPS-detector was connected to real-time digital pulse processor and digital multichannel analyzer nanoMCA (labZY, USA) in

370 order to acquire and analyze the the energy spectra of the alpha-particles.

In the present work two series of experiments were conducted. The first series was dedicated to the validation of the diffusion model and to the proof of concept of the differential method. For that purpose metallic diffusion chambers

375 with Kodak-Pathe LR115/II SSNTDs were used. These diffusion chambers are cylindrical metallic cans with radius  $r=3.5$  cm and height  $h=8$  cm. The cans have metallic caps and the SSNTDs are stuck on the inner surface of the caps. The caps close tight enough to ensure hermeticity of the cans for SDPs of radon and thoron. Diffusion chambers of this type were packed in LDPE foils of different thicknesses with known diffusion properties<sup>2</sup> (Georgiev et al., 2019) and

380 exposed in air with known radon concentration at three different temperatures.

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<sup>2</sup>Among the LDPE foils used in the current work, the diffusion properties of only the "100  $\mu\text{m}$ "-foil were studied in (Georgiev et al., 2019). All foils are from the same supplier's batch and the other two foils ("18  $\mu\text{m}$ " and "40  $\mu\text{m}$ ") are considered to have the same diffusion properties.

The thicknesses of the foils declared by the supplier were approximately 18  $\mu\text{m}$ , 40  $\mu\text{m}$  and 100  $\mu\text{m}$ , but the exact thicknesses of the foils were measured by a micrometer with a resolution of 1  $\mu\text{m}$ . The differences of the measured and the declared thicknesses were within 10%. In each exposure eight chambers were exposed – a couple of bare (not packed) chambers and the rest six – packed individually, each two in a foil of a given thickness. For all the experiments, the LDPE foils of each thickness were cut from a single bigger sheet (incl. the second series of experiments, described further in the manuscript). The exposures were carried out with the exposure setup shown in Figure 3 with the PIPS-system excluded. During the exposures, the pump flow-rate was constant in order to ensure constant activity concentration in the 50 L vessel with the diffusion chambers placed in it. The activity concentration and the temperature were monitored during the exposure by the AlphaGUARD. After the exposure, the chambers have been left at the same temperature in radon-free air for time sufficient for them to degas. The exposure conditions are summarized in Table 1.

Table 1: Exposure conditions in the experiments with diffusion chambers packed in LDPE foils of different thicknesses or exposed "bare". The time-integrated radon activity concentration is estimated as  $I_C = \bar{C}_{out} t_{exp}$ . The permeability of the LDPE foil is estimated from the dependence  $P(T)$  in (Georgiev et al., 2019). The measured average thickness of the foils is given with indexes "18", "40" and "100" corresponding to the thicknesses in [ $\mu\text{m}$ ] declared by the supplier. The average surface and the average volume of the packings are also given. All the uncertainties are at the level of  $1\sigma$ .

$T$ [ $^{\circ}\text{C}$ ]	$\bar{C}_{out}$ [ $\text{kBq}/\text{m}^3$ ]	$t_{exp}$ [h]	$I_C$ [ $\text{kBq h}/\text{m}^3$ ]	$P$ [ $10^{-13} \text{ m}^2/\text{s}$ ]
10(1)	19.7(15)	335.7	6600(500)	48(10)
20(1)	19.4(14)	264.0	5120(370)	108(22)
30(1)	18.4(14)	167.7	3080(240)	244(49)
$\bar{d}_{18}$ [ $\mu\text{m}$ ]	$\bar{d}_{40}$ [ $\mu\text{m}$ ]	$\bar{d}_{100}$ [ $\mu\text{m}$ ]	$\bar{S}$ [ $\text{cm}^2$ ]	$\bar{V}$ [ $\text{cm}^3$ ]
18.3(12)	41.4(17)	97.3(70)	379(23)	516(32)

The second series of experiments were aimed at the proof of concept of the applicability of the diffusion model for temperature correction of the monitors

with passive sampling. These experiments were also aimed at the additional  
400 validation of the diffusion model. In these experiments the PIPS-system was  
connected to the exposure setup as it is shown in Figure 3. The exposure  
setup was used to ensure known activity concentration in the lower chamber of  
the PIPS-system. The PIPS-system was consecutively loaded with the studied  
membranes and the PIPS-detector was used to measure the activity concentra-  
405 tion of radon/thoron in the upper chamber, which has penetrated through the  
membrane. In all experiments with radon, an exponentially decreasing activity  
concentration was used – in the beginning of the exposure the radon activity  
from the source was promptly introduced in the system, then the source was  
closed with valves and bypassed with an additional hose (not shown in Figure 3).  
410 The thoron exposures were carried out at constant thoron activity concentra-  
tion.

A calibration of the PIPS-system for radon and thoron measurements was  
conducted. In these experiments the two chambers of the PIPS-system were sep-  
arated by two mixed cellulose ester membrane filters (Millipore PHWP04700)  
415 with  $0.3 \mu\text{m}$  pore size and thickness of about  $150 \mu\text{m}$  each. These filters were  
chosen as they are transparent to both radon and thoron, efficiently stop their  
SDPs and are shown to be suitable for precise thoron penetration measurements  
(Mitev et al., 2020). For the calibration experiments pure radon or thoron at-  
mosphere was created and the activity concentration was monitored by the  
420 AlphaGUARD. The alpha-spectra from the PIPS-detector were acquired and  
processed with the nanoMCA and the labZY software.

Once the system was calibrated, LDPE foils of different thicknesses (the  
same as those used for the diffusion chambers packing) were studied at different  
temperatures. For that purpose a LDPE foil of a given thickness was loaded  
425 in the PIPS-system in addition to the two cellulose filters used in the calibra-  
tion. The temperatures of the air-conditioner and the thermostat were set to  
 $16^\circ\text{C}$  and when temperature equilibrium was reached the radon activity was  
promptly introduced in the experimental setup. The AlphaGUARD and the  
PIPS-detector followed the activity concentration in the lower and in the upper

430 chamber, respectively, until equilibrium is reached. Indication for that was the  
same rate of decrease of the two activity concentrations, see Eq. 10b. Then, the  
temperature was set to 21°C and some additional activity was promptly added  
and the same procedure was followed at that temperature. After that the same  
was repeated for 26°C. The additional input of activity between each exposure  
435 at different temperature was necessary because the radon activity concentration  
in the system was decreasing (due to decay and some leakage) while reaching  
equilibrium (up to 3-4 days). This procedure was followed for the studies of all  
foils.

Additionally, only the "18  $\mu\text{m}$ " LDPE foil was studied at 21°C with thoron  
440 as the modeling showed that less than one percent of thoron would penetrate  
in the upper chamber even with the thinnest foil available. The equilibrium  
with thoron was reached fast (in less than 10 minutes due to its short half-life),  
however the experiment continued a few days in order to get better counting  
statistic.

#### 445 **4. Results and Discussion**

After the exposure and the degassing of the diffusion chambers, their SS-  
NTDs were etched to reveal the tracks and the tracks were counted visually  
with an optical microscope. Unexposed SSNTDs were also processed for back-  
ground estimation. The track density  $n$  for each detector was estimated as the  
450 ratio  $n = N/A$  of the number of the counted tracks  $N$  and the observed area  
 $A$ . Then, the net track density  $n_0$  for each SSNTD was estimated as the dif-  
ference  $n_0 = n_s - n_b$  between the track density  $n_s$  of the exposed SSNTD and  
the background track density  $n_b$ . For all diffusion chambers the relative uncer-  
tainty of the track density was less than 5%. For each exposure the average net  
455 track density (hereafter the signal) of each couple of diffusion chambers (packed  
in foils of the same thickness or exposed bare) was estimated and is shown in  
Table 2. The ratios  $n_{0,d_i} : n_{0,bare}$  of the signals of the packed  $n_{0,d_i}$  and the  
bare  $n_{0,bare}$  chambers for each temperature were also estimated and is shown

in Table 2. The signal of the bare chambers is proportional to the activity  
460 concentration in the exposure media  $C_{out}$ , while the signal of the packed cham-  
bers is proportional to the activity concentration inside the given packaging  
 $C_{in}(d, T)$ . Thus, to validate the diffusion model the experimentally estimated  
ratios  $n_{0,d_i} : n_{0,bare}$  were compared with the model-estimated ratios  $C_{in}/C_{out}$   
(Eq. 14). The comparison is shown in Figure 4. As can be seen from Eq. 14  
465 for fixed  $S/V$  of the packing, the studied ratio is a function of the thickness  
 $d$  and the permeability  $P(T)$  of the packing. However, the temperature seems  
more intuitive and convenient parameter than the permeability and therefore in  
Figure 4 the temperature is used. In Figure 4(a) the studied ratios are presented  
as a function of the LDPE thickness for the three exposure temperatures and  
470 in Figure 4(b) the studied ratios are given as a function of the temperature for  
the three LDPE thicknesses. As can be seen there is a good agreement between  
the model and the experimental data within the uncertainties. This leads to  
the conclusion that the diffusion model describes well the behavior of diffusion  
chamber packed in anti-thoron packaging and proves the feasibility of the opti-  
475 mization approach described in Section 2.2. This also allows to speculate that  
foils of the same producer’s batch (even with different thicknesses) posses similar  
diffusion properties (permeabilities).

The results from these experiments were also used to check the applicability  
of the differential method described in Section 2.2. For that purpose the ratios  
480  $R_s$  was experimentally estimated as  $R_s(d_1, d_2; T) = n_{0,d_1} : n_{0,d_2}$ , (shown in  
Table 2). In Figure 5 these ratios are plotted versus the exposure temperature  
and compared with the model estimated dependence  $R_s(d_1, d_2; T)$  (Eq 17) using  
the already known  $P(T)$ -dependence (Georgiev et al., 2019). The results in  
the figure confirm that the packagings with bigger difference in the thicknesses  
485 ensure better temperature sensitivity of  $R_s(T)$  in agreement with what was  
noted in Section 2.2. Figure 5 shows how the exposure temperature could be  
estimated. This temperature could be used to estimate the ratio  $C_{in}/C_{out}$  for  
the given packing thickness (e.g. see Figure 4(b)) which actually accounts for  
the temperature bias of the packed passive devices. This approach was applied

Table 2: For each exposure temperature are given: The average net track density ( $n_{0,bare}$ ,  $n_{0,d_i}$ ) of diffusion chambers packed in foils of the same thickness or exposed bare; The ratios ( $n_{0,d_i} : n_{0,bare}$ ) of the net track densities of the packed and the bare chambers; The ratios ( $n_{0,d_i} : n_{0,d_j}$ ) of the net track densities of chambers with different packaging. The uncertainties are at the level of  $1\sigma$ .

		Packaging thickness			
		bare	18 $\mu\text{m}$	41 $\mu\text{m}$	97 $\mu\text{m}$
$T$ [°C]	$n_{0,bare}$ [ $\text{cm}^{-2}$ ]	$n_{0,18}$ [ $\text{cm}^{-2}$ ]	$n_{0,41}$ [ $\text{cm}^{-2}$ ]	$n_{0,97}$ [ $\text{cm}^{-2}$ ]	
10(1)	7160(260)	6430(260)	5800(240)	4380(180)	
20(1)	6360(240)	6100(240)	5480(220)	4960(250)	
30(1)	4440(190)	4400(200)	4280(180)	3870(180)	
$T$ [°C]	$n_{0,d_i} : n_{0,bare}$				
10(1)	1	0.899(48)	0.810(44)	0.612(34)	
20(1)	1	0.960(53)	0.861(48)	0.779(50)	
30(1)	1	0.992(62)	0.964(59)	0.873(57)	
$T$ [°C]	$n_{0,97} : n_{0,18}$		$n_{0,41} : n_{0,18}$		$n_{0,97} : n_{0,41}$
10(1)	0.681(39)		0.901(51)		0.756(44)
20(1)	0.812(52)		0.897(51)		0.905(59)
30(1)	0.880(58)		0.972(60)		0.905(58)

490 to the signal of the chambers shown in Table 2 and the results are given in  
Table 3. The ratios  $R_s(d_1 = 97 \mu\text{m}, d_2 = 18 \mu\text{m}; T)$  were used to estimate the  
temperature  $T_{est}$  in the three exposures. The estimated temperatures were used  
to estimate  $\kappa_{d_i}(T_{est}) = C_{in}/C_{out}$  similarly to the curves plotted in Figure 4(b).  
The corrected signal was estimated as  $n_{\kappa,d_i} = n_{0,d_i}/\kappa_{d_i}$  (Table 3). Although  
495 the temperature estimation is not very good, a very good agreement between  
the corrected signal of the packed chambers and the bare chambers is observed.  
Moreover, the uncertainty of the signal of the chambers packed in thinner foil  
is barely increased. This is due to the fact that the correction coefficient  $\kappa =$

$C_{in}/C_{out}$  and the ratio  $R_s$  are correlated (see Eq. 19) and the intermediate  
 500 poor estimation of the temperature does not affect the correct estimation of  
 $\kappa(R_s)$ . Additionally, for thin foil  $\kappa$  is close to 1 and changes slightly with  
 the temperature, and therefore the uncertainty of the temperature does not  
 contribute significantly to the uncertainty of  $\kappa$ . These observations are also  
 confirmed by Figure 6 (described in the next paragraph).

Table 3: Demonstration of the differential method (Section 2.2): The experimentally estimated  
 ratios  $n_{0,97} : n_{0,18}$  (from Table 2) are used to estimate the exposure temperature  $T_{est}$ . This  
 temperature is used to calculate  $\kappa_{d_i}$  by the model. The net track densities of the packed  
 diffusion chambers (from Table 2) are corrected  $n_{\kappa,d_i} = n_{0,d_i}/\kappa_{d_i}$  and are compared with  
 $n_{0,bare}$ . The uncertainties are at the level of  $1\sigma$ .

$n_{0,97} : n_{0,18}$	$T_{est} [^{\circ}\text{C}]$	$\kappa_{18}$	$\kappa_{41}$	$\kappa_{97}$
0.681(39)	9(3)	0.90(3)	0.79(4)	0.62(6)
0.812(52)	18(5)	0.95(2)	0.89(4)	0.77(6)
0.880(58)	25(6)	0.97(2)	0.93(4)	0.86(7)

$T_{est} [^{\circ}\text{C}]$	$n_{0,bare} [\text{cm}^{-2}]$ (to compare with)	$n_{\kappa,18} [\text{cm}^{-2}]$	$n_{\kappa,41} [\text{cm}^{-2}]$	$n_{\kappa,97} [\text{cm}^{-2}]$
9(3)	7160(260)	7150(370)	7340(480)	7070(750)
18(5)	6360(240)	6430(290)	6150(370)	6440(600)
25(6)	4440(190)	4540(230)	4600(280)	4500(430)

505 The other way to apply the diffusion method is the direct experimental  
 estimation of  $CF_i(R_s)$  and  $\kappa_i(R_s)$  during the calibration of the device cou-  
 ples. However, it is still based on the model: it is known that the dependence  
 $\kappa_i(R_s(T))$  exists and it applies to the calibration factors of the devices from the  
 couple as  $CF_i = CF\kappa_i$ . The device couple could be calibrated in their packag-  
 510 ings at a few different values of  $R_s$  (e.g. at different temperatures) and thus,  
 the  $CF_i(R_s)$  dependence could be estimated experimentally. Then in the field  
 measurements of radon, the ratio  $R_s(d_1, d_2; T) = n_{0,d_1} : n_{0,d_2}$  could be mea-  
 sured experimentally and the calibration factor could be determined directly

from  $CF_i(R_s)$ . An example for that application is given in Figure 6. For that  
 515 example the modeled dependence  $\kappa_i(R_s(T))$  for the diffusion chambers packed  
 in 18  $\mu\text{m}$  and 97  $\mu\text{m}$  foils is assumed to be the calibration-estimated dependence.  
 The experimentally obtained ratio  $R_s(97 \mu\text{m}, 18 \mu\text{m}; T) = n_{0,97} : n_{0,18}$  (shown in  
 Table 2) presented in the figure as black dashed line is used to estimate (dashed  
 arrows) the corresponding "calibration factors" ( $\kappa_i$  in this example). For the  
 520 comparison purposes the ratios  $n_{0,d_i} : n_{0,bare}$  (see Table 2) that should corre-  
 spond to the "true" values of  $\kappa_i$  are shown as dots. The very good agreement  
 observed in Figure 6 proves the applicability of the differential method for tem-  
 perature correction of the radon measurements with passive devices packed in  
 anti-thoron membranes.

525 The second series of experiments were dedicated to monitors with passive  
 sampling and for that purpose the upper chamber of the PIPS-system (see Fig-  
 ure 3) was considered to be such a monitor. The PIPS-system was calibrated for  
 radon and thoron measurement as described in Section 3. In Figure 7 examples  
 of the obtained alpha-spectra of radon and thoron with their SDPs are shown.  
 530 It is seen that in the case of pure radon or thoron atmosphere the alpha-peaks  
 of the radon SDPs –  $^{218}\text{Po}$  (6.00 MeV,  $T_{1/2}=3.071(22)$  min (Bé et al., 2016))  
 and  $^{214}\text{Po}$  (7.69 MeV,  $T_{1/2}=0.1623(12)$  ms (Bé et al., 2016)) and the thoron  
 SDP –  $^{216}\text{Po}$  (6.78 MeV,  $T_{1/2}=0.148(4)$  s (Bé et al., 2016)) are well separated,  
 which makes them suitable for measurements and therefore these peaks (nu-  
 535 clides) were used for the calibration. A drawback of  $^{214}\text{Po}$  is that it is preceded  
 in the decay chain of radon by  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$  ( $T_{1/2}=26.916(44)$  min (Bé et al.,  
 2016)) and  $^{214}\text{Bi}$  ( $T_{1/2}=19.8(1)$  min (Bé et al., 2016)) and, in order to be used  
 for measurement and calibration, at least 3 hours are needed to reach secular  
 equilibrium. However, in case of mixed radon-thoron atmosphere  $^{214}\text{Po}$  is more  
 540 suitable for radon measurement as the  $^{218}\text{Po}$  peak overlaps with the alpha-peaks  
 of thoron SDPs, see Figure 7(b).

In the calibration procedures with radon or thoron at least 10 equilibrium  
 spectra were obtained with counting statistics in Region of Interest (ROI) of  
 each calibration alpha-peak better than 1.5%. The counting rate  $\nu$  in each peak

545 was estimated as the ratio  $\nu = \Sigma/t_l$  between the total counts  $\Sigma$  in the ROI  
 of the alpha-peak (see the peak-marking in Figure 7(b)) and the live time  $t_l$   
 for the spectra acquisition. Background measurements were also carried out  
 and the background counting rate in each alpha-peak ROI was found to be  
 below 0.1% of the counting rate during the calibration. The net counting rate  
 550  $\nu_0$  was estimated as the difference  $\nu_0 = \nu_s - \nu_b$  between the counting rates in  
 the given alpha-peak ROI in the calibration spectrum  $\nu_s$  and the background  
 spectrum  $\nu_b$ . Two efficiencies for each nuclide were estimated: 1. The "volumic"  
 efficiency  $\epsilon_{v,i}$  was estimated as the ratio  $\epsilon_{v,i} = \nu_{0,i}/C_{cal}$  of the net counting rate  
 in the given alpha-peak and the activity concentration  $C_{cal}$  of radon or thoron  
 555 measured by the AlphaGUARD, and 2. The efficiency  $\epsilon_i$  was estimated as the  
 product  $\epsilon_i = \epsilon_{v,i}V$  of the "volumic" efficiency and the volume  $V=15.64(18)$  cm<sup>3</sup>  
 of the upper chamber. The volumic efficiency  $\epsilon_{v,i}$  is more convenient to work  
 with as it allows to estimate directly the activity concentration in the upper  
 chamber, while the efficiency  $\epsilon_i$  gives information about the particles detected  
 560 per one decay of radon or thoron. The obtained efficiencies were averaged over  
 all calibration spectra used and the average values are shown in Table 4.

Table 4: Efficiencies and volumic efficiencies of the PIPS-system for the radon SDPs – <sup>218</sup>Po  
 and <sup>214</sup>Po and the thoron SDP – <sup>216</sup>Po. The uncertainties of the obtained efficiencies are  
 given at the level of  $1\sigma$ . The alpha-particles energies  $E$  are also given according to Bé et al.  
 (2016).

Nuclide	$E_\alpha$ [keV]	$\epsilon_v$ [ $10^{-6}\text{m}^{-3}$ ]	$\epsilon$
<sup>218</sup> Po (radon)	6002.35(9)	6.80(37)	0.435(23)
<sup>214</sup> Po (radon)	7686.82(6)	6.71(37)	0.429(24)
<sup>216</sup> Po (thoron)	6778.4(5)	5.08(54)	0.325(35)

The next experiments were carried out with LDPE membranes of differ-  
 ent thickness separating the two chambers of the PIPS system as described in  
 Section 3. The purpose was to check the applicability of the diffusion model  
 565 for temperature correction of the readings of monitors with passive sampling

and to validate additionally the diffusion model with a different experimental setup. The activity concentrations in the lower and in the upper chambers of the PIPS system were continuously measured by the AlphaGUARD and the PIPS-detector, respectively. When an equilibrium was reached, the obtained activity concentrations were used to estimate the ratio  $C_{in}/C_{out}$  (i.e. the PIPS-system signal over the AlphaGUARD signal) and it was compared with the model expectation (shown in Figure 8). During the experiment a small activity leakage was observed in the upper detector chamber. To account for such leakage the decay constant  $\lambda$  of radon in Eq. 5 (which accounts for the decay/decrease of radon in the detector's chamber) was replaced by an effective decrease constant  $\lambda_{eff}$  which accounts for both decay and leakage. This lead to the replacement of  $\lambda$  with  $\lambda_{eff}$  in the solutions of Eq. 5. For instance, Eq. 10a takes the form:

$$C_{in}(t) = \frac{C_{out}(t)}{\left(1 + \frac{\lambda_{eff}}{\lambda_d}\right)} \left(1 - e^{-(\lambda_{eff} + \lambda_d)t}\right) \quad (20)$$

In the present experiments this constant was estimated to be  $\lambda_{eff} \approx 0.0135 \text{ h}^{-1}$ . In Figure 8(a) the effect of  $\lambda_{eff}$  is demonstrated: the dashed line represents the modeled ratio  $C_{in}/C_{out}$  if there was no leakage and  $C_{in}$  decreases only due to radon decays (i.e.  $\lambda_{eff} = \lambda_{radon} \approx 0.0076 \text{ h}^{-1}$ ) and the solid line is the modeled ratio  $C_{in}/C_{out}$  with the estimated  $\lambda_{eff} \approx 0.0135 \text{ h}^{-1}$ . The dots represent the experimental ratio for the LDPE foil of  $91.2(13) \mu\text{m}$  the permeability of which is known (the permeability of this foil is studied in (Georgiev et al., 2019)). As it is seen there is an excellent agreement between the diffusion model and the experimental results. The comparison for the three foils with the diffusion model (with the leakage taken into account) are shown in Figure 8(b). A good agreement is also observed for the  $18.4 \mu\text{m}$  foil, however the  $39.4 \mu\text{m}$  foil shows slight discrepancies. It was assumed earlier that the foils of the same batch should have the same permeability and this was supported by the diffusion chambers results. However, the results with the PIPS system indicated that such assumption could be incorrect. Therefore, it is highly recommended to characterize each membrane in terms of permeability before its usage as a diffusion barrier.

Figure 9 shows the temperature and the activity concentration measured  
595 by the AlphaGUARD and the activity concentration measured by the PIPS  
system during the study of the 91.2(13)  $\mu\text{m}$  membrane. When the temperature  
is changed, an activity is added to the system and the equilibrium is broken.  
After a while an equilibrium is reached again and an indication for that is the  
same slope of  $C_{in}$  and  $C_{out}$ , however they differ due to the diffusion resistivity  
600 introduced by the membrane. As the diffusion properties (i.e. the permeability)  
of the membrane is known, the diffusion model and the temperature data were  
used and readings of the PIPS system were corrected ( $C_{in,corr}$ ) to account for  
the diffusion resistivity of the membrane at the given temperature. The very  
good agreement between  $C_{in,corr}$  and  $C_{out}$  when equilibrium is reached shows  
605 that the proposed approach could be applied for monitors with passive sampling  
protected with anti-thoron membrane. It is also seen from the figure that if a  
membrane is used for anti-thoron protection, it introduces a time delay which  
could be significant if the diffusion parameters of the membrane are not chosen  
properly. This points out again that the diffusion properties of the membranes  
610 should be studied prior their usage as thoron barriers.

In the last experiment the same setup was used with the thoron source. The  
preliminary estimation by the model showed that if the thinnest foil (18.4(11)  $\mu\text{m}$ )  
is used and the temperature is set to 21°C the equilibrium ratio should be  
 $C_{in}/C_{out} \approx 0.4(1)\%$ . Such low penetration ratio is very sensitive to the exper-  
615 imental conditions and the diffusion properties of the membrane, which were  
actually transferred from the 91.2  $\mu\text{m}$  as already noted. However, the exper-  
imentally estimated ratio of 0.6(1)% was in good agreement with the model  
expectation, considering the low activity (about 0.03 Bq) in the upper chamber  
that was measured in order to estimate the ratio. Thus, the model seems also  
620 applicable for studies of thoron transport.

The results of this study also indicate that the PIPS system can be applied  
very well for continuous monitoring of the changes of radon and thoron concen-  
trations during calibrations and measurements. Thus, the system appears to be  
a valuable tool in the characterization of the timing response of active radon

625 monitors.

## 5. Conclusions

A verified model (Mosley, 1996) of the diffusion of radon through a membrane in a defined volume was modified and further developed to allow modeling the effect of anti-thoron membranes on the response of radon detectors to radon and thoron. The model is validated and three approaches are proposed that  
630 allow to use diffusion membranes with known diffusion properties to improve the performance of radon detectors.

The first approach allows to model and estimate the thoron reduction and the temperature bias which is introduced by a membrane with known diffusion  
635 properties if it is used as a packaging of a passive radon device. The approach allows to optimize the packaging to obtain reasonable reduction of the thoron influence while minimizing the temperature bias. It is shown that the thoron influence could be lowered to less than 2% (in terms of percentage of thoron that penetrates the packaging) with temperature bias within 2% considering  
640 calibration at 20°C with temperature values within 10-30°C.

The second approach (the differential method) uses a couple of passive devices packed in two packages with different diffusion properties. The ratio of the signal of the two devices is temperature dependent. Thus, this ratio could be used to deduce and account for the temperature bias. The applicability of  
645 the differential method is demonstrated and conclusions are drawn considering the optimal choice of the packings of the two detectors in the couple.

The third approach is applicable to radon monitors with passive sampling protected with anti-thoron membrane. The idea is to use the temperature measurements of the monitor (currently most radon monitors measure the temperature), the already known diffusion properties of the membrane and the diffusion  
650 model to calculate and apply on-line temperature correction of the measurements of the monitor. The applicability of the approach is demonstrated. A potential drawback of this approach is the time-delay of the response of the de-

tector due to the diffusion resistivity of the membrane. Therefore, the approach  
655 cannot be directly applied if a fast response of the monitor is required. It is  
speculated that a possible solution could be found if the differential signal of  
the detector (the difference between two consecutive measurements related to  
the time between them) is used in combination of the diffusion model to recover  
rapid changes. However, a dedicated study is required to study this opportunity.

660 In the course of this work, the PIPS system used in the studies was found  
to be very appropriate for study of the permeability of thin membranes. The  
system is almost the same as the French (CEA/LNHB) reference thoron detector  
and allows precise measurement of the activity penetrating the membrane, which  
is of high importance for the permeability estimation. It also allows precise  
665 studies of the diffusion dynamics including at non-constant temperature. A  
small drawback is that the PIPS system is applicable only for thin membranes  
with thickness much smaller than the diffusion length of radon in the membrane,  
which is fulfilled for the anti-thoron membranes. Additionally, the PIPS system  
is found to be a very useful tool for characterization of the timing response of  
670 active radon monitors.

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## Figure captions

**Fig. 1.** Scheme of the experimental setup used by Mosley (1996). It consist  
795 of a source chamber in which the radon concentration  $C_{out}$  is kept constant, an  
accumulation chamber with defined volume  $V$  and an alpha-detector inside (not  
shown) and a film with defined thickness  $d$  and surface  $S$  the diffusion properties  
of which are to be studied.

**Fig. 2.** Example for the thoron and the temperature influence on the  
800 radon signal of a passive device introduced by its LDPE package estimated by  
the diffusion model. The curves represent the percentage of thoron from the  
ambient media that penetrates inside the packing. The straight lines represent  
the relative temperature bias in the radon penetration inside the packaging of  
the passive device if the device is calibrated for radon measurements at 20°C.

**Fig. 3.** Scheme of the experimental setup. The right-side part is an enlarged  
805 scheme of the PIPS-system used to study the response of monitors with passive  
sampling protected by a polymer (LDPE) membrane. The left-side part is the  
Exposure setup used: 1. To create known radon/thoron activity concentration  
for the studies with the PIPS-system and 2. To expose the passive devices (in  
810 this case the PIPS-system was disconnected). Each element of the experimental  
setup has a couple of valves (not shown) that allows the element to be isolated  
without activity loss.

**Fig. 4.** Comparison of the experimental data for  $n_{0,d_i} : n_{0,bare}$  obtained  
with the diffusion chambers (dots), shown in Table 2 and the diffusion model  
815 estimated ratio  $C_{in}/C_{out}$  (solid lines), estimated by Eq. 14. In (a) these ratios  
are given as a function of the packaging thickness for the three exposure temper-  
atures and in (b) the ratios are given as a function of the exposure temperature  
for the three packaging thicknesses. The dashed lines mark the  $1\sigma$  uncertainty  
range of the corresponding solid lines, that is due to the uncertainty of the per-  
820 meability. The uncertainties of the experimental points are about 5–6% at the  
level of  $1\sigma$  for all points (not shown).

**Fig. 5.** Comparison of the experimental results (dots) and model estimated

(solid lines) dependence  $R_s(d_1, d_2; T) = n_{0,d_1} : n_{0,d_2}$  for three cases of device couple packing. The case with the biggest difference in the packaging thick-  
 825 nesses is used to demonstrate (dashed arrow) the estimation of the exposure temperature.

**Fig. 6.** Comparison of the experimental results (dots) and model estimated (solid lines) dependence  $\kappa_i(R_s)$  for the two devices of the device couple packed in LDPE membranes with  $d_1=97 \mu\text{m}$  and  $d_2=18 \mu\text{m}$ . It is also demonstrated  
 830 (dashed arrows) how the experimental value of  $R_{s,exp}$  could be used to estimate the value of  $\kappa_i(R_{s,exp})$  from the "calibration dependences". The uncertainties of the experimental points are about 5–7% at the level of  $1\sigma$  for all points (not shown).

**Fig. 7.** (a) A screenshot of a radon spectrum acquired with the labZY  
 835 software in a single measurement with the PIPS system. The alpha-peaks of  $^{218}\text{Po}$  (6.00 MeV) and  $^{214}\text{Po}$  (7.69 MeV) are marked. (b) Several radon and thoron spectra, acquired in the same way, are summed for better visualization of all alpha-peaks in radon and thoron chains. The alpha-particles of radon and thoron form continuum as their atoms are not charged and they are spread in  
 840 the volume of the chamber. On the other hand, the decay products are charged and therefore captured at the PIPS surface by the HV applied (see Figure 3) and form narrow, well resolved peaks. Due to the very small half-life of  $^{212}\text{Po}$  (300(2) ns (Bé et al., 2016)), its alpha-particles are registered in coincidence with the beta-particles of its predecessor  $^{212}\text{Bi}$  which results in a high-energy tailing of the  $^{212}\text{Po}$  alpha-peak with the shape of  $^{212}\text{Bi}$  beta-spectrum.  
 845

**Fig. 8.** Comparison between the experimentally obtained (dots) and the modeled (lines) ratio  $C_{in}/C_{out}$  as a function of the temperature for the LDPE foils with three thicknesses. During the experiments leakage was observed. In (a) the effect of the leakage is demonstrated for the 91.2  $\mu\text{m}$  membrane – the  
 850 dashed line is the model expectation without taking into account the leakage and for the solid line the leakage is taken into account. In (b) for all model-expectation curves the leakage is taken into account.

**Fig. 9.** Activity concentration and temperature follow-up during the ex-

periments with the 91.2  $\mu\text{m}$  LDPE foil. It is seen that after equilibrium the  
855 activity concentrations in the ambient media measured by the AlphaGUARD  
( $C_{out}$ ) and in the upper chamber of the PIPS system ( $C_{in}$ ) have the same slope  
but they differ. After taking into account the temperature ( $T^\circ\text{C}$ ) and using the  
model to estimate the temperature correction, the corrected activity concentra-  
tions in the upper chamber of the PIPS system ( $C_{in,corr}$ ) coincides very well  
860 with the ambient concentration measured by the AlphaGUARD.

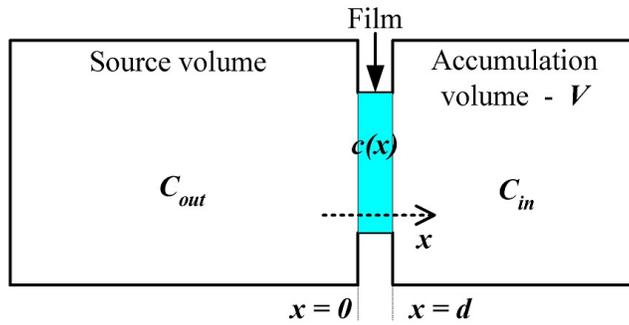


Figure 1:

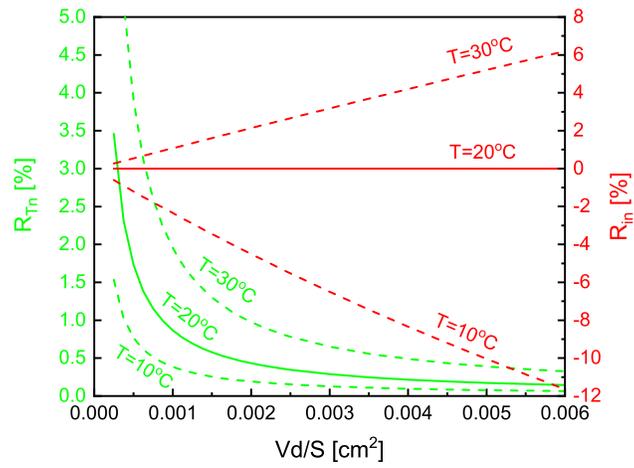


Figure 2:

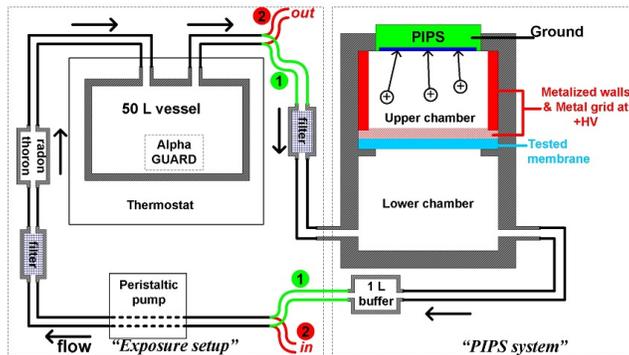
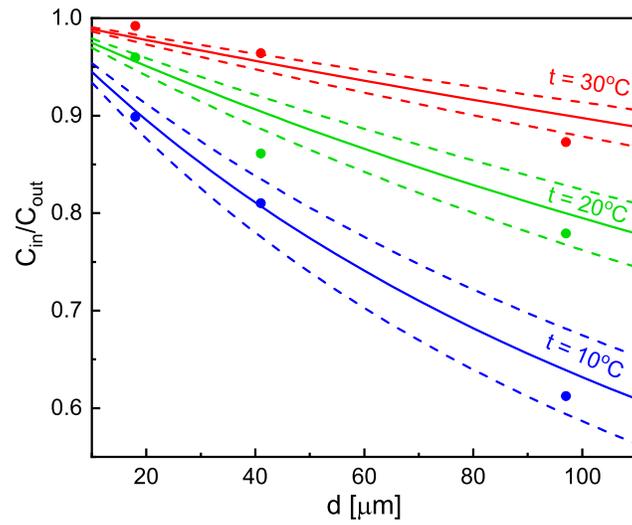
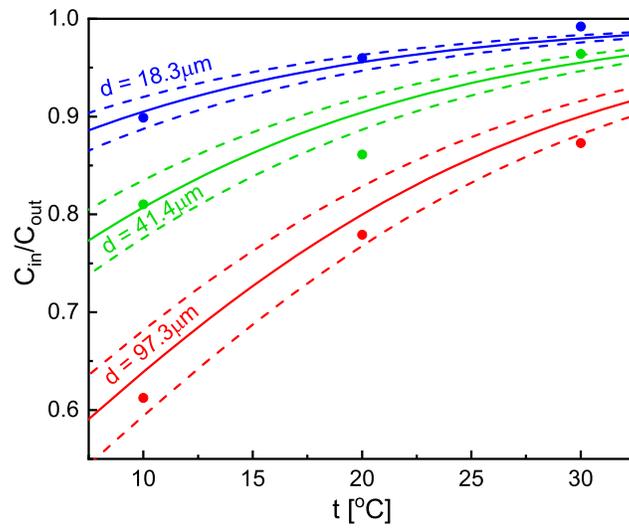


Figure 3:



(a)



(b)

Figure 4:

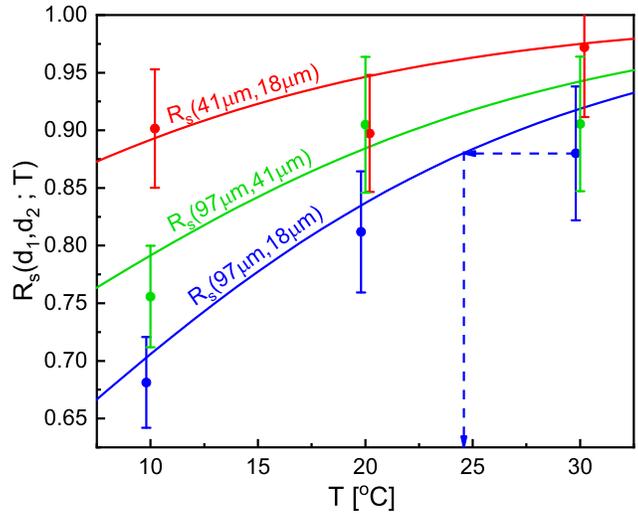


Figure 5:

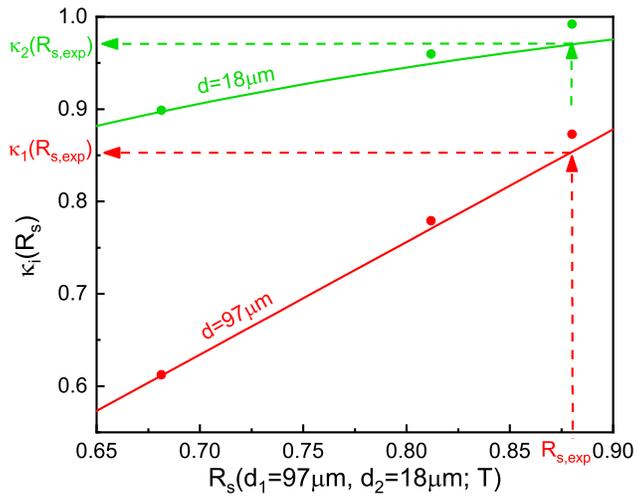
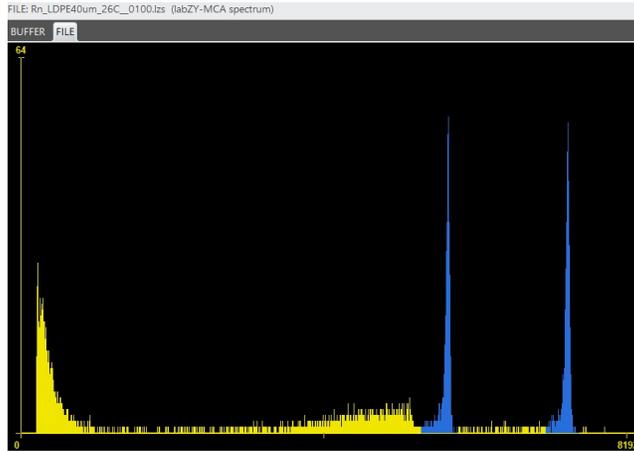
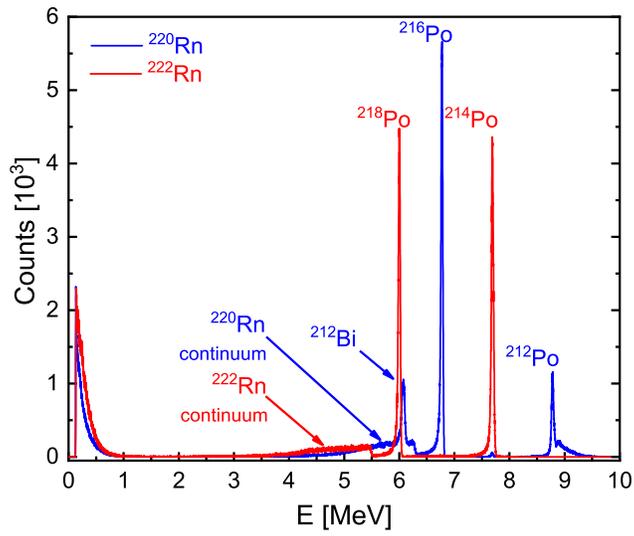


Figure 6:

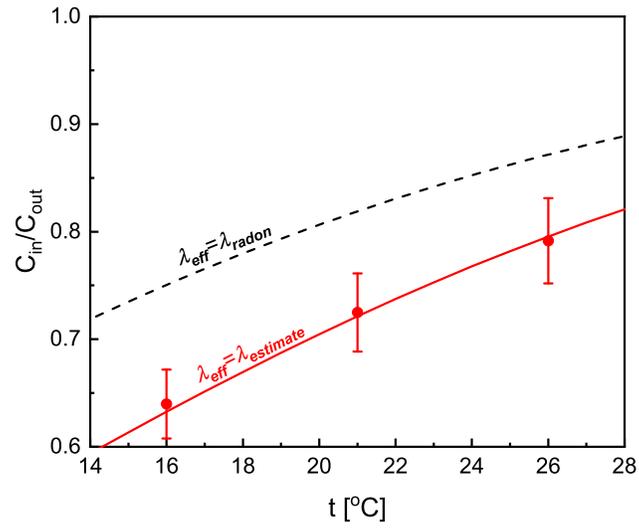


(a)

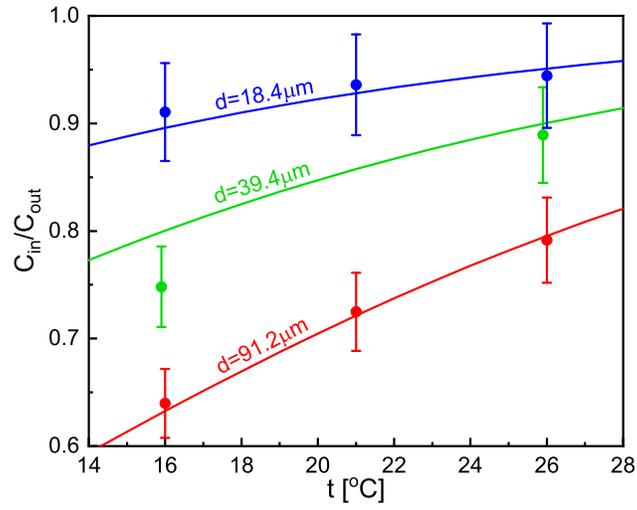


(b)

Figure 7:



(a)



(b)

Figure 8:

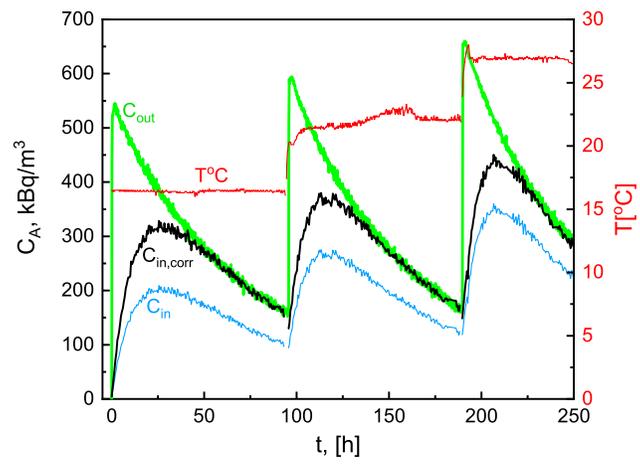


Figure 9: