

# RESEARCH PAPER Inertia in the response of radon monitors introduced by diffusion anti-thoron barriers

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### Abstract

The reduction of thoron (<sup>220</sup>Rn) influence on radon (<sup>222</sup>Rn) monitors by diffusion barriers may cause some deterioration of the quality of the radon measurements. Therefore, the best compromise has to be found between ensured anti-thoron protection and deterioration of the quality of the radon measurements. In this report, the focus is on the additional inertia in the response introduced by passive diffusion barriers against thoron. The characteristic inertia time introduced by diffusion barriers is theoretically modeled for the levels of thoron interference down to 1%. Experiments were carried out with an active monitor working in diffusion mode, using its built-in diffusion barrier and with an additional diffusion barrier added. The experimental results showed very good correspondence with the estimates based on the theoretical model. In summary, when using passive diffusion barriers against thoron, the greater is the reduction in the thoron interference, the larger is the inertia time in the response of active monitors that are introduced.

Keywords: radon; thoron; active monitors; inertia in response; anti-thoron barriers

Ithough making radon measurements not interfered by thoron has been a concern for decades, currently, many radon detectors that are used widely are still sensitive also to thoron (1). In the European Union, the implementation of the European Council Directive (2) needs dedicated research on metrology and quality assurance of radon measurements in the range of low concentrations (e.g. below 300 Bq/m<sup>3</sup>). To address these challenges, the European project MetroRADON was launched for the period 2017–2020. One of its work packages was focused on thoron interference on radon measurements and measures to reduce/eliminate it.

The concept of thoron interference may be illustrated by the following example: Consider a radon monitor which, when exposed to a certain radon activity concentration (or integrated activity concentration when the measurements are integrated), gives a signal/read-out  $E_{Rn}$ . When exposed to the same thoron (integrated) activity concentration, the signal/read-out is  $E_{Tn}$ . Then, the cross-interference of thoron on radon measurements is  $CI = E_{Tn}/E_{Rn}$ . Technical details of how this quantity is determined for active and passive radon monitors are given elsewhere (3). In the last years, different thoron exposure facilities were created, where the thoron interference can be quantitatively studied (4–7).

The techniques to reduce thoron influence may be classified as passive (placing a diffusion barrier in the pathway through which radon and thoron penetrate into the detection volume) and active (e.g., using spectrometry discrimination between radon and thoron, using 'delay lines', and counting in different time intervals) (3). There are two commonly used diffusion barriers to discriminate against thoron. In the first one, the diffusion is through thin polymer foil (8), and in the second one, the diffusion is through small gaps/holes of the chamber (9). The polymer membranes isolate the interior of the chamber from humidity, leaving radon to diffuse inside while stopping radon progeny, thoron, and thoron progeny (8). While such chambers may eliminate thoron interference, a significant temperature bias may be introduced in radon results as shown in Refs. (10–12). Other types of diffusion chambers that are preferred nowadays include diffusion through small gaps or pinholes (9). However, such chambers cannot isolate the detector volume from the environment making the response influenced by moisture/humidity, and in addition, the thoron interference is not constant and may depend on the movement and turbulence of the surrounding air (9, 13).

Overall, it appears that reducing the thoron interference may worsen the quality of the radon measurement.

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Therefore, the measures to reduce thoron influence should be considered, together with their effect on the quality of radon measurements. One quality expected from active radon monitors, especially those used for diagnostics and control of mitigation, is their fast response to changes in radon concentrations (14). In this work, results on the inertia in the radon response of active monitors introduced by diffusion barriers against thoron are reported.

### **Materials and methods**

The penetration of radon by diffusion through foils has been examined and described in the literature (8, 12, 15, 16). An analytical description of the penetration by diffusion through pinholes can be found in Ref. (9). Consider a volume V in which radon diffuses through a barrier. The growth (from zero) of radon activity concentration inside the volume  $C_{in}$ , after placing the volume at constant ambient radon concentration  $C_{out}$ , can be described by the expression (9, 17):

$$C_{in} = C_{out} \frac{1}{1 + \lambda \tau} \Big( 1 - \exp(-\lambda_{eff} t) \Big), \tag{1}$$

and when placed at  $C_{out} = 0$ , the decrease of  $C_{in}$  [from initial level of  $C_{in}(0)$ ] is:

$$C_{in}(t) = C_{in}(0) \exp(-\lambda_{eff} t), \qquad (1')$$

where  $C_{in}$  and  $C_{out}$  are the concentrations in the volume and outside,  $\lambda$  is the decay constant ( $\lambda_{Rn}$  for radon and  $\lambda_{Tn}$  for thoron, respectively), *t* is the time after the start of exposure,  $\lambda_{eff} = \lambda + \lambda_d = \lambda + \tau_d^{-1}$ , where  $\lambda_d$  is the constant for the diffusion process and  $\tau_d = \lambda_d^{-1}$  is the 'mean permeation time' (7). The expressions for these parameters are as follows (17):

1. Volume *V*, one or more sides with total area *S* of which are covered by a foil of thickness *h* made of a material of radon permeability *P* (8, 12):

$$\lambda_d = \frac{pS}{hV} \Longrightarrow \tau_d = \frac{hV}{pS},\tag{2}$$

2. Volume *V*, in which radon/thoron diffuses through holes of total area *A* and length of one hole *d*:

$$\lambda_d = \frac{AD_a}{dV} \Longrightarrow \tau_d = \frac{dV}{AD_a},\tag{3}$$

where  $D_{a}$  is the diffusion coefficient of radon in air.

At equilibrium, the transmission or attenuation factor *R* for either radon or thoron is expressed as:

$$R = \frac{C_{in}}{C_{out}} = \frac{1}{1 + \lambda \tau_d} \tag{4}$$

It should be noted that equation (2) is approximate and valid when  $h \ll L_D$ , where  $L_D$  is the diffusion length of radon/thoron in the material of the membrane

 $(L_D = \sqrt{D/\lambda})$ , where *D* is radon diffusion coefficient in the material). The general case has been considered elsewhere (12, 16). The case  $h << L_D$  is usually valid for <sup>222</sup>Rn, and foils are commonly used as protective barriers (usually polyethylene foil of thickness <100 µm). For instance, at room temperature, the diffusion length of <sup>222</sup>Rn in low-density polyethylene is about 1,500 µm, and in high-density polyethylene, it is about 700 µm (18). Due to the much smaller half-life, the diffusion length of <sup>220</sup>Rn is about two orders of magnitude smaller than that of <sup>222</sup>Rn, and the approximation might not be valid for thoron so the exact expression for *R* given in Ref. (12) should be used for this isotope.

Equation (4) gives an insight to the concept of reducing the thoron interference by diffusion barriers. Due to the great difference in decay constants of <sup>222</sup>Rn and <sup>220</sup>Rn, the transmission factor of thoron  $R_{Tn}$  is always less than the transmission factor of radon  $R_{Rn}$ :  $R_{Tn} < R_{Rn}$  and can be  $R_{Tn} << R_{Rn}$ . If a detector of inherent thoron interference *CI* is additionally protected by a diffusion barrier of transmission factor of thoron  $R_{Tn}$  and that of radon  $R_{Rn}$ , the thoron interference of such an assembled monitor will be reduced to:

$$CI' = CI \frac{R_{In}}{R_{Rn}}$$
(5)

### Results

As seen from equation (1), using diffusion barriers (gap/pinholes as well as polymer foils) always introduces an additional inertia in the response to a sudden change of radon. The reaction of the detector to a sudden change of radon concentration may be characterized by  $\tau = 1/\lambda_{eff}$  which is the 'characteristic inertia time' (usually in practice  $\lambda_d >> \lambda_{Rn}$ ) so  $\tau \approx \tau_d$ ). This is, for example, the time needed for the concentrations in the protected volume to reach 63% (i.e., *1-e*<sup>-1</sup>) of the equilibrium level, after a sudden change of the ambient concentration from zero to a constant nonzero value. We have used numerical modeling to calculate  $\tau$  at different levels of reduction of the thoron interference, quantified by  $R_{T_{r}}/R_{P_{r}}$ [see equation (5)]. Figure 1 shows the introduced characteristic inertia time. If thoron interference of less than 5% is targeted, the additional inertia time introduced by the diffusion process may exceed 30 min, and if the target interference is <1%, the additional inertia time introduced may be >2 h.

These results should be taken into account when aiming at reducing thoron interference of active radon monitors. To study the reaction of instruments to sudden change of the radon concentration, experiments were carried out by an active monitor that cannot discriminate between radon and thoron. The instrument was an old version of AlphaGUARD (Genitron GmbH, Germany) radon monitor, which cannot discriminate between radon and thoron. The AlphaGUARD monitors use alpha spectroscopy by pulse ionization chamber of active volume of 0.56 L. The active volume is protected by a glass fiber filter that is permeable for radon but stops dust and radon progeny from the exterior. The instruments employ a sophisticated algorithm for data processing based on alpha spectroscopy and pulse shape analysis (19). This algorithm discriminates the signal from radon progeny and sufficiently eliminates the inertia due to the radon progeny decay chain. The monitor operated in a diffusion mode with a 10-min measuring cycle. In this mode, radon/thoron diffuses into the detection volume through a filter that covers the entrance of the instrument, which serves as a built-in protective and diffusion barrier of the instrument. In experiments described elsewhere (3), it was found that in this mode, the thoron interference of the studied instrument is  $CI = (8.8 \pm 1.3)$  %. To reduce this barrier, an additional diffusion barrier was made of a solid plate with a 1.3 cm<sup>2</sup> square hole in the center (Fig. 2). With this additional diffusion barrier, the thoron interference was reduced about twofold to  $(4.23 \pm 0.84)$  %.

To study how this affects the reaction of the instrument to a sudden change in radon concentration, an experiment was performed in an isolated room with elevated levels of <sup>222</sup>Rn (in closed room conditions, the <sup>222</sup>Rn concentrations are usually within the range 1,200–2,200 Bq m<sup>-3</sup>). First, the instrument was started in the room and operated until the



*Fig. 1.* Characteristic 'inertia time' introduced by diffusion barriers versus the reduction of the thoron interference by protecting the detectors with diffusion barriers.

readout is stabilized at a level typical for this room. After that, the instrument was quickly (within 1 min) taken outside the building – outdoors where the radon concentrations were <10 Bq m<sup>-3</sup>. The change in the read-outs is shown in Fig. 3 (normalized to the first reading, that is, the concentration indoors). As clearly demonstrated, the decrease of readings is slower with the additional diffusion barrier placed, and the correspondence to the theoretical model (Fig. 1) is very good. This result also demonstrates that the inertia in the monitor response is mostly due to the inertia in the radon diffusion through the diffusion barrier.



*Fig. 3.* Decrease of the instrument readout without ( $\blacktriangle$ ) and with (•) additional barrier, after fast moving of the instrument from a closed room where radon concentrations are high to outdoors at radon concentration <10 Bq m<sup>-3</sup>. The read-out is normalized to the last reading in the room – just before moving the instrument (2,160 ± 180 Bq m<sup>-3</sup> for the experiment without additional barrier and 1,330 ± 100 Bq m<sup>-3</sup> for the experiment with the diffusion barrier added). The curves represent the dependence  $exp(-t/\tau)$ , where  $\tau$  is the characteristic inertia time, that correspond according to the model (Fig. 1), to the experimentally observed *CI* of 8.8% ( $\tau = 13.9$  min) and 4.23% ( $\tau = 30.4$  min) without and with the added diffusion barrier, respectively.



Fig. 2. The radon monitor used in the experiments with the additional diffusion barrier.

# Conclusions

In this report, the additional inertia introduced in radon monitors by diffusion anti-thoron barriers was studied theoretically and experimentally. The experimental results about the introduced inertia correspond very well to the theoretical model. The greater is the reduction in the thoron interference by a diffusion barrier, the larger is the inertia in the response introduced. For instance, if the cross-interference of <1% is to be reached, the characteristic inertia time may be more than 2 h. Although adding diffusion barriers to reduce thoron interference may be an easy and cost-efficient option, this is more suitable when a fast reaction is not strongly required, for example, for radon monitors used for long-term measurements. This and other related studies coherently suggest that reducing/eliminating the thoron interference on radon monitors is a matter of finding the best compromise between thoron elimination and worsening the quality of radon measurements.

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## **Conflict of interest**

The author declares no potential conflicts of interest.

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