Review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations

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MetroRadon WP 2, Report on the activity A2.3.1

Contents

Background

This work is a part of MetroRadon project (Metrology for radon monitoring) supported by the European Metrology Programme for Innovation and Research (EMPIR), JRP-Contract 16ENV10 MetroRADON (www.euramet.org). The EMPIR initiative is co-funded by the European Union’s Horizon 2020 research and innovation program and the EMPIR Participating States.

This work is a part of the Activity A2.3.1 at WP 2: “STUK and SUBG will undertake a literature review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations. Based on these findings, STUK and SUBG will perform an analytical analysis of the different techniques/materials and will identify the most promising ones, based on the effectiveness of the relative differentiation between thoron and radon.”

“In Task 2.3 the properties of different filters/foils/membranes that might potentially serve as efficient barriers for thoron, whilst not reducing radon permeability significantly, will be investigated in order to propose methods for reducing the influence of thoron on the radon measurements.”

Introduction

Radon (^{222}Rn) in indoor air is a well known problem. In most cases, the most important source of indoor radon is the soil below the building. Thoron (^{220}Rn) has a short half life of 55.6 sec and therefore the transport distance of thoron in the soil is short. Hence, the indoor thoron concentration due to transport from the soil is in most cases negligible. However, in some cases, building materials emit radon and/or thoron increasing the indoor concentrations significantly (Wiegand et al. 2000, Reddy et al. 2004, Shang et al. 2005, Gierl et al. 2014).

It is known that some radon detectors are sensitive to thoron. Number of thoron interference tests has been conducted (Tokonami et al. 2001, Ishikawa 2004, Bochicchio et al. 2009, Chen et al. 2009, Chen and Moir 2012, Sumesh et al. 2012, Michielsen and Bondiguel 2015). Thoron interference varies typically in the range 0.4 % - 74 % for alpha track detectors and in the range 4 % - 66 % for radon monitors based on ionization chamber or semiconductor detector.

In many cases, the error caused by interference is smaller than the measurement uncertainty. On the other hand, the error would be systematic and hence it would bias the measurement results increasing the measurement result.

In this document, we present a literature review of potential techniques and materials to reduce the influence of thoron on radon measurements and calibrations. More than 60 scientific articles were reviewed, but only a part was included in this document. First discriminative radon-thoron detectors are discussed shortly. After that diffusion through membranes, air gaps and pin holes as well as different membrane materials are discussed. Problems that need more detailed research within the MetroRADON project are identified.

Discriminative radon and thoron detectors

Both radon and thoron can be measured using discriminating radon-thoron detectors. Many measurement techniques have been developed. McLaughlin (2010) has divided them in to two categories. The first category includes techniques using two passive alpha track detectors, i.e., solid state nuclear track detectors (SSNTD). The two detectors have essentially identical geometry, but some differences exist. One of the detectors is designed to have low diffusion barrier, and therefore, it measures both radon and thoron gas concentrations. The other detector has higher diffusion barrier that eliminates the entry of thoron into the detector but allows the entry of radon. The difference is due to the different half-lives of radon and thoron. Based on the difference between the signals in the two detectors, both radon and thoron gas concentrations can be determined. Several authors have reported such radon-thoron detectors (Guo et al. 1995, Zhou et al. 2002, Tokonami et al. 2005, Eappen and Mayya 2004, Calamosca and Penzo 2009, Sciocchetti et al. 2010, Sahoo et al. 2013, Griel et al. 2014).

The second category includes active techniques. These can be based on the analysis of the time sequence of the alpha signals recoded in the devices such as Lucas scintillation cells (Tokonami et al. 2002, Eappen et al. 2008, Zhang et al. 2010 and Shumes et al. 2014) and ionization chambers. Time sequence differences arise due to the different half-lives of the alpha emitters in the radon and thoron chains. Some commercially available instruments utilize alpha spectroscopy to discriminate the radon and thoron-related signal, such as RAD 7 (Durridge Company Inc.). The discrimination is based on the measurement of alpha energies emitted by the radon and thoron progeny, that has been collected onto the surface barrier detector by an electric field.

A delayed coincidence technique has also been used for thoron measurements (Falk et al. 1992, Bochicchio et al. 1996). The method consists of a multiple time analysis of the pulse events detected by a flow-through scintillation cell and a phototube. It takes advantage of the relatively short time delay between the alpha particles emitted by ^{220}Rn and ^{216}Po (0.145 s half time).

A method utilizing a thin walled plastic tube has been reported by Falk et al. (2008). The technique is similar with the double-filter method based on the collection of decay products of thoron (Knutson et al. 1994 Eappen 2007: Kotrappa 1979). The setup consists of a plastic tube, an entrance filter and an exit filter. The air is sucked through the filters and tube with a constant flowrate. Inside the tube, thoron decays and its progeny is attached to the inner walls of the tube as well as on the exit filter. After the sampling period (typically 8 h), the exit filter and the inner plastic tube are folded and compressed into a standard vial for gamma counting.

Diffusion through an air gap or a pin hole

Many discriminative radon-thoron alpha track detectors are designed to possess optimal diffusion properties, Table 1. Different designs have been published. Guo et al. (1995) used an opening of 5 mm in diameter for radon detectors and four openings of 20 mm in diameter for the thoron detectors. The openings were covered with filter to allow entry only of the radon and thoron gases, but not of their progenies.

Tokonami et al. (2005) and Gierl et al. (2014) used a small gap between the lid and bottom of the detector for radon detection and several holes for radon and thoron detection. Diffusion through pin holes were used by Sahoo et al. (2013). They have calculated theoretically and verified experimentally the diffusion of radon and thoron into the detectors.

Table 1. Different designs for discriminative radon-thoron detectors.

Publication	Low diffusion rate	High diffusion rate
Guo et al. 1995	Opening of 5 mm in diameter	Four openings of 20 mm in diameter
Tokonami et al. 2005	Small air gap between the lid and bottom of the detector	Six holes of 6 mm in diameter
Sahoo et al. 2013	Four small pin holes, e.g. diameter of 2 mm	-
Gierl et al. 2014	Small air gap between the lid and bottom of the detector	Several holes, diameter not specified

Foil materials used as diffusion barriers.

Possibly, the first attempt to stop thoron in radon detectors is by the use of membrane foils as diffusion barriers (Ward et al., 1977). Several types of plastic foils (Table 2) have been studied by Hafez and Somogyi (1986). Considerable differences in radon diffusion coefficient was found due to different chemical structures. They concluded that the polyethylene proved to have the highest gas diffusion coefficient. Arafa (2002) has defined permeability constants P for 16 different materials and compared them to values found in the literature. The permeability constant is P=KD, where D is the radon diffusion coefficient in the material and K is its “partition coefficient” (this is the dimensionless solubility of radon in the material, equal to the ratio of the radon concentration in the material to that in the ambient air). Thoron separation was not reported.

Table 2. Characteristics of foils according to Hafez and Somogyi (1986).

Short name	Chemical name	Mean foil thickness (μm)	Radon permeability P_{exp} (10^{-12} m^2/s)	Radon attenuation R (%)	Thoron separation = C(Thoron) / C(Radon)
PE	polyethylene	70	7.8 ± 1.5	96.6	0.53
PC-G	polycarbonate	15	2.4 ± 0.1	89.5	0.12
HC	hydrate cellulose	25	0.97 ± 0.06	77.7	0.15
CA	cellulose acetate	25	0.75 ± 0.1	72.9	0.055
PVC1	polyvinyle chloride	10	0.58 ± 0.13	67.6	0.044
PVC2	polyvinyle chloride	10	0.61 ± 0.1	68.7	0.058
PC-KG	polycarbonate	15	0.55 ± 0.15	66.4	0.045
PET	polyethylene-terephthalate	12	0.30 ± 0.05	51.9	0.038

Table 3 summarizes the reported values of the radon permeability constant. Some variation may be observed. A review of different measurement methods has been published by Rovenska and Jiranek (2012). They concluded that differences in results can mainly be attributed to insufficient duration of the tests, insufficient radon concentration to which the samples are exposed and the use of steady state calculation procedures for data measured under non-steady state conditions. The results in Table 4 show that the differences in the values of *D*, *K* and *P* in different materials could be orders of magnitude. They can differ even when the chemical composition of the materials is the same (e.g Makrofol DE and Makrofol N polycarbonate). Moreover, the publication of Minelli and Doghieri (2017) of a study with stable gases indicate that, for a given polymer, the polymer pre-treatment and its prior history have an effect on the resulting gas solubility. The publication of Laot et. al. (2003) discusses the effects of the cooling rate and physical aging on the gas transport properties of bisphenol A polycarbonates.

Table 3. Radon permeability constants (10^{-12} m^2/s) reported by different authors.

Short name	Chemical name	Hafez and Somogyi 1986	Giridhar et al. 1982	Abdel-Fattah et al. 1987	Ramachandran et al. 1987	Wojcik 1991	Arafa 2002
	aluminized polycarbonate						0.4
	aluminized mylar						0.02
CA	cellulose acetate	0.75	0.38	0.55	0.38		2.1
CN	cellulose nitrate		12.4		12.5		1.6
HC	hydrate cellulose	0.97					3.6
PC	polycarbonate/ macrofol	0.55 - 2.4					0.03 - 0.06
PE	polyethylene	7.8		7.8	0.3		0.2 - 3.6
PET	polyethylene-terephthalate	0.30	0.08	0.3	0.08		3.0
	polyester		0.2		0.2		4.3
PVC	polyvinyle chloride	0.58 - 0.61	5	0.6	5	42	0.5

Table 4. Radon diffusion coefficient *D*, partition coefficient *K* and permeability *P* values for Nylon 6,Makrofol DE and Makrofol N polycarbonates.

Material		Partition coeffiicent	Diffusion coefficient	Permeability constant , calculated		
Short name	Chemical name	*K*	*D* (10^{-12} m^2/s)	*P=D*S*, (10^{-12} m^2/s)	Reference	Comments
N6	Nylon 6	5	0.0001	0.0005	Wojcik et. al 2000	T= 17.3 ^{O}C
K4079	Karlez comp. 4079	12.1	0.00012	0.0015	Wojcik et. al 2000	T not specified
MAK_DE	Makrofol DE	25.4	0.0072	0.18	Pressyanov et. al 2011	T= 25 ^{O}C
MAK_DE	Makrofol DE	26.2	0.0057	0.15	Mitev et. al 2016	T= 20 ^{O}C
MAK_N	Makrofol N	112	0.0032	0.36	Mitev et. al 2016	T= 20 ^{O}C

Radon detectors have been enclosed in plastic (LDPE) bags to prevent the entry of radon and thoron progeny and to reduce entry of thoron in the detector. The sensitivity of these detectors to thoron was 0.4 % of their sensitivity to radon (Bochicchio et al. 2009).

A semi-permeable membrane filter has also been used as a diffusion barrier (e.g. latex or cellulose nitrate (SN)) in radon measurement techniques. Thickness of the membrane was 25 μm and diffusion coefficient in the range of 10^{-8} - 10^{-7} cm^2/s (Eappen and Mayya 2004). It allows the build-up of about 90 % of the radon gas and suppress the thoron gas concentration by more than 99 %. The mean time for radon to reach the steady-state concentration inside the detector is about 4.5 h.

Furthermore, aluminized mylar film with thickness of 76 μm (Harley et al. 2005) and polyethylene film with thickness of 40 μm (Leung et al. 2007) have been used to separate radon and thoron. Tyvek membrane can also be used to separate radon and thoron (Kotrappa et al. 2014). A 1-mm-thick and a 4-mm-thick Tyvek membrane is reported to attenuate thoron approximately by 50 % and 95 % respectively.

Diffusion through polymer foils.

The penetration of radon through solids has been considered by many authors (e.g. Beckman, 1981; Durcik and Havlik, 1996). A useful approach to describe the radon/thoron penetration through membrane in a volume in which the detector is placed (e.g. the internal volume of a diffusion chamber with alpha track detector inside) has been described by Fleischer and Likes (1979). According to the theory described the growth of radon/thoron concentration in a volume V can be described by the expression:

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

where C_{in} and C_{out} are the concentrations in the volume and outside, λ is the decay constant, t is the time after the start of exposure, $\lambda_{eff} = \lambda + 1/\tau$ and τ is the "mean permeation time" (Tommasino 2016) which is given by the expression:

$$\tau = \frac{hV}{PS}, \qquad (2)$$

where h is the thickness of the membrane, V is the internal volume that is "protected" by the membrane, S is the area of the membrane and P is the radon permeability in the membrane material. It follows that the equilibrium "radon/thoron transmission factor (attenuation)" R is expressed as:

$$R = \frac{C_{in}}{C_{out}} = \frac{1}{1+\lambda\tau}. \qquad (3)$$

Ideally, the membrane foil that stops thoron but not radon should be chosen so that R (for radon) is close to one, while R (for thoron) is close to zero. Notably, this depends not only on the permeability and thickness of the foil, but also on its area and the volume that is "protected" by this foil. For instance at 20^0 C the NRPB monitor has R (for radon) of 0.80, while the ENEA monitor it is 0.96. However, there is another problem that we aim to address within the MetroRADON project. This is the large temperature dependence of the diffusion coefficient of many polymer materials which results to large variation of their radon permeability with the temperature, as first demonstrated by Fleischer et al. (2000). Table 5 (Tommasino, 2016) shows the temperature dependence of the permeability of polyethylene foils used in three passive radon monitors and its effect on R (for radon).

Table 5. Permeability and radon transmission factor (R) of polyethylene at different temperatures (Tommasino, 2016).

TEMPERATURE (°C)	PERMEABILITY ($x10^{-7}$cm2/s)	*R* Cup	*R* NRPB	R ENEA
0	0.15 + 0.04	0.32	0.33	0.73
20	1.20 + 0.04	0.80	0.80	0.96
40	3.60 + 0.50	0.92	0.92	0.99

As seen in Table 5 *R* (for radon) can vary by a factor of three within the temperature interval $0 \div 40^0$ C. This is a challenge, since to avoid complicated calibration adjusted to the temperature during exposure, it should be ensured that both *R* (for thoron) << 1 and *R* (for radon) ≈ 1 and their values do not vary substantially with the temperature. In order to study this problem and to be able to find practical solutions at different situations, the permeability of different polymer foils at different temperatures will be studied within the MetroRADON project.

Diffusion through pin hole or material

Diffusion through a pin hole has been calculated by Sahoo et al. (2013) and through a membrane by Arafa (2002) and Sumesh et al. (2012). If the exposure time is long enough, the transmission factor (or ratio) *R* can be approximated in both cases by the following equation:

$$R = \frac{C_{in}}{C_{out}} \cong \frac{1}{1+\frac{\lambda}{x}}, \qquad (4)$$

where C_{in} and C_{out} is the radon or thoron concentration inside and outside of the detector, respectively, λ is the decay constant and

$$x = \frac{AD}{Vd}, \qquad (5)$$

where *A* is the effective area of the pin hole or membrane, *D* is the diffusion coefficient or the permeability constant, *V* is the volume of the detector and *d* is the thickness of the membrane or length of the pin hole. Hence, radon and thoron concentrations will be different due to their different half-lives.

Sahoo et al. (2013) also calculated the transmission time T_{95} for which radon/thoron reach 95 % of its final steady state concentration in the detector:

$$T_{95} = \frac{3}{\lambda + x}. \quad (6)$$

Table 6 presents some values of the transmission factor and time. If thoron is eliminated almost completely, i.e., if diffusion is slow, the sensitivity of the detector to radon is also reduced andthe response time of the detector increases. For example, for x = 0,02 ms^{-1}, the transmission factor for radon and thoron is 90.5 % and 0.2 %, respectively, but the transmission time for radon is 38 hours.

Table 6. Transmission factors R and transmission time T_{95} for diffusion of radon and thoron through a membrane or a pin hole.

x (ms^{-1})	R(Radon)	T_{95}(Radon) (min)	R(Thoron)	T_{95} (Thoron) (min)
0.01	0.826	4128	0.001	4.0
0.02	0.905	2261	0.002	4.0
0.04	0.950	1187	0.003	4.0
0.08	0.974	609	0.006	4.0
0.16	0.987	308	0.013	4.0
0.32	0.993	155	0.025	3.9
0.64	0.997	78	0.049	3.8
1.3	0.998	39	0.093	3.6
2.6	0.999	20	0.170	3.3
5.1	1.000	10	0.291	2.8
10	1.000	4.9	0.451	2.2
20	1.000	2.4	0.622	1.5
41	1.000	1.2	0.767	0.9

Delay due to air flow in a pipe

In active detectors thoron can be eliminated using a long pipe or hose. Thoron or radon concentration $C(t)$ at the end of the pipe at time t is

$$C(t) = C_0 e^{-\lambda t}, \quad (7)$$

where C_0 is the concentration at the beginning of the pipe. Assuming a flow rate Q in a pipe with length L and diameter Φ, the delay time can be expressed as:

$$t = \frac{V}{Q} = \frac{\pi L \Phi^2}{4Q}, \quad (8)$$

where V is the inner volume of the pipe. Solving L from this equation gives:

$$L = \frac{4tQ}{\pi \Phi^2}. \quad (9)$$

Table 7 shows the relative radon and thoron concentration $C(t)/C_0$ at the end of the pipe for some values of the delay time *t*. After 8 minutes of flow in the pipe, the thoron concentration is reduced to 0.3 % of the original concentration C_0. The influence of this delay on the radon concentration is negligible. Flow time of 8 minutes can be achieved by varying the different parameters as shown in Table 8. Doubling of the length of the pipe or flow rate, doubles the delay time.

Radon concentration at the beginning of the pipe can be calculated, when the dimension of the pipe, the flow rate and the radon concentration at the end of the pipe are known.

Table 7. Relative radon and thoron concentration at the end of the pipe as a function of the delay time *t*.

t (min)	$C(t)/C_0$ Radon	$C(t)/C_0$ Thoron
1	1,000	0,473
2	1,000	0,224
4	0,999	0,050
8	0,999	0,003
16	0,998	6,34E-06

Table 8. Length of the pipe necessary to achieve delay time of 8 minutes at air-flow rate 0.5 l/min for different pipe diameters.

Diameter (mm)	Length (m)
10	50,9
20	12,7
30	5,7

Discussion and future tasks/activities

The influence of thoron on radon detectors can be significantly reduced with the discussed techniques, but a philosophical question remains: shouldn't both radon and thoron be measured, if it is known that is thoron present. If thoron concentration is small, only radon can be measured and in some cases radon detectors sensitive to thoron could be used. If radon detectors sensitive to thoron are used where thoron is present, this leads to a systematic error, which is a confounding factor in epidemiological studies. The epidemiological studies are only one area in which the application of thoron discrimination techniques is important. Other areas are radon surveys and mapping. Another important application is for the aims of radon measurements in dwellings and work places that indicate if radon remediation is needed or not. Buildings with high thoron but low radon concentrations have been observed (Pressyanov et al., 2013). If radon identification or diagnostics in such buildings is made by a detectors sensitive to thoron wrong conclusions will be drawn and resources can be wasted. However, keeping in mind the optimization principle, the possible bias due to thoron interference can be accepted in some cases. For example, a bias of ± 20 Bq/m^3 causes only minor error in the estimation of health risk and decisions about radon remediation.

The diffusion properties of radon detectors or thoron barrier membranes may be optimized to eliminate the transport of thoron. However, if the diffusion is too slow, the response time of the detector increases and the sensitivity of the detector to radon is reduced. In some cases, it is important to have a fast time response (Tommasino and Pressyanov, 2018). For example, a work place could have a mechanical supply and exhaust ventilation system, which operates only at daytime and is shut down at nights and weekends to save heating energy. This leads to a situation, where radon concentration is on higher during the nights and weekends than during the workdays. In this case, a significant error could be induced when measuring the daytime radon concentration, if the response time of the radon monitor is too slow.

Regarding calibration exposures, the influence of thoron can be eliminated using pure radon sources. If this is not possible and natural materials are used instead, the above mentioned techniques can be used to eliminate thoron from the calibration chamber. The natural sources can be placed in a plastic bag or connected to the exposure chamber through a pipe or hose that is long enough.

With respect to the usage of polymer foils as thoron barriers, the major challenge identified is the temperature dependence of the radon permeability of the polymer materials. This can change not only their properties as thoron barriers, but also the sensitivity to radon of monitors,the volume of which is “protected” by such membranes. Dedicated experimental and modeling research is planned to select materials and membrane design, so that this effect is sufficiently minimized.

In the planned tasks of the MetroRadon projects:

- The influence of thoron on active radon monitors that implement some of the reviewed techniques for thoron discrimination will be studied (A2.2.1);
- The influence of thoron on passive integrating radon detectors that implement some of the reviewed techniques for thoron reduction will be studied (Task 2.2.2);
- The properties of the discussed above filters/foils/ membranes as selective thoron barriers will be studied (Task 2.3.2 and Task 2.3.3);

Based on the results from these studies recommendations on the construction of radon detectors will be developed (Taks 2.3.4.).

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