Influence of various factors on the value of thoron emanation coefficient

Karolina Danyłec¹ · Jadwiga Mazur¹ · Krzysztof Kozak¹ · Dominik Grządziel¹ · Mariusz Mroczek¹

Received: 16 June 2021 / Accepted: 24 August 2021 / Published online: 13 October 2021
© The Author(s) 2021

Abstract

The “powder sandwich” method was adapted to the measuring equipment at the Laboratory of Radiometric Expertise, IFJ PAN (Kraków, Poland). The versatility of the method has been confirmed. The values of thoron emanation coefficients (f) for 16 materials have been determined. It has been found that the value of thoron emanation coefficient is not directly dependent on the concentration of ^{224}Ra , other factors are also important, e.g. the size of the grains, the inter-grain space and the distribution of radium atoms in the grain. The influence of the grain size on the value of thoron emanation coefficient was investigated.

Keywords Radon · Thoron · Thoron emanation coefficient · Grain size · Powder sandwich technique

Introduction

Radon is a natural radioactive gas, occurring commonly in the environment. It originates from alpha decay of radium that is present in natural uranium/thorium decay chains. There are four natural isotopes of radon: ^{222}Rn , ^{220}Rn , ^{219}Rn and ^{218}Rn . They have the same chemical properties, but they differ from radiological point of view [1]. Thoron isotope (^{220}Rn) is created during ^{224}Ra decay in ^{232}Th decay chain. It is one of the carcinogenic factors. Due to the short half-life (55.6 s) and the low concentration in the environment, its impact on human health is often neglected [2, 3]. However, there are places where there are high thoron concentrations in the ground, and therefore often also in building materials. In those places, the effective dose due to thoron inhalation is quite significant and may contribute to the increased risk of cancers of the respiratory tract [4–6]. Some authors stress the importance of thoron indoor research, due to its not negligible impact on human health [7]. It is therefore important to study the release of this gas from the ground to the atmosphere [8, 9]. This process consists of three steps; the first of them is emanation, i.e. release of thoron atom from a grain into

a pore space between grains. Emanation of thoron is a complex phenomenon, which depends on many factors, the most important of which are grain size, the distribution of ^{224}Ra atoms within a grain, humidity and porosity of material. Emanation also depends on recoil energy of thoron atom after decay of radium ^{224}Ra [10]. Sometimes this energy is not enough for thoron atom to get out of the grain or it is sufficient but thoron atom will be absorbed by the neighbouring grain or by the water located between the grains [11, 12]. For quantitative description of emanation process the so called emanation coefficient f is used. This is a ratio between a number of thoron atoms that are released out of grains into pore volume to the total number of thoron atoms that are created in the grains. The determination of the emanation coefficient for thoron is not easy. There are several articles about the processes of emanation or exhalation of radon, whereas those for thoron are not numerous. They deal more with thoron exhalation than emanation [13, 14]. The investigation of thoron emanation is rather difficult mainly due to a very short half life of thoron, i.e. 55.6 s. The most common method used for determination of thoron emanation coefficient is based on “accumulation chamber” together with alpha particle monitor [15, 16]. Another technique, based on an “open system” was presented by Wang et al. [17]. Some researchers used passive SSNTD detectors for studying the emanation process of thoron [18]. The interesting measurement technique was applied by Sakoda et al. [19]. They

✉ Jadwiga Mazur
Jadwiga.Mazur@ifj.edu.pl

¹ Institute of Nuclear Physics Polish Academy of Sciences,
Radzikowskiego 152, 31-342 Krakow, Poland

placed a sample in a closed circuit where air was pumped through the sample and thoron released from the sample was registered by a thoron monitor. The similar technique, so called “powder sandwich technique” was introduced by Kanse et al. [20].

The paper presents the adaptation of this “powder sandwich technique” to the equipment of the Laboratory of Radiometric Expertise (Institute of Nuclear Physics PAN, Kraków, Poland) and the determination of the values of thoron emanation coefficients for different materials. The universality and effectiveness of this method depending on various parameters of the measuring system has been investigated as well as the influence of grain size and moisture content of the sample on the values of thoron emanation coefficient.

Experimental

The scheme and view of measurement setup is presented in Fig. 1.

The idea of the “powder sandwich technique” is that a sample must be very thin (max 2 mm) and it must be placed in a closed circuit where airflow is forced by a pump. In that way thoron atoms created in a sample can be pushed out of the sample and measured by a thoron detector despite their short half-life. As a thoron monitor we used AlphaGUARD DF200 (Saphymo), RAD7 or EQF 3220 (SARAD) devices. Special correction factors for thoron monitors were determined, taking into account the influence of the system volume and air flow velocity on the obtained results of thoron concentration. The powdered sample was placed between two filters in a special holder, designed and constructed for that purpose. Before the measurement the sample was dried to a dry mass. All investigated samples were naturally in a powder form, so there was no need to grind them. The details of the setup as well as the way of determination of thoron emanation coefficient can be found in the previous paper [21]. The

following equation [20] was used to calculate thoron emanation coefficient f :

$$f = \text{Slope} \cdot V/Q \quad (1)$$

where *Slope*—a slope of straight line fitted to experimental points of the relationship between thoron concentration in the measuring setup and the sample mass [$\text{Bq m}^{-3} \text{g}^{-1}$], V —volume of the measuring setup [m^3], Q —concentration of ^{224}Ra in the sample [Bq g^{-1}].

Different sample masses have been used for measurements, the maximum mass did not exceed 2 g. The bigger mass caused the increase of sample thickness, which is not acceptable in this technique, as it was explained above.

The activity concentration of Ra-224 was measured using low-background gamma spectrometry with HPGe semiconductor detector. The GENIE 2000 + ISOCS software was used for spectrum analysis. The sample was prepared, sealed and stored two weeks before measurement to achieve radioactive equilibrium between Th-232 and its decay products in the radioactive chain. The activity concentration of Ra-224 was determined by activity concentration of Ac-228.

Results and discussion

Experimental part of the study consisted of carrying out measurements of thoron concentrations in the measuring system depending on the sample mass. Figure 2 presents the example of this relationship for PC1 sample. In that sample thoron concentration was measured using three different monitors. The slope of the straight line fitted to experimental points is needed to determine the value of thoron emanation coefficient f (Eq. 1). The different slopes in Fig. 2 for different thoron monitors are the result of different volumes of the detector and different flow rates.

During the study the influence of some factors to the value of thoron emanation coefficient has been investigated in order to confirm the versatility of the “powder sandwich”

Fig. 1 Scheme and view of measurement setup

Fig. 2 Relationship between thoron concentration in the measuring system and sample mass

Table 1 Thoron emanation coefficient determined for PC1 sample using different filters holding the sample

Kind of filter Pore size	f [-]
Fiberglass 2.7 μm	0.16
Fiberglass 0.7 μm	0.13
Fiberglass 0.3 μm	0.13
Membrane filter 0.7 μm	0.12
PCTE membrane filter 10 μm	0.16

Table 2 Thoron emanation coefficient determined for three samples using different lengths of connecting pipes

Sample code	Total length of connecting pipes: 3.26 m	Total length of connecting pipes: 0.5 m
	f [-]	
PC1	0.0896	0.0875
KP1	0.112	0.113
AP1	0.017	0.021

method. The following factors (that can possibly influence the result) have been checked:

- Type of thoron monitor
- Type of filters used in the sample holder
- Length of connection pipes
- Time and cycle of measurement (in case of EQF 3220 monitor)
- Position of the sample in the measuring system (vertical or horizontal)

The obtained results showed that the value of thoron emanation coefficient is not dependent on various thoron

Table 3 Thoron emanation coefficient determined for three samples using different positions of sample holder

Sample code	Sample holder: horizontal	Sample holder: vertical
	f [-]	
GI1	0.032	0.030
GJ1	0.17	0.19
PC1	0.12	0.12

Table 4 Tested materials and their country of origin

Sample code	Kind of material	Country of origin
AP1	thorium salt	Poland
GI1	soil	Indonesia
GJ1	soil	Japan
KP1	thorium salt	Poland
PB1	sand	Bangladesh
PC1	monazite sand	China
PJ1	monazite sand	Japan
PJ2	monazite sand	Japan
PM1	sand	Madagascar
PM2	sand	Madagascar
PM3	sand	Madagascar
PM4	sand	Madagascar
PM5	sand	Madagascar
PM6	sand	Madagascar
TW1	volcanic tuff	Italy
TW2	volcanic tuff	Italy

monitors, different work cycles, types of filters, lengths of connection pipes and sample orientation in the measuring system. In Tables 1, 2 and 3 the results of using different filters holding the sample, different lengths of connecting pipes and different position of the sample holder in the setup are presented. The uncertainties of emanation coefficient is ca. 10%. It can be seen that the maximum difference in emanation coefficient values is 0.04. The statistical significance test for correlation coefficient (at the 0.05 significance level) made for filters showed that the correlation between the filter pore size and the value of f is not significant. In the further study the fiberglass filter 0.7 μm and the shorter connecting pipes were used.

Having confirmed the versatility of the method, the thoron emanation coefficient has been determined for 16 samples. These were mainly sand (including monazite sand) and soil. Two samples of volcanic tuffs from Italy and two samples of thorium salts have also been studied. All samples with their country of origin are gathered in Table 4.

The values of thoron emanation coefficient f for all tested materials determined using different thoron monitors are

Table 5 Values of thoron emanation coefficient (f) and ^{224}Ra concentration (Q) for tested materials

Sample code	^{224}Ra concentration Q [Bq/g]	AlphaGUARD DF 2000 f [-]	SARAD EQF 3220 f [-]	RAD7 f [-]
PC1	63.01 ± 1.57	$0.13 \pm 5 \cdot 10^{-3}$	$0.09 \pm 6 \cdot 10^{-3}$	$0.11 \pm 5 \cdot 10^{-3}$
GJ1	$0.18 \pm 3 \cdot 10^{-3}$	0.15 ± 0.04	$0.18 \pm 1 \cdot 10^{-3}$	–
PJ2	5.8 ± 0.2	$5 \cdot 10^{-3} \pm 7 \cdot 10^{-4}$	$5 \cdot 10^{-3} \pm 3 \cdot 10^{-4}$	–
PJ1	5 ± 0.13	$3 \cdot 10^{-3} \pm 3 \cdot 10^{-3}$	$5 \cdot 10^{-3} \pm 5 \cdot 10^{-4}$	–
GI1	5.9 ± 0.02	$0.04 \pm 13 \cdot 10^{-4}$	$0.03 \pm 7 \cdot 10^{-4}$	–
PB1	50.03 ± 0.4	$9 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	$4 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	–
TW1	0.3 ± 0.01	0.12 ± 0.03	0.1 ± 0.02	0.09 ± 0.03
TW2	$0.2 \pm 6 \cdot 10^{-3}$	0.18 ± 0.06	0.18 ± 0.03	0.13 ± 0.07
AP1	1201 ± 26	$0.036 \pm 3 \cdot 10^{-3}$	$0.02 \pm 7 \cdot 10^{-3}$	–
KP1	14.62 ± 0.05	0.09 ± 0.03	0.11 ± 0.01	–
PM1	11.0 ± 0.2	$10 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	$11 \cdot 10^{-4} \pm 4 \cdot 10^{-5}$	–
PM2	12.0 ± 0.2	$12 \cdot 10^{-4} \pm 2 \cdot 10^{-4}$	$11 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	–
PM3	12.6 ± 0.2	$11 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	$9 \cdot 10^{-4} \pm 4 \cdot 10^{-5}$	–
PM4	12.4 ± 0.2	$12 \cdot 10^{-4} \pm 3 \cdot 10^{-4}$	$12 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	–
PM5	15.7 ± 0.2	$14 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	$11 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	–
PM6	24.4 ± 0.4	$10 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	$7 \cdot 10^{-4} \pm 1 \cdot 10^{-4}$	–

Fig. 3 Dependence of the thoron emanation coefficient on the concentration of radium ^{224}Ra activity in the sample

presented in Table 5. In the table there are also radium ^{224}Ra concentration in the samples.

Thoron emanation coefficient for samples PM1–PM6 are small in comparison with samples e.g. TW1, TW2, GJ1, although these samples have lower ^{224}Ra content. It shows that high ^{224}Ra concentration in a sample does not always mean also high value of emanation coefficient. This can also be seen in Fig. 3 where the relationship between thoron emanation coefficient and radium ^{224}Ra concentrations in the samples is presented. There is no direct correlation. Other factors are also important, e.g. the size of the grains, the inter-grain space and the distribution of radium atoms in the grain.

Samples PJ1, PJ2, GI1 have similar levels of ^{224}Ra concentrations. Samples PJ1 and PJ2 show also similar f values, while sample GI1 has much higher value of f . It may be related to the differences in grain size of these materials.

The thoron emanation coefficients for PB1 material, calculated on the basis of the results obtained from the indications of the AlphaGUARD DF2000 and EQF 3220 m, differ by more than 100%. This is probably due to the very low thoron emanation in this material. The values of thoron emanation coefficient are the lowest, despite the fact that the ^{224}Ra concentration in this material is 10 times higher than for materials from Japan (PJ2, GJ1 and PJ1) or Indonesia (GI1) for which the emanation coefficients are much higher. This may be due to the differences in the distribution of radium atoms in the grains or the grain structure.

The size of material's grains has an influence on the f value. During this study this relationship has been investigated for PC1 sample. Using a sieve, the material of PC1 sample was separated into four grain fractions (I–IV) with diameters:

- I: greater than 0.5 mm,
- II: 0.5–0.355 mm,
- III: 0.355–0.18 mm
- IV: less than 0.18 mm.

For each grain fraction, the concentration of ^{224}Ra activity in the sample and the value of f were determined. In Fig. 4 the dependence of thoron emanation coefficient f on grain size of PC1 sample is shown.

It can be seen that there is an optimum grain size for which emanation coefficient has the maximum value. The similar

Fig. 4 Dependence of thoron emanation coefficient f on grain size of PC1 sample

Table 6 The values of thoron emanation coefficient for dried and not dried PC1 sample determined using AlphaGUARD and EQF 3220 thoron monitors

	Alphaguard DF 2000 f [-]	Sarad EQF 3220 f [-]
Not dried PC1 sample	$0.13 \pm 5 \cdot 10^{-3}$	$0.09 \pm 6 \cdot 10^{-3}$
Dried PC1 sample	0.14 ± 0.01	0.13 ± 0.01

result was obtained previously by Sakoda et al. [22] and Markkanen and Arvela [23].

The attempts have also been undertaken to investigate the effect of sample moisture on its f value. However, the “powder sandwich” method is not good for that purpose. It is due to the fact that the sample is very thin and air flow through it makes it almost dry after some minutes. It was only possible to study in a general way the relationship between f and relative humidity of the sample PC1. The results are presented in Table 6. In the case of Alphaguard DF 2000 monitor, the difference in the f values for the undried and dried sample is 0.01 and it is within the measurement uncertainty. For Sarad EQF 3220 monitor, however, this difference is greater and amounts to 0.06. This may be rather the effect of air humidity in the closed measuring setup than of humidity of the sample which was small (ca. 6%). The manufacturer of EQF 3220 device does not recommend the use of dehumidifiers or any correction of the obtained thoron concentrations in terms of the influence of humidity. As the presented results show, it is important to ensure that the air in the measuring system is dry in order to obtain the reliable values of thoron concentrations.

Conclusions

The results of the presented research confirm that emanation process in the medium is not simple to observe and measure.

The “powder sandwich” technique was adapted to the LER IFJ equipment by making a special holder for low mass samples, choosing optimal length of connection pipes (minimizing the volume of the measuring system) and by elimination of redundant elements, such as dryer or sample container.

The versatility of the method has been confirmed, the results are not dependent on various thoron monitors, different work cycles, types of filters, lengths of connection pipes and sample orientation in the measuring system.

Correction factors for thoron monitors were determined, taking into account the influence of the system volume and air flow velocity on the obtained results of thoron concentration.

The values of thoron emanation coefficients were determined for 16 samples. The lowest emanation coefficient ($f = 9 \cdot 10^{-4}$) was observed for a sand sample from Bangladesh, and the highest ($f = 0.18$) for volcanic tuff from Italy.

It has been found that the value of thoron emanation coefficient is not directly dependent on the concentration of radium (^{224}Ra)—other factors are also important, e.g. the size of the grains, the inter-grain space and the distribution of radium atoms in the grain.

The influence of the grain size on the value of thoron emanation coefficient was investigated.

It was found that, despite the lack of recommendations from the manufacturer of the EQF 3220 m, it was necessary to reduce the humidity in the measuring system before the measurement.

Due to the very small mass of samples, it was not possible to determine the dependence of the thoron emanation coefficient on the sample moisture content. The continuation of this research will be the improvement of the measuring system in order to enable the study of the influence of sample humidity on the thoron emanation coefficient.

The results obtained in the study may contribute to the improvement of the modelling of thoron emanation process by taking into account the parameter describing the effect of the volume and shape of intergranular spaces.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- Haynes WM (2011) 92nd ed. CRC Handbook of Chemistry and Physics, vol.122. CRC Press, Boca Raton. FL., p 4, ISBN 1439855110
- UNSCEAR (2000) United Nations Scientific Committee on the Effects of Atomic Radiation Sources. In: Effects and Risks of Ionizing Radiation. Report to the General Assembly. United Nations, New York
- WHO (2009) WHO handbook on indoor radon: a public health perspective. WHO Press, Geneva
- Mayya YS, Mishra R, Prajith R, Gole AC, Sapra BK, Chougaonkar MP, Nair RR, Ramola RC, Karunakara N, Koya PK (2012) Deposition-based passive monitors for assigning radon, thoron inhalation doses for epidemiological studies. *Radiat Prot Dosim* 152(1–3):18–24
- Ramola RC, Gusain GS, Rautela BS, Sagar DV, Prasad G, Shahoo SK, Ishikawa T, Omori Y, Janik M, Sorimachi A, Tokonami S (2012) Levels of thoron and progeny in high background radiation area of southeastern coast of Odisha, India. *Radiat Prot Dosim* 152(1–3):62–65
- Hosoda M, Kudo H, Iwaoka K, Yamada R, Suzuki T, Tamakuma Y, Tokonami S (2017) Characteristic of thoron (^{220}Rn) in environment. *Appl Radiat Isot* 120:7–10. <https://doi.org/10.1016/j.apradiso.2016.11.014>
- Ramola RC, Prasad M (2020) Significance of thoron measurements in indoor environment. *J Environ Radioact* 225:106453
- Singh P, Singh P, Singh Bajwa B, Sahoo BK (2017) Radionuclide contents and their correlation with radon-thoron exhalation in soil samples from mineralized zone of Himachal Pradesh, India. *J Radioanal Nucl Chem* 311:253–261
- Singh B, Kant K, Garg M, Sahoo BK (2020) Quantification of radon/thoron exhalation rates of soil samples collected from district Faridabad of Southern Haryana, India. *J Radioanal Nucl Chem* 326:831–843
- Fleischer RL (1983) Theory of alpha recoil effects on radon release and isotopic disequilibrium. *Geochim Cosmochim Acta* 47:779–784
- Sahoo BK (2013) Modeling and measurement of radon and thoron emission from naturally occurring radioactive materials (PhD. Thesis). <https://doi.org/10.13140/RG.2.2.33980.54400>
- Greeman DJ, Rose AW (1996) Factors controlling the emanation of radon and thoron in soils of the eastern U.S.A. *Chem Geol* 129:1–14
- Rout RP, Sahoo BK, Pal R, Dhabekar BS, Bakshi AK, Datta D (2020) Investigation of ^{220}Rn emanation and exhalation from soil samples of Larsemann Hills region, Antarctica. *J Environ Radioact* 214–215:106175. <https://doi.org/10.1016/j.jenvrad.2020.106175>
- De With G, Kovacs T, Csordas A, Tschiersch J, Yang J, Sadler SW, Meisenberg O (2021) Intercomparison on the measurement of the thoron exhalation rate from building materials. *J Environ Radioact* 228:106510. <https://doi.org/10.1016/j.jenvrad.2020.106510>
- Jónás J, Sas Z, Vaupotic J, Kocsis E, Somlai J, Kovács T (2016) Thoron emanation and exhalation of Slovenian soils determined by a PIC detector-equipped radon monitor. *Nukleonika* 61(3):379–384
- Tuccimei P, Moroni M, Norcia D (2006) Simultaneous determination of ^{222}Rn and ^{220}Rn exhalation rates from building materials used in Central Italy with accumulation chambers and a continuous solid state alpha detector: influence of particle size. Humidity and precursors concentration. *Appl Radiat Isot* 64:254–263
- Wang Y, Zhang L, Guo Q (2017) The experimental study on the emanation power of a flow-through thoron source made from incandescent gas mantles. *J Radiol Prot* 37(4):918–926
- Misdaq MA, Amghar A (2005) Radon and thoron emanation from various marble materials: impact on the workers. *Radiat Meas* 39:421–430
- Sakoda A, Meisenberg O, Tschiersch J (2016) An approach to discriminatively determine thoron and radon emanation rates for a granular material with a scintillation cell. *Radiat Meas* 89:8–13
- Kanse SD, Sahoo BK, Sapra BK, Gaware JJ, Mayya YS (2013) Powder sandwich technique: a novel method for determining the thoron emanation potential of powders bearing high ^{224}Ra content. *Radiat Meas* 48:82–87
- Danyłec K, Mazur J, Kozak K, Grządziel D (2018) Determination of the thoron emanation coefficient using a powder sandwich technique. *J Environ Radioact* 195:109–113
- Sakoda A, Ishimori Y, Hanamoto K, Kataoka T, Kawabe A, Yamaoka K (2010) Experimental and modeling studies of grain size and moisture content effects on radon emanation. *Radiat Meas* 45:204–210
- Markkanen M, Arvela H (1992) Radon emanation from soils. *Radiat Prot Dosim* 45:269–272

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.