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The experimental study on the emanation power of a flow-through thoron source made from incandescent gas mantles

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Abstract

To improve the quality of the calibration of thoron concentration activity measurement, an experimental study on the emanation power of a flow-through thoron source based on incandescent gas mantles was carried out in this paper. The thoron activity concentrations of the outflowed air from the flow-through source were measured using RAD7, and the quantitative relationship between thoron concentrations and flowrates was studied through theoretical analysis, and the thoron emanation powers were obtained from the fitting of the relationship above. Results show that the thoron concentration decreased with the increasing flowrate in the gas path, and the thoron emanation powers of three batches of gas mantles obtained by fitting were 1.33% ± 0.17%, 0.77% ± 0.10% and 0.57% ± 0.07% respectively in low humidity condition. Those results were checked using the gamma spectroscopy method, and were consistent within the error range.

Keywords: thoron, emanation power, flow-through source, incandescent gas mantle, exhalation fraction

(Some figures may appear in colour only in the online journal)

1. Introduction

Thoron (^{220}Rn) is a natural isotope of radon (^{222}Rn) with a half-life of 55.6 s. The calibration of thoron activity concentration is of great importance to the accurate measurement of thoron concentration and the estimation of thoron exposure dose. However, the determination of thoron activity concentration in the air is difficult due to thoron's short half-life and relatively low concentration in the environment, and thoron has no widely accepted gas activity standard like radon [1–4]. Therefore the traceable calibration of thoron measurement devices is still a difficult point. Although there are some ongoing work about thoron and its progeny reference chamber [5], it cannot be easily achieved actually. In order to improve the quality of thoron's measurement and calibration, some flow-through thoron sources have been developed [6–8]. A flow-through source contains a radon/thoron exhalation unit and necessary gas path. It usually has a relatively small volume, making it more portable and more suitable for calibration or measurement *in situ*.

During our outdoor radon measurement process, we found that the ambiance thoron concentration cannot be ignored, but we did not have a ready-made field calibration method for thoron measurement. To solve the problem, we intended to use incandescent gas mantles as a thoron calibration source to fix it. An incandescent gas mantle is a kind of material for generating incandescent light when it is heated, which is made of artificial silk net immersed in thorium nitrate solution [6, 9]. It is commercially available and commonly used in some portable camping lanterns to improve the luminous brightness due to its simple production process and low production cost. Gas mantles can also be used as a kind of thoron source because of its considerably high ^{232}Th content. In case of this article, the thoron exhalation unit of our flow-through source is an exhalation pipe filled with gas mantles. The thoron emanation power is the ratio of the number of thoron atoms released to the pore spaces, to the number of all thoron atoms generated by decay, which is the key parameter of the source and needs to be determined. However, there are few reports on the measurement of thoron emanation power [8, 10–12], especially for gas mantles.

In this paper, we set up a flow-through thoron source measurement system based on the above-mentioned exhalation pipe, and measured the thoron emanation power of three batches of gas mantles, and then verified our results by gamma spectroscopy method.

2. Materials and methods

2.1. Theory

For a flow-through thoron source, when the carrier gas enters into the exhalation pipe filled with gas mantles (as shown in figure 1), the thoron atoms precipitated by gas mantles will be carried by the carrier gas and discharged from the outlet of the pipe. Under the condition of stable temperature and humidity, the thoron precipitation capacity of gas mantles can be considered as a constant. Therefore, the thoron activity concentration of the outflowed air will decrease with increasing flowrate theoretically.

Assume that ^{232}Th activity in incandescent gas mantles is A_0 (Bq), the thoron emanation power of gas mantles is η $(0 < \eta < 1)$, which remains stable in same temperature and humidity condition, the decay constant of thoron is λ (s^{-1}), the volume of the exhalation pipe is V (ml), and the flowrate in the pipe is q (l min^{-1}), then despite the heterogeneity of this system, the number of free thoron atoms N in the pipe can be calculated by equation (1)

$$\frac{\mathrm{d}N(t)}{\mathrm{d}t} = \eta A_0 - \lambda N(t) - \frac{q}{V} \cdot N(t), \tag{1}$$

where, the first item on the right side of the equal sign is newly generated part by gas mantles, the second and third items are thoron decayed and removed by the airflow respectively. Then we get equation (2)

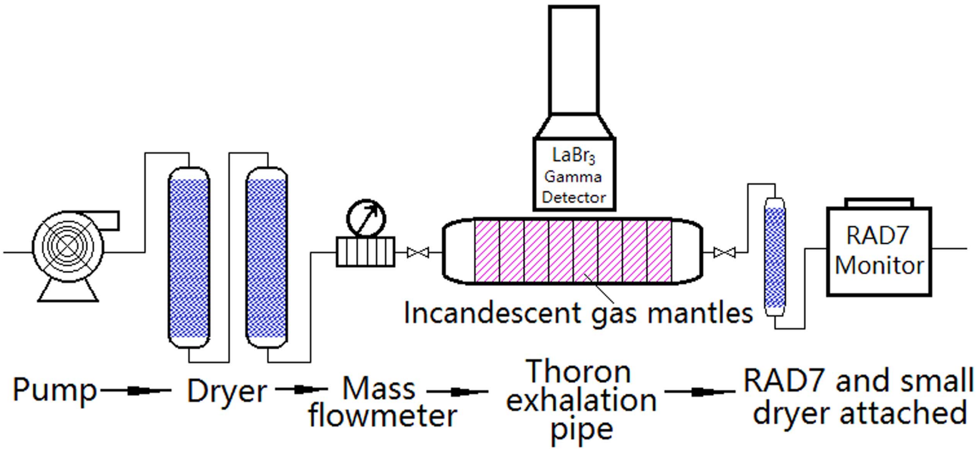

Figure 1. Schematic diagram of the thoron exhalation and measurement system.

$$N(t) = \frac{\eta A_0}{\lambda + q/V} \cdot (1 - \mathrm{e}^{-(\lambda + q/V)t}) \tag{2}$$

which shows that the number of free thoron atoms N or the thoron activity A (Bq) in the exhalation pipe will be at equilibrium after a sufficiently long period of time and can be given in equations (3) and (4)

$$N = \frac{\eta A_0}{\lambda + q/V}, \tag{3}$$

$$A = \lambda N = \frac{\lambda \eta A_0}{\lambda + q/V}. \tag{4}$$

Therefore, the thoron activity concentration C (Bq m^{-3}) in the exhalation pipe or at the outlet of the pipe can be given in equation (5)

$$C = \frac{A}{V} = \frac{\lambda \eta A_0}{\lambda V + q}. \tag{5}$$

Equation (5) shows that the thoron concentration C in the pipe is a function of flowrate q, which means if we measure a series of thoron concentrations at different flowrates, we can use equation (5) to get thoron emanation power η with the known activity A_0 and volume V by fitting. This method of measuring the emanation power of a flow-through thoron source is the main idea of this paper.

On the other hand, using the mixing model [13], we can get the relation between thoron emanation power η and thoron exhalation fraction χ ($0 < \chi < 1$), which is given in equation (6)

$$\chi = \frac{1}{1 + \dfrac{\lambda \cdot V}{q}} \cdot \eta = \frac{q}{q + \lambda \cdot V} \cdot \eta. \tag{6}$$

The thoron exhalation fraction χ is a ratio of outflowed thoron atoms number to totally generated thoron atoms number. If we can get the thoron exhalation fraction with another method, such as gamma spectroscopy, then we can get the thoron emanation power by equation (6), which gives us a way to verify the emanation power value given by equation (5).

2.2. Experimental setup and procedure

In order to verify the above theory, a thoron exhalation and measurement system is set up, whose schematic diagram is shown in figure 1. The thoron exhalation pipe is the core component of this system. A certain weight of gas mantles (Captain Stag, Japan) [14] were placed inside the exhalation pipe as a thoron source. As two important factors affecting the thoron emanation power, the humidity in the pipe was controlled between 3% and 5% by a filter of allochroic silica gel (Qingdao Haiyang Chemical Co. Ltd, China), which dried the air from the pump before it entered into the pipe, and the temperature fluctuated around 20 °C ± 5 °C which had little effect on the emanation power. The MP-∑300N pump (Sibata, Japan) can provide the airflow with different flowrate between 0.5 and $3\,\mathrm{l\,min^{-1}}$ in unloaded condition, and in our actual experiment condition, the flowrate was between 0.2 and $1.5\,\mathrm{l\,min^{-1}}$, which was measured by TSI-4146 mass flowmeter (TSI, US). A RAD7 radon/thoron monitor (Durridge, US) was placed at the end of the gas path to measure the thoron concentration of the air outflowed.

To verify that the experimental system is able to meet our measurement purpose, we measured the thoron concentration at the outlet of the exhalation pipe continuously for 12 h and observed its stability at six different flowrates using a batch of 23.7 g gas mantles as thoron source.

Due to thoron's relatively short half-life of 55.6 s, thoron concentration given by RAD7 is more or less lower than that at the outlet of the exhalation pipe, so a correction is necessary for the accurate measurement. For this purpose, we used a small dryer vessel attached to the RAD7 monitor, whose correction factor has been studied in detail [15] and given in equation (7)

$$C_{\mathrm{true}} = R \cdot C_{\mathrm{RAD7}} = (0.650 + 0.347 \cdot q^{-1} + 0.00365 \cdot q^{-2}) \cdot C_{\mathrm{RAD7}}, \tag{7}$$

where R is the correction factor of the small dryer vessel; C_{true} ($\mathrm{Bq\,m^{-3}}$) and C_{RAD7} ($\mathrm{Bq\,m^{-3}}$) are thoron concentrations at the outlet of the exhalation pipe and given by RAD7 respectively; q is the value of flowrate in L/min.

In order to verify the results of above measurement, a $\mathrm{LaBr_3(Ce)}$ scintillator detector (Saint-Gobain, France) with digiBASE-E digital gamma spectrometer (ORTEC, US) was installed next to the exhalation pipe as a confirmatory device (figure 1). It can be used to measure the thoron exhalation fraction χ according to the ^{212}Pb activity A (Bq) before and after the inlet air flow and using equation (8)

$$\chi = \frac{A(^{220}\mathrm{Rn})}{A(^{224}\mathrm{Ra})} = 1 - \frac{A(^{212}\mathrm{Pb})}{A(^{224}\mathrm{Ra})} = 1 - \frac{A(^{212}\mathrm{Pb})_{\mathrm{ventilated}}}{A(^{212}\mathrm{Pb})_{\mathrm{sealed}}} \tag{8}$$

^{212}Pb is one of decay products of thoron and its activity can be measured by using the counting rate of 238.6 keV peak in the gamma spectrum after three days of stable condition (ventilated or sealed). This method is also used in the determination of radon emanation factor [16, 17].

3. Results and discussion

3.1. The stability of thoron concentrations under different flowrates

The measurement result is shown in figure 2. As can be seen from the figure, the thoron concentration is relatively stable at the same flowrate and decreases with the increasing

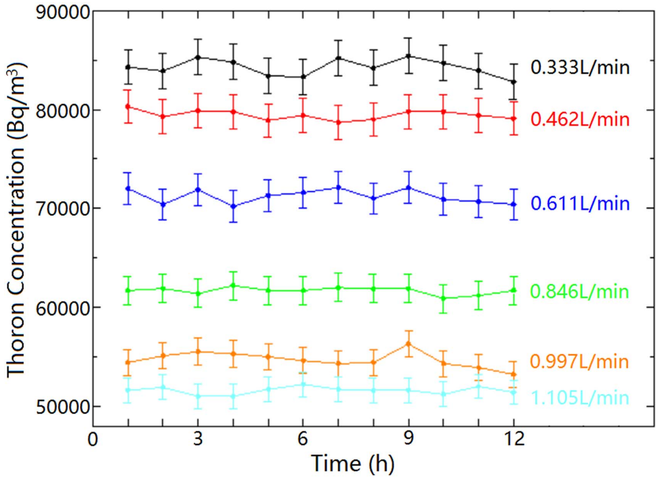

Figure 2. Variation of thoron concentration over time at different flowrates.

Table 1. Results of measured gas mantles mass, ^{232}Th mass activities and the volume of pipes.

Sample no.	Mass of gas mantles m (g)	^{232}Th mass activity A_0/m (Bq g^{-1})	Volume of exhalation pipes V (ml)
1	23.70 ± 0.06	212.5 ± 9.9	163 ± 2
2	29.93 ± 0.06	178.8 ± 8.3	169 ± 1
3	7.30 ± 0.06	257.9 ± 12.1	163 ± 2

flowrate, which means it is feasible to quantitatively measure the thoron emanation power using equation (5) by this measurement system.

3.2. Measurement results of thoron emanation powers

In our experiment, three exhalation pipes filled with three batches of gas mantle samples were tested. The volume of pipes, the mass and the ^{232}Th mass activity of three gas mantle samples and their errors were measured and are shown in table 1.

The measurement results are shown in figure 3. For each sample, we have 5 or 6 data points, which are stable thoron concentrations corresponding to different flowrates. Fitting curves of three samples have the same trend, and are consistent with equation (5) derived above. According to equation (5), we can get thoron emanation power η from the data of each sample by fitting, which are calculated to be 1.33% ± 0.17%, 0.77% ± 0.10% and 0.57% ± 0.07% respectively. The difference between these three values is mainly because of different concentrations of the thorium nitrate solution used in the production process of different batches of gas mantles.

For further study, we put the third sample into a smaller exhalation pipe, whose volume was measured to be 66.8 ± 0.4 ml, and hoped to find out if the volume of exhalation pipe affects the emanation power measured. Figure 4 is the measurement result of the third sample in different size of exhalation pipes. Two thoron emanation powers, 0.57% ± 0.07% and

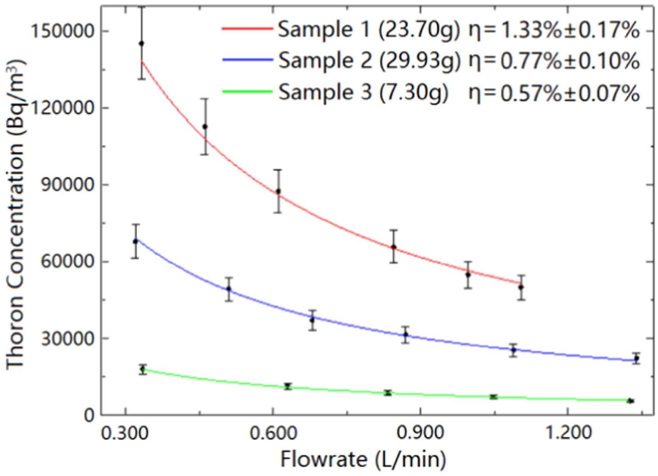

Figure 3. Emanation power fitting curves of three gas mantles samples.

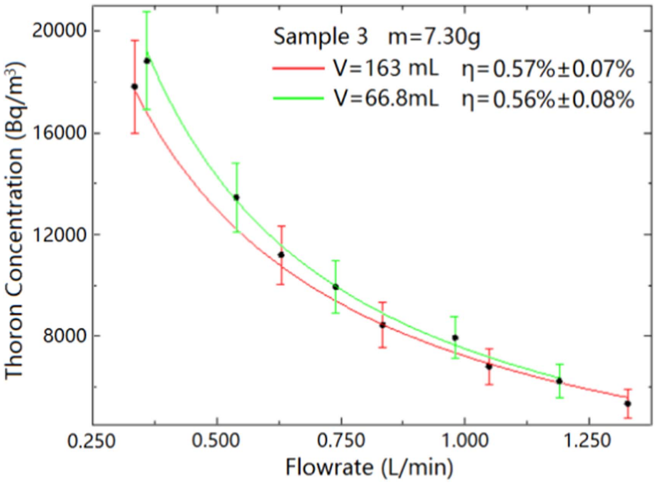

Figure 4. Emanation power fitting curves of the third sample in different pipes.

0.56% ± 0.08%, are very close, which show that the volume of the exhalation pipe has little effect on the emanation power, and the emanation power is quite stable for the same sample.

There are mainly three sources contributing to the errors of emanation powers above. The first is the error of flowrate of each data point, which can be ignored because of the accuracy of TSI mass flowmeter and multiple measurements. The second part is statistical error of fitting procedure, which leads to the standard errors of 0.03%–0.06% on emanation powers (given by ORIGIN). The third part is the uncertainty of thoron concentration measurements. The relative error of a single measurement of thoron concentration by RAD7 is around 2%–18% in our experiment, which can be reduced by means of multiple measurements. Then take the calibration error of RAD7 into account, we can get that total relative error of thoron concentration of each data point is around 9.6%–10.7%, which leads to the standard errors of 0.07%–0.16% on emanation powers (by using error transmission formula). Considering all

Table 2. Comparison between the emanation powers calculated by exhalation fraction and by fitting, and other parameters.

Sample no.	1		2		3	
Volume of exhalation pipes V (ml)	163 ± 2		169 ± 1		163 ± 2	
Flowrate (ventilated only) q (l min^{-1})	1.035 ± 0.011		1.083 ± 0.011		1.024 ± 0.030	
Everage count of 238.6 keV peak in 2 h	Ventilated	Sealed	Ventilated	Sealed	Ventilated	Sealed
	830 996 ± 2330	841 120 ± 1062	1171 663 ± 2576	1181 138 ± 1683	323 678 ± 732	325 708 ± 543
Thoron exhalation fraction χ	1.20% ± 0.30%		0.80% ± 0.26%		0.63% ± 0.28%	
Thoron emanation power by exhalation fraction η_{χ}[a]	1.35% ± 0.34%		0.90% ± 0.29%		0.70% ± 0.31%	
Thoron emanation power by fitting η[b]	1.33% ± 0.17%		0.77% ± 0.10%		0.57% ± 0.07%	

[a] The emanation power calculated by equation (6).

[b] The emanation power fitted by equation (5).

the aspects above, total standard errors of emanation powers can be calculated to be 0.07%–0.17%, corresponding to the relative errors of 12.3%–13.6%.

3.3. Verification experiment result by gamma spectroscopy method

In order to confirm the reliability of the results above, the thoron exhalation fraction were measured by $LaBr_3$ detector. When the pipe was ventilated, the flowrate was set to around $1\,\mathrm{l\,min^{-1}}$, and when the pipe was sealed, valves at both ends of the pipe were closed. After three days of stabilising process, the $LaBr_3$ detector began to work and gave a count value every two hours. For each sample, we got two average count values in ventilated condition and sealed condition. Then using equations (6) and (8), the thoron exhalation fractions and corresponding emanation powers can be calculated respectively. Measurement conditions and results are shown in table 2. We can see that thoron emanation powers calculated by exhalation fraction are consistent with the fitting results within the error range. It is worth mentioning that the difference between the counting rates in ventilated condition and in sealed condition is small, which leads to the relatively large error in emanation power results obtained by gamma spectroscopy. This also shows the advantage of the fitting method by equation (5) on the other hand.

4. Conclusions

In this study, a method has been demonstrated to determine the thoron emanation power of flow-through thoron sources by RAD7 monitor. Our research shows that the thoron concentration of the flow through source will decrease with the increasing flowrate in the gas path, and the quantitative relationship between the concentration and the flowrate can be used to determine the thoron emanation power. The results given by fitting are 1.33% ± 0.17%, 0.77% ± 0.10% and 0.57% ± 0.07% respectively, which are consistent with the results given by gamma spectroscopy within the error range. Furthermore, for a flow-through thoron source with the known emanation power, it can be used to provide long-term stable thoron gas with different target activity concentrations by adjusting the flowrate of the gas path, which can greatly enhance the convenience of the calibration for thoron measurement devices. It should be noted that, this method is applicable to the calibration of a flow-through device, but it cannot be used for a diffusion device because its measurement principle is different from a flow-through device.

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References

[1] Picolo J L 1996 Absolute measurement of radon 222 activity *Nucl. Instrum. Methods Phys. Res.* **369** 452–7

[2] Dersch R and Schotzig U 1998 Production and measurement of ^{222}Rn standards *Appl. Radiat. Isot.* **49** 1171–4

[3] Dersch R 2004 Primary and secondary measurements of ^{222}Rn *Appl. Radiat. Isot.* **60** 387–90

[4] IEC 61577-4 2009 *Radiation Protection Instrumentation—Radon and Radon Decay Product Measuring Instruments—Part 4: Equipment for the Production of Reference Atmospheres Containing Radon Isotopes and their Decay Products (STAR)* (Geneva: International Electrotechnical Commission)

[5] Röttger A, Honig A and Arnold D 2009 The German thoron progeny chamber—concept and application *Appl. Radiat. Isot* **67** 839–42
[6] Sorimachi A, Sahoo S K and Tokonami S 2009 Generation and control of thoron emanated from lantern mantles *Rev. Sci. Instrum.* **80** 015104
[7] Röttger A, Honig A, Dersch R, Ott O and Arnold D 2010 A primary standard for activity concentration of ^{220}Rn (thoron) in air *Appl. Radiat. Isot.* **68** 1292–6
[8] Kanse S D, Sahoo B K, Sapra B K, Gaware J J and Mayya Y S 2013 Powder sandwich technique: A novel method for determining the thoron emanation potential of powders bearing high ^{224}Ra content *Radiat. Meas.* **48** 82–7
[9] Wikipedia: Gas mantle https://en.wikipedia.org/wiki/Gas_mantle
[10] Greeman D J and Rose A W 1996 Factors controlling the emanation of radon and thoron in soils of the eastern USA *Chem. Geol.* **129** 1–14
[11] Iimoto T, Nagai T, Sugiura N and Kosako T 2005 Emanating power of ^{220}Rn from a powdery radiation source *Int. Congr.* **1276** 303–4
[12] Ujić P, Čeliković I, Kandić A and Žunić Z 2008 Standardization and difficulties of the thoron exhalation rate measurements using an accumulation chamber *Radiat. Meas.* **43** 1396–401
[13] RAD7 radon detector, Owner's Manual
[14] Zhang L, Wu J, Guo Q and Zhuo W 2010 Measurement of thoron gas in the environment using a Lucas scintillation cell *J. Radiol. Prot.* **30** 597–605
[15] Ma R, Zhang L and Guo Q 2012 Study on accurate measurement of ^{220}Rn activity concentration by using RAD7 radon monitor *At. Energy Sci. Technol.* **46** 1397–401 (in Chinese)
[16] Mostafa M Y, Vasyanovich M and Zhukovsky M 2016 Prototype of a primary calibration system for measurement of radon activity concentration *Appl. Radiat. Isot.* **107** 109–12
[17] Mostafa M Y A, Vasyanovich M and Zhukovsky M 2017 A Primary standard source of radon-222 based on the HPGe detector *Appl. Radiat. Isot.* **120** 101–5