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NOTE

ESTIMATION OF THORON CONCENTRATION USING SCINTILLATION CELL

C. G. Sumesh*, A. Vinod Kumar, R. N. Nair, R. M. Tripathi and V. D. Puranik
Environmental Assessment Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400085, India

*Corresponding author: sumeshcg@barc.gov.in

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Two counting techniques are proposed in this paper to estimate thoron (^{220}Rn) concentration using a Lucas scintillation cell. The alpha activity build-up inside the cell is calculated theoretically by using Bateman equations. The first method is having a minimum detection limit of 325 Bq m^{-3} and can be used for thoron measurement in thorium-processing plants. In the second method, thoron concentration is calculated using the alpha counts from thoron progenies and is a reference to the first method. The results obtained by these techniques compare well with the double filter method.

INTRODUCTION

The radioactive inert gases, radon and thoron, are present everywhere. The concentration of the gases is measured with different techniques, namely scintillation detectors, pulse ionisation chambers, alpha spectrometric technique and with solid-state nuclear track detectors. Of these, the Lucas scintillation cell (LSC) is one of the most reliable and simple techniques commonly used all over the world for the estimation of radon and thoron.

The cell was originally devised by Vandilla and Taysum[1]. The cell has since been modified by others[2–4]. An air sample is admitted inside the cell through a filter and the concentration is evaluated from the measured disintegration rates and the calibration factor obtained from the theoretical build-up of radon decay products due to a pure radon source. The principle of detection is the counting of photons resulting from the interaction of alpha particles produced by radon and its progeny with the ZnS (Ag) phosphor. A photomultiplier tube assembly counts the photon events and the events are converted into respective concentrations.

Several techniques for the estimation of thoron with the scintillation cell have been attempted by others[5–7]. However, these techniques are not useful in the measurement of thoron gas alone. In this study, estimation of thoron using two techniques are explored, tested and compared with the conventional double filter method (DFM)[8].

In the first method, theoretical estimation of thoron concentration is made by using total alpha counts obtained due to thoron and its first progeny (^{216}Po). In the second method, the same is calculated using the alpha counts only due to thoron progenies (^{212}Pb/^{212}Bi) by applying Bateman's equations. Counts obtained for any interval of time from the start of sampling will correspond to the alpha decay of thoron and progeny concentrations present in the cell during that counting interval. The first method will give an estimation of thoron concentration and the second will be a reference to the concentration measured by the first method. This reveals the presence of counts due to thoron progenies in the cell even after the thoron gas has decayed out.

MATERIALS AND METHODS

Mathematical formulation

An air sample containing only thoron (^{220}Rn) atoms is sampled to a scintillation cell at t=0. If A_0 is the initial activity given by $A_0 = N_1^0\lambda_1$. Let $A_1(t)$, $A_2(t)$, $A_3(t)$, $A_4(t)$ and $A_5(t)$ denote the activities of ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi and ^{212}Po at any time t, respectively. They can be related to A_0 using Bateman's equations[9] as:

$$A_1(t) = A_0 e^{-\lambda_1 t} \tag{1}$$

$$A_2(t) = A_0\lambda_2\left\{\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)}\right\} \tag{2}$$

$$A_4(t) = A_0\lambda_2\lambda_3\lambda_4\left\{\begin{array}{c}\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)} \\ +\frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)} \\ +\frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)} \\ +\frac{e^{-\lambda_4 t}}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)}\end{array}\right\} \tag{3}$$

$$A_5(t) = A_0\lambda_2\lambda_3\lambda_4\lambda_5\left\{\begin{array}{l}\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)(\lambda_5-\lambda_1)}\\ \quad+\frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)(\lambda_5-\lambda_2)}\\ \quad+\frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)(\lambda_5-\lambda_3)}\\ \quad+\frac{e^{-\lambda_4 t}}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)(\lambda_5-\lambda_4)}\\ \quad+\frac{e^{-\lambda_5 t}}{(\lambda_1-\lambda_5)(\lambda_2-\lambda_5)(\lambda_3-\lambda_5)(\lambda_4-\lambda_5)}\end{array}\right\} \quad (4)$$

$A_3(t)$ is not considered since it is a beta emitter. Where t is in seconds and activities are in Becquerel. λ_1, λ_2, λ_3, λ_4 and λ_5 are the decay constants per second (s^{-1}) of ^{220}Rn, ^{216}Po, ^{212}Pb, ^{212}Bi and ^{212}Po, respectively. The decay series of thoron is shown in Table 1. The total number of disintegrations, D_t, emitted during a time interval from beginning of sampling to any time 't' can be estimated using the integral:

$$D_t = \int_0^t (A_1(t) + A_2(t) + 0.36A_4(t) + 0.64A_5(t))\,dt \quad (5)$$

Weighting to the branching of ^{212}Bi through alpha and beta decay in the thoron decay series are taken care by using appropriate fractions in eq. (5).

On integrating eq. (5), the following is obtained:

$$D_t = A_0\left\{\begin{array}{l}C_1(1-e^{-\lambda_1 t}) + C_2(1-e^{-\lambda_2 t}) + C_3(1-e^{-\lambda_3 t})\\ \quad + C_4(1-e^{-\lambda_4 t}) + C_5(1-e^{-\lambda_5 t})\end{array}\right\} \quad (6)$$

where C_1, C_2, C_3, C_4 and C_5 are constants given by

$$C_1 = \frac{1}{\lambda_1} + \frac{\lambda_2}{\lambda_1(\lambda_2-\lambda_1)} + \frac{0.36\lambda_2\lambda_3\lambda_4}{\lambda_1(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)} + \frac{0.64\lambda_2\lambda_3\lambda_4\lambda_5}{\lambda_1(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)(\lambda_4-\lambda_1)(\lambda_5-\lambda_1)}$$

$$C_2 = \frac{1}{(\lambda_1-\lambda_2)} + \frac{0.36\lambda_3\lambda_4}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)} + \frac{0.64\lambda_3\lambda_4\lambda_5}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)(\lambda_4-\lambda_2)(\lambda_5-\lambda_2)}$$

$$C_3 = \frac{0.36\lambda_2\lambda_4}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)} + \frac{0.64\lambda_2\lambda_4\lambda_5}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)(\lambda_4-\lambda_3)(\lambda_5-\lambda_3)}$$

$$C_4 = \frac{0.36\lambda_2\lambda_3}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)} + \frac{0.64\lambda_2\lambda_3\lambda_5}{(\lambda_1-\lambda_4)(\lambda_2-\lambda_4)(\lambda_3-\lambda_4)(\lambda_5-\lambda_4)}$$

$$C_5 = \frac{0.64\lambda_2\lambda_3\lambda_4}{(\lambda_1-\lambda_5)(\lambda_2-\lambda_5)(\lambda_3-\lambda_5)(\lambda_4-\lambda_5)}$$

On substituting the values of decay constants, $C_1{=}160{\cdot}6806$, $C_2{=}-0{\cdot}2170$, $C_3{=}88{\cdot}7698$, $C_4{=}-8{\cdot}5407$ and $C_5{=}3{\cdot}78\times10^{-34}$ are obtained.

First method (prompt method)

If the counting is started immediately (within a few seconds) after sampling, the build-up of activity inside the cell depends only on the activity of ^{220}Rn and ^{216}Po since in this case the contribution due to thoron daughters can be assumed as zero. Therefore, eq. (6) can be rewritten as

$$D_t = A_0\{160.6806(1-e^{-\lambda_1 t}) - 0.217(1-e^{-\lambda_2 t})\} \quad (7)$$

Let T be the counting time from the sampling period then the total number of disintegrations during this period 0 to $t{+}T$ is

$$D_{t+T} = A_0\{160.6806(1-e^{-\lambda_1(t+T)}) - 0.217(1-e^{-\lambda_2(t+T)})\} \quad (8)$$

Therefore, the number of disintegrations during the counting period is

$$D_{t+T} - D_t = A_0\{160.6806e^{-\lambda_1 t}(1-e^{-\lambda_1 T}) - 0.217e^{-\lambda_2 t}(1-e^{-\lambda_2(t+T)})\} \quad (9)$$

If the counting period (T) is $>$60 s then the exponential term involving the decay constant of ^{216}Po becomes almost zero, i.e. $e^{-\lambda_2 t}=0$, In that case eq. (9) reduces to

$$D_{t+T} - D_t = A_0\{160.4636e^{-\lambda_1 t}(1-e^{-\lambda_1 T})\} \quad (10)$$

or

$$A_0 = \frac{D_{t+T} - D_t}{160.4636e^{-\lambda_1 t}(1-e^{-\lambda_1 T})} \quad (11)$$

Let $D = (D_{t+T} - D_t)$ be the counts obtained in the counting interval T, then D is related to the initial

Table 1. Decay series of thoron.

Radionuclide	Decay constant	Notation	Decay mode	Half-life (s)	Product of decay
^{220}Rn	λ_1	A_1	α	55·6	^{216}Po
^{216}Po	λ_2	A_2	α	0·15	^{212}Pb
^{212}Pb	λ_3	A_3	β	38304	^{212}Bi
^{212}Bi	λ_4	A_4	α	3636 (36 %)[a]	^{208}Tl
			β	3633 (64 %)[a]	^{212}Po
^{212}Po	λ_5	A_5	α	$3{\cdot}05\times10^{-7}$	^{208}Pb

[a]Branching ratio.

activity of the sample through the relation:

$$A_0 = \frac{D}{160.4636\mathrm{e}^{-\lambda_1 t}(1-\mathrm{e}^{-\lambda_1 T})} \tag{12}$$

or

$$A_0 = \frac{6.22\times10^{-3}\times D}{\mathrm{e}^{-\lambda_1 t}(1-\mathrm{e}^{-\lambda_1 T})} \tag{13}$$

Taking into account volume V and efficiency E of the scintillation cell, eq. (13) can be used to estimate the concentration of ^{220}Rn in the cell as:

$$C = \frac{6.22\times10^{-3}\times D\times100}{E\times V\times\mathrm{e}^{-\lambda_1 t}(1-\mathrm{e}^{-\lambda_1 T})} \tag{14}$$

where C is the concentration of ^{220}Rn in the cell (Bq m^{-3})

Second method (delayed method)

It is seen that if $t \geq 17$ min (1020 s) the exponential terms involving the decay constant of ^{220}Rn, ^{216}Po and ^{212}Po in eq. (6) becomes almost zero. $\mathrm{e}^{-\lambda_1 t}=0, \mathrm{e}^{-\lambda_2 t}=0, \mathrm{e}^{-\lambda_5 t}=0$

Hence, for 't' >17 min, say 20 min, eq. (6) can be modified as

$$D_t = A_0\{160.4636 + 88.8722(1-\mathrm{e}^{-\lambda_3 t}) -8.5457(1-\mathrm{e}^{-\lambda_4 t})\} \tag{15}$$

Let T be the counting time after the delay period of 't' (>17 min), then the total number of disintegrations emitted is

$$D_{t+T} = A_0\{160.4636 + 88.8722(1-\mathrm{e}^{-\lambda_3 (t+T)} -8.5457(1-\mathrm{e}^{-\lambda_4 (t+T)})\} \tag{16}$$

Therefore, the number of disintegrations during the counting period T is

$$D_{t+T} - D_t = A_0\{88.8722\mathrm{e}^{-\lambda_3 t}(1-\mathrm{e}^{-\lambda_3 T}) -8.5457\mathrm{e}^{-\lambda_4 t}(1-\mathrm{e}^{-\lambda_4 T})\} \tag{17}$$

or

$$A_0 = \frac{D_{t+T}-D_t}{88.8722\mathrm{e}^{-\lambda_3 t}(1-\mathrm{e}^{-\lambda_3 T}) - 8.5457\mathrm{e}^{-\lambda_4 t}(1-\mathrm{e}^{-\lambda_4 T})} \tag{18}$$

If $D = (D_{t+T} - D_t)$ be the counts obtained in the counting interval T, 'V' be the volume and 'E' be the % efficiency of the scintillation cell, then eq. (18) can be used to estimate the concentration of ^{220}Rn in the cell as:

$$C = \frac{D\times100}{E\times V\times Z} \tag{19}$$

where $Z = 88.8722\mathrm{e}^{-\lambda_3 t}(1-\mathrm{e}^{-\lambda_3 T}) \quad -8.5457\mathrm{e}^{-\lambda_4 t}(1-\mathrm{e}^{-\lambda_4 T})$ and C is the concentration of ^{220}Rn in the cell (Bq m^{-3})

Experimental set-up

Laboratory scale experiments were carried out for the estimation of thoron using LSC and DFM (DF method). A leak proof cubical chamber of 512 l capacity was used to contain the gas for the experiments. Thorium oxalate powder (^{232}Th) separated from monazite, packed in a porous container was used to generate thoron (^{220}Rn) in the chamber. By varying the thorium oxalate powder quantity, ^{220}Rn concentration was varied inside the chamber. A constant ventilation rate was maintained in the chamber to keep the thoron concentration at a constant level. A small fan was used for uniform mixing of the gas inside the chamber. A sampling port at one side of the chamber was used for sample

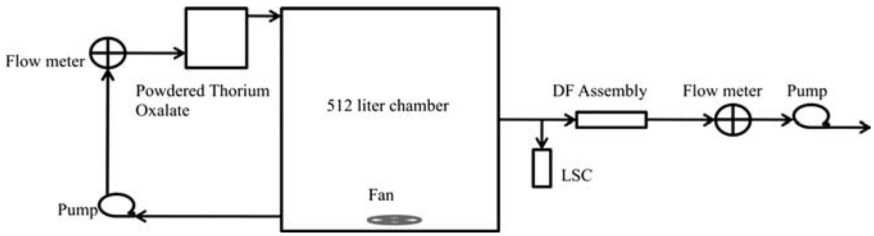

Figure 1. Experimental set-up used for the thoron concentration measurement.

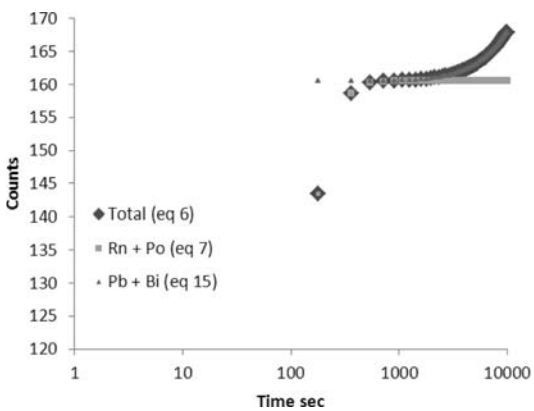

Figure 2. Integrated counts obtained for different counting periods for eq. (6) (total), eq. (7) (Rn+Po) and eq. (15) (Pb+Bi).

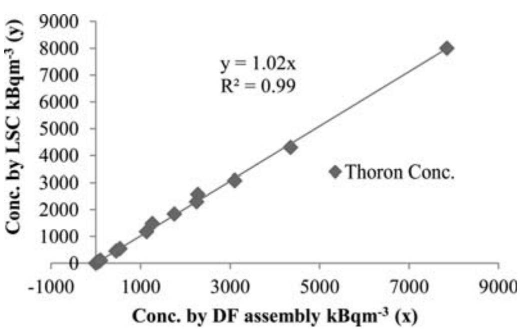

Figure 3. Linear fit of thoron concentration obtained in LSC (prompt method) and DFM.

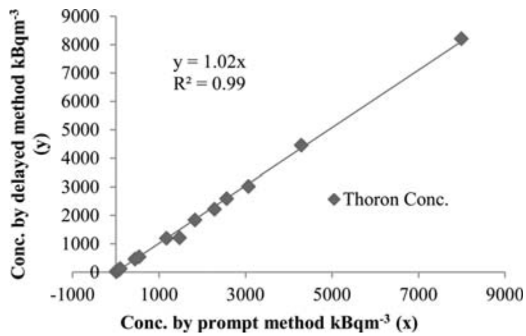

Figure 4. Linear fit of concentration obtained in LSC (prompt method) and delayed method.

collection. Figure 1 shows the experimental set-up used for the thoron concentration measurement.

A double filter (DF) assembly connected to the chamber measured thoron concentration simultaneously with the Lucas cell. The flow rate of gas through the DF assembly was kept at 8–12 litres per min and sampling time for the DF assembly was fixed at 1 min. As the volume sucked out was small compared with the chamber volume, the concentration of ^{220}Rn in the air sample was assumed to be same from the start to end of the sampling. The exit filter was counted immediately after sampling. The Lucas cell is counted immediately after sampling and continued for 200 min.

The experiments were also repeated with various flow rates of gas through DF assembly to see the reproducibility of the results. Similarly, thoron concentration is calculated with different delay periods and different counting period in LSC was also carried out to ascertain the reproducibility of the results.

RESULTS AND DISCUSSION

The results of theoretical computations carried out using eqs (5), (6) and (15) are shown in Figure 2. In the initial stage, total disintegration is mainly dependent on the disintegrations of thoron and polonium. Later on it depends on the disintegration of lead and bismuth. Figure 2 show that thoron gas concentration can also be estimated using eqs (7) and (15). In case of eq. (7), the counting has to start immediately after sampling, whereas a delay period not $<$17 min is required for eq. (15). The thoron concentrations estimated using scintillation cell using a prompt method is compared with samples taken simultaneously through the DF method. A good correlation is seen as shown in Figure 3. Similarly, thoron concentration is calculated using a delayed counting method also gives a good correlation with the prompt method as shown in Figure 4.

The normalised mean square error (NMSE) of the standard method DFM to the prompt method is found to be $7{\cdot}7\times10^{-4}$ and NMSE for prompt method to delayed method is found to be $6{\cdot}05\times10^{-7}$. For a perfect model, NMSE should be zero. Since the calculated values are close to zero, uncertainties in measuring thoron in this method is very low. The coefficient of variation for repeated measurement in scintillation cell was found to be $<12\%$ indicating a high reproducibility of the measurement technique. Within 240 s, immediately after sampling, a good measurement can be obtained using this method whereas other methods, such as DF, require a few hours for the same measurements. The scintillation cell system can be used for thoron-alone measurements in situations where the radon concentration is nil or negligible. In the present experimental set-up, a background count rate of 6 counts per hour and a counting period of 420 s will result in a minimum detection limit (MDL) of 325 Bq m^{-3} for thoron concentrations, whereas the second method has a higher MDL of 10000 Bq m^{-3}. However, this value can be brought down to a much lower value if a large volume Lucas cell[10] is used. The present method cannot be used for environmental samples as the MDL for the system is above the normal atmospheric concentrations.

CONCLUSION

The methods developed in this study to estimate thoron concentration using LSC are simple and reliable. The method is simple and found to be as accurate as conventional methods like the DFM. The method is tested successfully against the DFM technique. This method can be used to estimate the thoron concentration in thoron calibration chamber as well as in Monazite-processing plants where the expected radon concentration is negligible. The second method will be a reference to the concentrations obtained from the first set of measurements and enable to estimate the thoron concentration even after the gas has decayed appreciably (after seven half-lives of thoron). This will confirm the presence of thoron daughters in the cell even after the thoron has completely decayed out.

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