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A simple method for calibration of Lucas scintillation cell counting system for measurement of ^{226}Ra and ^{222}Rn

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ABSTRACT

Known quantity of radium from high grade ore solution was chemically separated and carefully kept inside the cavity of a Lucas Cell (LC). The ^{222}Rn gradually builds up and attain secular equilibrium with its parent ^{226}Ra. This gives a steady count after a suitable buildup period (>25 days). This secondary source was used to calibrate the radon counting system. The method is validated in by comparison with identical measurement with AlphaGuard Aquakit. The radon counting system was used to evaluate dissolved radon in ground water sample by gross alpha counting in LC. Radon counting system measures the collected radon after a delay of >180 min by gross alpha counting. Simultaneous measurement also carried out by AlphaGuard Aquakit in identical condition. AlphaGuard measures dissolved radon from water sample by constant aeration in a closed circuit without giving any delay. Both the methods are matching with a correlation coefficient of >0.9. This validates the calibration of Lucas scintillation cell counting system by designed encapsulated source. This study provides an alternative for calibration in absence of costly Radon source available in the market.

1. Introduction

Lucas scintillation cell has been used to measure ^{226}Ra by growth of ^{222}Rn in sealed, evacuated glass vial (Butts, Todd, Lerche, Moore, & Moore, 1988; Lucas, 1957). The Lucas Scintillation cell (LC) and its modifications are among the most sensitive instruments to detect low levels of radon (Semkow, Parekh, Schwenker, Dansereau, & Webber, 1994). Lucas cell is a cylindrical chamber in which internal surface is coated with silver activated zinc sulfide. The ZnS (Ag) has a scintillation property of 4500 A° maximum wave length with a decay time of 5 S (Manquing, 1991). When an alpha particle collides with the ZnS phosphor, it produces a light signal which is detected by a photomultiplier tube which translates the photon into an electrical count (Damkjaer and Korshech, 1985; George, 1976; Quindos, Fernandez, & Soto, 1994). Since Rn decays to Po in the scintillation cell, which subsequently produces Pb, up to three (for ^{222}Rn) alpha particles can be counted from one Rn nuclide. Therefore, the scintillation cell

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has a high efficiency (0.83 for ^{222}Rn; Mathieu, Biscaye, Lupton, & Hammond, 1988). The sample containing radium is allowed to hold in an airtight container. It allows the decay of radium present in the sample to form radon in an airtight device for a known period of time. The inbuilt radon from the container is transferred to a calibrated Lucas cell (Kristan & Kobal, 1973). Counting of gross alpha activity in the Lucas cell is carried out after 200 min of delay to ensure equilibrium between ^{222}Rn and its short lived progeny (Jha et al., 2010; Sethy et al., 2011, 2013). The method is known as emanometry (Markose, 1981; Raghavayya, Iyenger, Markose, 1979; Sedlacek, Sebesta, & Benes, 1980; Tripathi et al., 2012), is convenient and cost effective. The Lucas cells were coupled with a radon alpha counting system to get the alpha counts. Gross alpha counts obtained is converted to activity of ^{226}Ra present in the sample. A radon alpha counter consists of a PM tube which can be coupled with the transparent side of the Lucas cell. The radon inside the Lucas cell decays forming alpha emitting daughters. The alpha particles are interacting with the wall coated scintillating materials and produces photon. As in the radon counting system Lucas cell is counted the system is need to be calibrated using a known activity of radon in a Local cell (or standard). In this study a Radium–Radon source is designed and used for calibration of the Radon counting system. The calibrated instrument is compared with alpha guard measured values by simultaneous measurement of dissolved radon in ground water.

1.1. *Theory*

If a known activity of ^{226}Ra is kept inside a Lucas cell (Fig. 1) and the cell is closed and airtight the radon gas formed will attains equilibrium within next few weeks. ^{226}Ra in this study was derived from high grade uranium ore in which uranium is in secular equilibrium with its daughters. Let us consider the decay of ^{226}Ra, denoted by subscript 1, to produce ^{222}Rn, denoted by subscript 2. Thus,

$$-\frac{dN_1}{dt} = \lambda_1 N_1$$

$$N_1 = N_1^0 e^{-\lambda_1 t}$$

where, N_1^0 to represent the value of ^{226}Ra at $t = 0$.

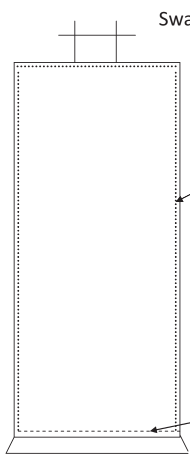

Fig. 1 – Schematic diagram of Lucas cell.

Now the radon is formed at the rate at which the ^{226}Ra decays, $\lambda_1 N_1$, and also it decays at the rate of $\lambda_2 N_2$. Hence

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

$$\frac{dN_2}{dt} = \lambda_1 N_1^0 e^{-\lambda_1 t} - \lambda_2 N_2$$

$$\frac{dN_2}{dt} + \lambda_2 N_2 = \lambda_1 N_1^0 e^{-\lambda_1 t}$$

By multiplying both sides by $e^{\lambda 2t}$:

$$e^{\lambda_2 t}\frac{dN_2(t)}{dt} + \lambda_2 N_2 e^{\lambda_2 t} = \lambda_1 N_1^0 e^{(\lambda_2-\lambda_1)t}$$

$$\frac{d}{dt}\left(N_2 e^{\lambda_2 t}\right) = \lambda_1 N_1^0 e^{(\lambda_2-\lambda_1)t}$$

Intergrating:

$$N_2 e^{\lambda_2 t} = \frac{\lambda_1}{\lambda_2-\lambda_1} N_1^0 e^{(\lambda_2-\lambda_1)t} + C$$

$$N_2 = \frac{\lambda_1}{\lambda_2-\lambda_1} N_1^0 e^{-\lambda_1 t} + Ce^{-\lambda_2 t}$$

When $t = 0$, $N_2 = N_2^0$:

$$C = \frac{\lambda_1}{\lambda_2-\lambda_1} N_1^0 + N_2^0$$

$$N_2 = \frac{\lambda_1}{\lambda_2-\lambda_1} N_1^0\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^0 e^{-\lambda_2 t}$$

$\frac{dN_2}{dt} - \lambda_1 N_1 + \lambda_2 N_2 = 0$, is a linear differential equation of the first order and the solution of which is given by

$$N_2 = \frac{\lambda_1}{\lambda_2-\lambda_1} N_1^0\left(e^{-\lambda_1 t} - e^{-\lambda_2 t}\right) + N_2^0 e^{-\lambda_2 t}$$

where, N_2^0 is the value of no of ^{222}Rn atoms at $t = 0$. The first group of terms shows the growth of ^{222}Rn from the ^{226}Ra. This can be also written in terms of activity as

$$A_{Rn}(t) = A_{Ra}^0 \frac{\lambda_{Rn}}{\lambda_{Rn}-\lambda_{Ra}}\left(e^{-\lambda_{Ra} t} - e^{-\lambda_{Rn} t}\right) + A_{Rn}^0 e^{-\lambda_{Rn} t} \quad (1)$$

1.2. *Secular equilibrium in uranium ore*

Freshly collected undisturbed uranium ore was selected to ascertain the secular equilibrium (Jha et al., 2010; Jha, Tripathi, Sethy, Sahoo, & Puranik, 2013; Narayana & Rajashekara, 2010; Sethy et al., 2013) status of the uranium series radionuclides (Table 1). Three long lived alpha emitting radio nuclides were selected viz. U (nat), ^{226}Ra and ^{210}Po for equilibrium study.

Table 1 – Secular equilibrium in uranium ore.

Sl. no.	Sample identification	^{238}U	^{226}Ra	^{210}Po
		Bq g^{-1}	Bq g^{-1}	Bq g^{-1}
1.	Low grade ore (a)	6.58	6.25	6.43
2.	Low grade ore (b)	5.59	5.49	5.24
3.	High grade ore (a)	12.18	14.66	12.49
4.	High grade ore (b)	12.52	13.98	14.63

Details of analysis of these radionuclide and quality control of analytical procedures are described elsewhere (Jha et al., 2010; 2013; Kolthoff & Eiving, 1962; Markose, 1990, p. 62; Sethy et al., 2011, 2013).

A known quantity of ore was dried, ground, sieved (100 mesh) and homogenized. 1 g of the representative sample was digested in microwave reaction system (Antaon paar multiwave 3000) and the aliquot (8.7 mg ml^{-1}) was preserved in 0.25 N nitric acid for radiochemical analyses. Uranium (nat) was analyzed fluorimetrically after separation of uranium by solvent extraction process (Sethy et al., 2011, 2013). ^{226}Ra was analyzed by allowing the buildup of its progeny ^{222}Rn in a bubbler and counting the total alpha activity by collecting the radon gas into a scintillation cell (Jha et al., 2010, Raghavayya et al., 1979; Sethy et al., 2011). ^{210}Po was analyzed by spontaneous electrodepositing (Figgins, 1961; Jha et al., 2013; Narayana & Rajashekara, 2010) on a brightly polish silver disc and alpha counting. The details of analysis of ore are presented in Table 1.

2. Experimental

The LCs used in this study (Fig. 1) was having internal height 7 cm and radius of 2.2 cm. It has one end connected to a Swagelok quick connecter for sampling/evacuation of radon and opposite window is sealed by transparent glass for coupling with counting device. The Swagelok quick is removed to keep a known activity of ^{226}Ra in the Swadge luck cavity in the inside of the Lucas cell (Fig. 2). The modified cells were opened from glass window side. From the stock solution known activity of ^{226}Ra was transferred drop wise to the area earlier occupied by Swagelok quick connecter in Lucas cell. The active solutions are allowed to dry under infrared lamps. The dried activity is carefully covered by a thin slice of sponge to prevent its loss from the cavity. Use of sponge as a radon penetrating media has been described by in a number of studies worldwide. (Al-Jarallah, Fazal-ur-Rehman, & Abdalla, 2008; Ismail, 2006; Yamamoto, Tarutani, Yamasoto, Iskandar, & Iida, 2000) sponge slice is given as cover in order to prevent loss of planchated ^{226}Ra in the cavity inside the LS. There is a decrease in efficiency of LS expected due to loss in effective area of Zns (Ag) coating. The Lucas cells were made airtight and kept for buildup of ^{222}Rn inside it. After a period of 3–4 weeks a steady count was obtained due to secular equilibrium. The measurement of radon and its progeny in Lucas cell method considers homogeneous distribution of ^{222}Rn inside the cell volume and attachment of ^{218}Po and ^{214}Po to the cell wall (Al-Jarallah et al., 2008; Lenzen & Neugebauer, 1996a; Lenzen & Neugebauer, 1996b). The Lucas cell containing the activity was counted continuously until a steady count is obtained. The counts are become steady after equilibrium between ^{226}Ra.

^{222}Rn and its daughters are established within 30 days. After 23 days, >98% from radium–radon equilibrium is attained (Moldovan, Cosma, Encian, & Dicu, 2009). These ^{226}Ra–^{222}Rn source giving steady counts corresponding to the activity present inside it were used to calibrate the radon alpha counting system.

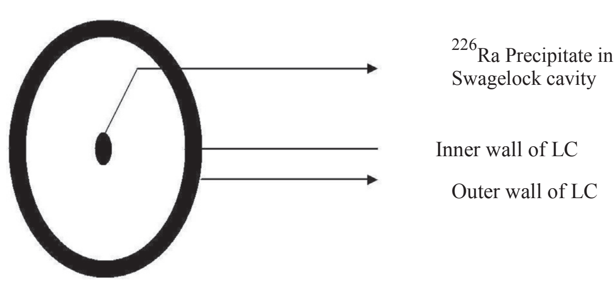

Fig. 2 – Schematic diagram of Lucas cell with sealed source.

3. Results

The prepared sources are kept for decay of ^{226}Ra and growth of ^{222}Rn for 30 days to obtained secular equilibrium between ^{226}Ra and ^{222}Rn. The possible interference due to ^{224}Ra and ^{223}Ra was eliminated during this growth period. In the mean time the sources are alpha counted daily to known the growth pattern of ^{222}Rn inside the local cell. The counts of the sealed source were grown gradually with attending a steady count within three weeks. The results of gross alpha counting are presented in Figs. 3, 4 and 5. It is observed that the counts obtained are directly proportional to the amount of activity present inside the LC. Figs. 3, 4 and 5 represents typical growth of radon inside the Lucas cell and the counts are becoming steady as the equilibrium between ^{226}Ra and ^{222}Rn approaches. The activity of U (nat) and ^{226}Ra are seems comparable from the analytical results indicating near secular equilibrium in the ore sample. As discussed earlier about expected decrease in efficiency due to sponge cover (dead area) over the radium source, an approximation is made in order to quantify the loss in efficiency. Dead area is the portion of the zinc sulfide coating in which alpha particle interaction remain undetected due to sponge cover. The total effective surface are of ZnS (Ag) inside a Lucas cell is about 112 square centimeter ($h = 7$ cm and $r = 2.2$ cm), out of which the area occupied by the sponge is 15.2 square centimeter. A standard Lucas cell of identical geometry with plated radium source inside with known rated counts and efficiency was used for calibration. The rated count of the said cell was 21,500 CPM at 75% efficiency. Four different scintillation counters were used for calibration of the setup (scintillation cell coupled with PMT &

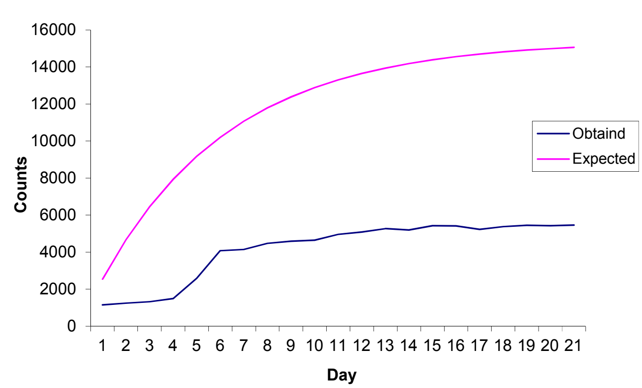

Fig. 3 – Expected count vs actual counts from Source-1.

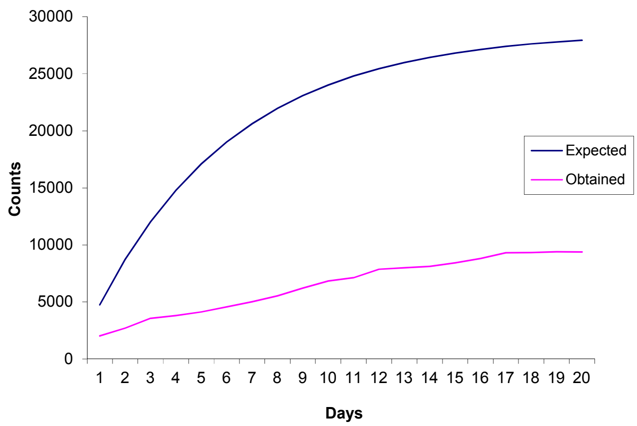

Fig. 4 – Expected count vs actual counts from Source-2.

counter). The average efficiency was worked out to be 88%. Since an effective area of 112 square centimeter provides 88% efficiency in the radon alpha counter, 12% decrease in efficiency was calculated due to reduction in effective area by sponge cover. An average efficiency of 76% thus worked out was used for actual activity evaluation. The counts obtained shows similar growth pattern as theoretically expected with a correlation coefficient of $r = 0.948$ ($p < 0.02$). As evident from the plot, alpha counts were increasing and then attains steady counts within 20 days. The efficiencies of sealed sources were (Table 2) varied from 0.15 to 0.30 cps Bq^{-1}.

4. Field application

In order to validate the prepared encapsulated $^{226}Ra–^{222}Rn$ source the radon alpha counting system was calibrated considering these LCs. Measurement of dissolved radon in community ground water samples were carried out using radon bubbler and subsequent measurement in radon alpha counting system. Radon concentration in water was also measured simultaneously using the professional radon monitor AlphaGuard (made by GENITRON, Germany) at identical conditions.

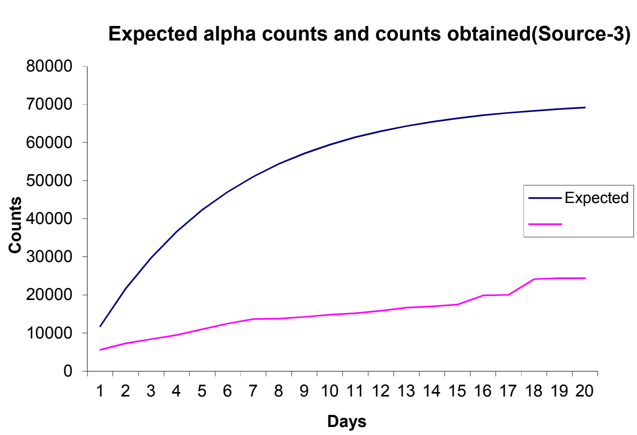

Fig. 5 – Expected count vs actual counts from Source-3.

Table 2 – Details of counting and efficiencies of Source prepared.

Source	Source-1	Source-2	Source-3
Activity of ^{226}Ra inside LC	9.87	19.74	49.35
Expected counts after 30 days at 76% E of LC	13,492	26,985	67,462
Mean counts obtained after 30 days	5464	6813	14,101
Counts obtained due to ^{222}Rn alpha	1821	2271	4700
Counting time (min)	10	10	10
Efficiency of LC for ^{222}Rn (cps Bq^{-1})	0.30	0.19	0.15
Efficiency of Radon alpha counter	0.40	0.25	0.20

4.1. *Measurement by alpha guard Aquakit*

Alpha guard is an ionization chamber, designed for measuring radon in air, soil and water. For water measurements the additional equipment Aqua KIT was used. The reliability of Alpha guard is studied by different researcher (Jilek, Thomas, & Brabec, 2008; Kochowska, Kozak, Kozlowska, Mazur, & Dorda, 2009; Shubert, Buerkin, Pena, Lopez, & Balcazar, 2006; Yasuoka et al., 2010) and can be considered for comparison in field studies. Ground water samples of 100 mL were placed for radon activity measurements in appropriate glass vessels (Aqua KIT) connected to detector through the air pump, which following the recommendations of the manufacturer (Genitron, 2005) and was adjusted to continuous air flow of 0.5 L min^{-1}. It directly draws radon from the water sample that feeds to the ionization chamber of the Alphaguard in a closed circuit. The Alpha guard displays the radon concentration in air which in turn gives the activity concentration of Radon in water by following equation.

$$C_{water} = C_{air}\left[\frac{(V_{system} - V_{sample})}{V_{sample}} + K\right] - Co$$

where

C_{water} is the dissolved radon in water (Bq l^{-1})
C_{air} is the radon concentration in the measuring setup or AlphaGuard reading (Bq m^{-3})
Co is background of alpha guard (Bq m^{-3})
V_{system} is interior volume (ml) of measuring setup including ionization chamber of alpha guard, alpha pump, sampling vessels and tubing
K is radon distribution coefficient (0.26)

4.2. *Measurement by radon bubbler-radon alpha counting system*

The radon alpha counters were calibrated using the prepared encapsulated radon source. The estimation of radium by radon bubbling method have been described elsewhere (Jha et al., 2010; Markose, 1981; Raghavayya, Iyengar & Markose, 1980; Tripathi et al., 2012; Sethy et al., 2013; USEPA). In the bubbler method the water sample is carefully transferred into a radon bubbler. Radon from the bubbler was collected in previously evacuated scintillation cell without any delay. The

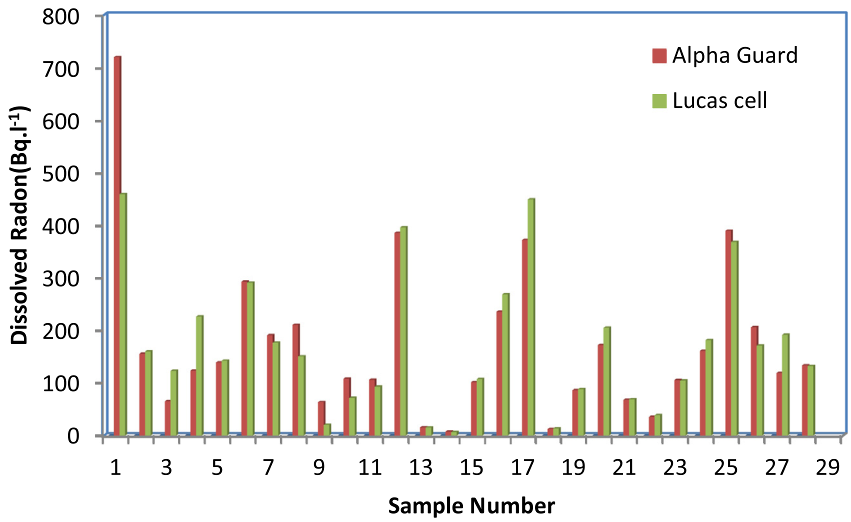

Fig. 6 – Dissolved radon measured in Alphaguard and Lucas cell method.

LCs containing the samples were given 200 min delay in order to attains equilibrium between radon and its progeny. Then LCs was counted for alpha activity in the radon alpha counting system. The dissolved radon concentration in water samples is given by

$$^{226}\mathrm{Ra}(Bq) = \frac{1.883 \times 10^{-3} \times C \times 0.037}{E \times (1 - e^{-\lambda T}) \times e^{-\lambda T}}$$

C = is the net counts obtained after subtraction of the background
E = Efficiency of the cell (75%)
t = counting delay in minutes
T = counting duration in minutes
λ = Decay constant of ^{222}Rn (1.258×10^{-4} min^{-1})
V = Volume of water (lit) collected in the bubbler

The gross alpha counts obtained in the bubbler method is converted to activity concentration of radon in water and compared with the values obtained by alpha guard measurement. Using the mean count rate (cpm) from the source the efficiency of the counting system was estimated. The gross counts obtained from the sample were converted to dissolve radon concentration.

5. Conclusions

The activity concentration of radon in water samples collected in identical conditions were compared with that of values measured by Alpha guard (Fig. 6). The result of both gross counting method using calibrated radon counter and alpha guard measurement were closely matching with a correlation >0.9. Following conclusions can be made out of this study.

1. The technique is to calibrate the Lucas scintillation counting system for measurement ^{226}Ra and ^{222}Rn.
2. The efficiencies of sealed sources were varied from 0.15 to 0.30 cps Bq^{-1}.
3. Good agreement (r^2 = 0.91) with alpha guard measured values validates this method for use in routine radiochemical measurement of ^{226}Ra and ^{222}Rn.
4. This is a simple cost effective method to calibrate the radon alpha counting system using encapsulated ^{226}Ra–^{222}Rn source.

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