Determination of radon leakage from sample containers for gamma spectrometry measurement of $^{226}$Ra

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02 MATERIALS AND METHODS
03 RESULTS AND DISCUSSION
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Introduction
Abstract

Marinelli Beaker Release Fraction ($MB_{LR}$)
### Background Theory

#### Measuring Radium in Soil Sample by HPGe

<table>
<thead>
<tr>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>✓ Versatile</td>
<td>✓ weak yields γ-line 186.2 keV (3.59%),</td>
</tr>
<tr>
<td>✓ easy to use</td>
<td>✓ an interference with 235U direct line 185.7 keV</td>
</tr>
<tr>
<td>✓ non-destructive</td>
<td>✓ longer time needed to achieve secular equilibrium (at least 21 days)</td>
</tr>
<tr>
<td>✓ relatively cheap method</td>
<td>✓ radon leakage from the measurement container causes the equilibrium cannot be reached.</td>
</tr>
<tr>
<td>✓ Repeatability</td>
<td></td>
</tr>
<tr>
<td>✓ easy sample preparation</td>
<td></td>
</tr>
<tr>
<td>✓ easier spectrum analysis</td>
<td></td>
</tr>
</tbody>
</table>

### Table 1. Advantages and disadvantages of gamma spectrometry methods

<table>
<thead>
<tr>
<th>Gamma spectrometry</th>
<th>Advantage</th>
<th>Disadvantage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direct measurement</td>
<td>✓ analyzing Ra-226 by its 186 keV peak.</td>
<td>✓ weak yields γ-line 186.2 keV (3.59%), ✓ an interference with 235U direct line 185.7 keV</td>
</tr>
<tr>
<td>Indirect measurement</td>
<td>✓ analyzing Ra-226 by its progenies’ gamma rays using radioactive equilibrium.</td>
<td>✓ longer time needed to achieve secular equilibrium (at least 21 days), ✓ radon leakage from the measurement container causes the equilibrium cannot be reached.</td>
</tr>
</tbody>
</table>
Samples and sealing methods

Table 2. Samples and sealing methods for HPGe and radon chamber measurement

| Soil sample | Uljin soil sample  
|             | Uljin 1: 522.6 gram (Radon Chamber)  
|             | Uljin 2: 512.4 gram (HPGe)  
| Reference material | IAEA 434 Phosphogypsum, 250 gram  
| Marinelli beaker | Polypropylene  
|                 | Snap-on lid with inner lid  
|                 | Vol: 450 mL  
| Sealing method  | MB0: open  
|                 | MB1: only lid without sealing  
|                 | MB2: lid and sealed with paraffin film  
|                 | MB3: sealed with vacuumed plastic bag  

Figure 1. Marinelli beaker sealing method (a) lid without sealing, (b) lid and paraffin film sealing, (c) vacuumed plastic bag sealing
Materials and Methods

HPGe measurement system

Detector specification
- ORTEC GEM 15P4 coaxial HPGe
- 70mm diameter endcap: equipped with 16384 channels MCA
- 0.82 keV for the 122 keV-peak resolution and is 15% for the 1.33 MeV Co-60 peak relative efficiency.

Energy and efficiency calibration
Using CRM volume source in 450mL Marinelli Beaker of agar medium, certified by KRISS, consists of $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{139}$Ce, $^{51}$Cr, $^{113}$Sn, $^{85}$Sr, $^{137}$Cs, $^{60}$Co, and $^{88}$Y with nominal density of 1.001 g/cm$^3$.

Measurement set up
- Samples’ gamma ray spectra were taken by HPGe detector for each of 86400 seconds during 21 days.

Figure 2. Experimental set up for radon leakage measurement.
Materials and Methods

Radon chamber measurement system

- **Radon accumulation chamber**
  - Acrylic material
  - Dimension = 30 cm X 30 cm X 30 cm
  - Sealed with rubber
  - Effective Volume = 23.41L
  - Averaged BKGRND air radon = 12.38 Bq/m³

- **Chamber tightness test**
  - Measuring decaying radon concentration in the chamber.
  - Radon leakage from chamber was determined by comparing the theoretical decay graph of radon and the measured decay.

- **Measurement set up**
  - Radon leakage from Marinelli Beaker was measured by RAD7 detector each of every hour continuously for 21 days

**Figure 3.** Experimental set up for gamma ray spectrum measurement.
Results and Discussion
HPGe measurement
Figure 4. IAEA-434 reference material gamma spectrum measured by HPGe and analyzed by Aptec program
## Results and Discussion

### HPGe measurement result (IAEA-434)

![Graph](image)

Figure 5. IAEA-434 reference material measurement result (MB3).

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Certified Value [Bq/kg]</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{210}$Pb</td>
<td>680</td>
<td>58</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>780</td>
<td>62</td>
</tr>
<tr>
<td>$^{230}$Th</td>
<td>211</td>
<td>9</td>
</tr>
<tr>
<td>$^{234}$U</td>
<td>120</td>
<td>9</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>120</td>
<td>11</td>
</tr>
</tbody>
</table>

#### Table 2. IAEA-434 reference material description.

Resulted direct measurement value of $^{226}$Ra was not agreed well with certified value of IAEA-434 possibly caused by low efficiency of P-type HPGe for lower gamma energy.

Radon progeny result (indirect measurement) is used for further analysis.

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>24th day measured result</th>
<th>Calculated concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{226}$Ra</td>
<td>$885.5 \pm 48.8$</td>
<td>$776.1 \pm 62$ Bq/kg</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>$722.8 \pm 47.8$</td>
<td>$766.0 \pm 61$ Bq/kg</td>
</tr>
<tr>
<td>$^{214}$Pb</td>
<td>$744.2 \pm 39.1$</td>
<td></td>
</tr>
</tbody>
</table>

2% difference
Results and Discussion

HPGe measurement result (Soil)

Figure 6. $^{214}$Bi and $^{214}$Pb build-up for different sealing methods measured by HPGe.

- **Direct measurement ($186$ keV of $^{226}$Ra)**
  - Results from 3 different sealing methods show similar results with averaged values which were $108.91$, $110.04$ and $106.31$ Bq/kg respectively for MB1, MB2, and MB3.
  - Interference from $185.7$ keV peak of $^{235}$U.

- **Indirect measurement ($^{214}$Bi and $^{214}$Pb peaks)**
  - Results show large discrepancy between $^{226}$Ra and $^{222}$Rn progenies.
  - Radon leakage from beaker so that equilibrium cannot be reached.
  - Radon progenies slightly built-up for MB2 and MB3, while for MB1 did not.

- **Free radon inside leak tight Marinelli beaker (MB3)**

  \[
  C_{MB} = EF \cdot C_{Ra} \cdot \frac{m}{V} \\
  C_{MB} = 0.124 \times 50.12 \frac{Bq}{Kg} \times 1.156 \frac{g}{mL} = 7184Bq/m^3
  \]

Better sealing
Radon Chamber measurement
The radon accumulation chamber has a non negligible radon leakage, therefore the radon buildup must be corrected using the following equation (Scholten et al, 2013):

\[
\dot{C}(t) = C_\infty \times (1 - e^{-\lambda \times (1 + a_v) \times t})
\]

\[
\bar{C} = C_\infty \times (1 + a_v)
\]

Normalized leak rate:

\[
a_v = \frac{\lambda_{eff}}{\lambda_{Rn}} = 1.97
\]
Figure 8. Radon chamber measurement result.

Less sealing
More leakage

\[
C_{\infty} = \text{equilibrium radon concentration in chamber (measured)}
\]
\[
\bar{C} = \text{non leakage radon concentration in chamber (calculated with correction factor of } \lambda_V) \]

Leak-rate corrected radon concentration in radon chamber:

\[
C = \bar{C} \times (1 - e^{-\lambda t})
\]
### Results and Discussion

#### Radon chamber measurement result

Marinelli beaker release fraction

\[ MB_{LR} = \frac{\bar{C}}{C_{MB}} \]

\[ C_{MB} = 7184 \text{Bq/m}^3 \]

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**Table 4. Comparing Marinelli beaker release fraction with 4 measurements**

<table>
<thead>
<tr>
<th>Sample and sealing methods</th>
<th>( \bar{C} , (\text{Bq/m}^3) )</th>
<th>Marinelli beaker Release Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uljin1 - MB0</td>
<td>225</td>
<td>0.031</td>
</tr>
<tr>
<td>Uljin1 - MB1</td>
<td>115</td>
<td>0.016</td>
</tr>
<tr>
<td>Uljin1 - MB2</td>
<td>65</td>
<td>0.009</td>
</tr>
<tr>
<td>Uljin1 - MB3</td>
<td>10 (~Background)</td>
<td>&lt;</td>
</tr>
</tbody>
</table>
Conclusion
1. **Direct measurement** and **indirect measurement** by P-type HPGe shows different concentration between soil and reference material which may caused by:
   - Lower efficiency for low gamma energy measurement (≤ 200 keV)
   - Interference from other $^{235}$U gamma peak to $^{226}$Ra gamma peak
   - Radon leakage from Marinelli beaker that inhibit secular equilibrium between Radium and Radon progenies

2. IAEA-434 indirect measurement results using MB3 sealing show very low difference with certified value (± 2 %) therefore **MB3 can be considered as radon leak tight**.

3. The $^{214}$Pb concentrations in soil obtained by HPGe indirect measurement were **MB3 > MB2 > MB1**. This result was verified by radon chamber measurement which showed accumulated radon concentration (leaked radon) **MB3 < MB2 < MB1**.

4. **Radon release fraction (MB$_{LR}$)** were determined as MB3 > MB2 > MB1. For low $^{226}$Ra activity samples the three sealing methods have low radon leakage rate (≤ 5 %). Further study will be needed for relatively high $^{226}$Ra activity samples.

5. The results show that simple and cheap sealing method using **vacuumed plastic bag can effectively minimize the leakage**.


Thank You

Q & A