Practical Self-absorption Correction Method for Various Environmental Samples in a 1 000 cm³ Marinelli Container to Perform Accurate Radioactivity Determination with HPGe Detectors

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The self-absorption of large volume samples is an important issue in gamma-ray spectrometry using high purity germanium (HPGe) detectors. After the Fukushima Daiichi Nuclear Power Plant accident, a large number of radioactivity measurements of various environmental samples have been performed using 1 000 cm³ containers. This study uses Monte Carlo simulations and a semiempirical function to address the self-absorption correction factor for the samples in the 1 000 cm³ Marinelli container that has been widely marketed after the accident. The presented factor was validated by experiments using test sources and was shown to be accurate for a wide range of linear attenuation coefficients $\mu$ (0.05 - 1.0 cm$^{-1}$). This suggests that the proposed correction factor is applicable to almost all environmental samples. In addition, an interlaboratory comparison where participants were asked to determine the radioactivity of a certified reference material demonstrated that the proposed correction factor can be used with HPGe detectors of different crystal sizes.

Key Words: HPGe detector, self-absorption, gamma-ray spectrometry, Marinelli container

1. Introduction

Radioactivity measurements of various environmental samples have been widely performed in many institutes. In these measurements, high purity germanium (HPGe) detectors have been commonly used. The low detection limit is the most significant advantage of this method.

A 100-cm³ cylindrical container ¹ has been widely used in daily monitoring activities to achieve overall high efficiency. To enable a precise determination even for low activity, the sample volume is reduced and the sample is placed into the container.

Conversely, rapid measurement must be performed immediately in emergency situations, without any time-consuming pretreatment (e.g., ashing). In this case, the sample volume should be large to maximize the peak count. In many such cases, re-entrant (Marinelli) containers are used to maximize the counting
efficiency.

In Japan, 2 000 and 700 cm$^3$ Marinelli containers have been used for rapid activity determination with the HPGe detectors. Conversely, 1 000 cm$^3$ Marinelli containers have been preferred for use in food inspection laboratories such as quarantine stations or co-op laboratories.

In addition, a large number of screening devices with NaI(Tl) detectors have been newly installed in Japan for extensive inspection of food products after the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. Around one-third of these devices use the 1 000 cm$^3$ sample containers and additional inspections using HPGe detectors are requested for the samples exceeding screening levels. Thus, the use of 1 000 cm$^3$ containers in radiation activity measurements has increased significantly after the accident.

For accurate radioactivity determinations, efficiency calibration of the detector should ideally be performed under the same conditions as those for the sample. However, preparation of a standard source with the same chemical composition and density as the sample to be measured is difficult. Typically, aluminum oxide or polymer matrices are used in standard commercially available sources; these are often quite different from environmental samples. Numerous studies have been performed for the evaluation of the self-absorption to experimentally and/or theoretically correct this difference.

Guidelines for the use of gamma-ray spectrometry in Japan have also provided a correction method for the 700 and 2 000 cm$^3$ Marinelli containers. The self-absorption correction factor as a function of the linear attenuation coefficient $\mu$ (cm$^{-1}$) is specified in this method. However, the guidelines do not provide any information on the correction for the 1 000 cm$^3$ Marinelli container. To determine the self-absorption correction factors, several standard sources with different matrices are required. However, the use of this process increases the amount of radioactive waste.

In this study, we use Monte Carlo simulations to investigate the self-absorption correction for a range of sample volumes and in particular for the Marinelli containers, which can be easily applied to the method specified in gamma-ray spectrometry guidelines. We determined the self-absorption correction factor for the 1 000 cm$^3$ Marinelli container and validated it experimentally. Furthermore, the accuracy of the proposed correction factor was evaluated by comparing the results obtained by five laboratories with different-sized HPGe detectors for two certified reference material (CRM) samples.

2. A semiempirical self-absorption correction factor for Marinelli containers

The self-absorption correction factor for the sample is given by

$$\varepsilon_s(E, \mu_s) = \frac{f(\mu_s)}{f(\mu_{ref})} \cdot \varepsilon_{ref}(E, \mu_{ref})$$

(1)

where $\varepsilon_s(E, \mu_s)$ is the peak efficiency for the sample with the linear attenuation coefficient $\mu_s$ at a photon energy of $E$, and $\varepsilon_{ref}(E, \mu_{ref})$ is the peak efficiency for a reference source with the linear attenuation coefficient, $\mu_{ref} \cdot f(\mu)$ is the self-absorption correction factor as a function of the linear attenuation coefficient. Note that there is basically no difference in the counting geometry between the sample and the standard source.

Many studies have suggested theoretically...
calculated or empirical formulae for self-absorption correction function of the Marinelli containers. The gamma-ray spectrometry guidelines provide the following formula:

$$f(\mu) = \frac{1}{1 + a \cdot \mu + b \cdot \mu^2}$$  \hspace{1cm} (2)

where \(a\) and \(b\) are constants that depend on the size of the Marinelli container.

This formula has been used in the gamma-ray spectrometry to correct the self-absorption in the containers especially in Japan. However, the accuracy of this formula for a wide range of the linear attenuation coefficients has not been studied. Therefore, experimental confirmation of the accuracy of this formula is another motivation for our study.

The geometric configuration of the 1000 cm\(^3\) Marinelli container examined in this study is shown in Fig. 1.

### 3. Materials and methods

#### 3.1 HPGe spectrometer

An HPGe detector (GEM-30-70, ORTEC, US) installed in a 160-mm thick iron shield was mainly used in this study. Relative efficiency and energy resolution, at the photon energy of 1332 keV obtained from \(^{60}\)Co source, for this detector were 32\% and 1.7 keV, respectively. The diameter and length of the detector crystal were 56.4 mm and 63.7 mm, respectively. Pulse height spectra were obtained through a multichannel analyzer (MCA-7, SEIKO EG&G, JP) with a conversion gain of 4096 channels; the maximum detectable energy was set at 2048 keV (i.e., 0.5 keV per channel). No artificial radionuclide peaks including those originating from the FDNPP accident were observed in a background measurement with a counting time of one million seconds.

#### 3.2 Monte Carlo simulation

Monte Carlo simulations were performed to determine the \(a\) and \(b\) constants in the self-
where ( and ) are the calculated peak efficiencies for SiO₂ with a nonzero and the void ( ) respectively. Note that is one in the case of void (no self-absorption). The two constants in the factor were then determined by the least-squares method based on the calculated values.

The matrix in the 1000-cm³ Marinelli container was the source region, which was virtually set as SiO₂ for Monte Carlo simulation. Since SiO₂ has been commonly used as an alternative material to an environmental sample, the elemental weight fraction is unknown in the simulation. Photons were uniformly generated within the source cell and each full energy deposition event in the active volume of the crystal was scored using a pulse height tally (deposited energy spectrum in the detector crystal). Peak efficiencies were then determined from the resulting pulse height spectra. The simulations were performed individually for each peak energy from the reference source, with the density of the source region changing from 0 (void) to 4 g cm⁻³ in 1 g cm⁻³ steps. One million histories were performed in each simulation so that the statistical standard deviation (1σ) of the calculated peak efficiency was mostly less than 1%.

Equation (1) can be modified as follows:

\[
\frac{\varepsilon_c(E, \mu_c)}{\varepsilon_{\text{void}}(E, 0)} = \frac{f(\mu_c)}{f(0)} = \frac{1}{1 + a \cdot \mu_c + b \cdot \mu_c^2}
\]  

(3)

where \(\varepsilon_c(E, \mu_c)\) and \(\varepsilon_{\text{void}}(E, 0)\) are the calculated peak efficiencies for SiO₂ with a nonzero \(\mu_c\) and the void \((\mu = 0)\), respectively. Note that \(f(\mu)\) is one in the case of void (no self-absorption). The two constants in the factor were then determined by the least-squares method based on the calculated \(f(\mu_c)\) values.

### 3.3 Sources

Two types of certified sources in the 1000 cm³ Marinelli container were used in this study. The exteriors of these sources are shown in Fig. 3. The first source comprised aluminum oxide powder (as a matrix) with a density of 1 g cm⁻³. This source was used as the reference source described above and was also used to perform efficiency calibration of the HPGe detector. It contained the following nine radionuclides:¹⁰⁶Cd (88 keV),³²Co (122 keV, 136 keV),

![Fig. 3 Exteriors of the reference source (left) and a set of the test sources (right).](image-url)
Table 1 Composition ratio for each source (>1%)

<table>
<thead>
<tr>
<th>Source #1</th>
<th>Source #2</th>
<th>Source #3</th>
<th>Source #4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g cm(^{-3}))</td>
<td>0.7</td>
<td>1.0</td>
<td>1.6</td>
</tr>
<tr>
<td>Composition</td>
<td>H: 9.7%</td>
<td>H: 9.4%</td>
<td>H: 3.6%</td>
</tr>
<tr>
<td></td>
<td>C: 49.4%</td>
<td>C: 46.7%</td>
<td>C: 18.2%</td>
</tr>
<tr>
<td></td>
<td>O: 16.1%</td>
<td>O: 18.3%</td>
<td>O: 34.0%</td>
</tr>
<tr>
<td></td>
<td>Si: 24.8%</td>
<td>Si: 25.6%</td>
<td>Si: 10.5%</td>
</tr>
<tr>
<td></td>
<td>Na: 19.8%</td>
<td>Ca: 12.9%</td>
<td></td>
</tr>
<tr>
<td></td>
<td>S: 13.9%</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^{137}\)Cs (168 keV), \(^{51}\)Cr (320 keV), \(^{85}\)Sr (514 keV), \(^{137}\)Cs (662 keV), \(^{54}\)Mn (836 keV), \(^{48}\)Y (898 keV, 1 836 keV), and \(^{60}\)Co (1 173 keV, 1 332 keV). We also used a set of four sources comprising different matrices with densities ranging from 0.7 to 3.0 g cm\(^{-3}\). The density and elemental weight fraction of the matrices used in these four sources (Source #1 - #4) are given in Table 1. This set was used as test sources to validate the constants determined in this study. In these sources, \(^{54}\)Mn (836 keV) was replaced by \(^{113}\)Sn (392 keV), whereas the other radionuclides were the same as those in the reference source. As a result, this set exhibited a wide range of linear attenuation coefficient values from 0.0357 (for 1 836 keV for Source #1) to 0.929 (for 88 keV for Source #4). Linear attenuation coefficients of materials were calculated using the XCOM\(^{16}\) database and the reference data of the density and elemental weight fraction. The total radioactivity was 70 kBq for the reference source and 60 kBq for each of the test sources; the uncertainties for each radionuclide in the certified sources were in the 3% - 5.3% range (coverage factor \(k = 2\)).

3.4 Interlaboratory comparison

We performed an interlaboratory comparison to evaluate the validity of the proposed self-absorption correction factor by providing two identical test samples made of brown rice that was prepared as a CRM related to the FDNPP accident\(^{17}\). The brown rice with certified radioactivities of \(^{134}\)Cs and \(^{137}\)Cs was placed in two 1 000 cm\(^3\) Marinelli containers.

Five laboratories participated in this test. Table 2 listed the relative efficiency and crystal size of 11 different HPGe detectors owned by these laboratories. Participants were requested to perform radioactivity evaluations using the self-absorption correction factors determined in this study.

4. Results and discussions

4.1 Self-absorption correction factor

Fig. 4 shows the self-absorption correction factor for the 1 000 cm\(^3\) Marinelli container as a function of the linear attenuation coefficient. Each point in the figure corresponds to the \(f (\mu_c)\) value calculated by Monte Carlo simulation for the particular value of the linear attenuation coefficient. As a result, the constants \(a\) and \(b\) were determined to be 1.362 and 0.6774, respectively. As shown in the figure, the calculated \(f (\mu_c)\) values were fit well by the semiempirical formula.

Fig. 5 illustrates two peak efficiency functions for the HPGe detector. The first function
Table 2  Relative efficiency and crystal size of 11 different HPGe detectors owned by five laboratories

<table>
<thead>
<tr>
<th>Relative efficiency (%)</th>
<th>Crystal size</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Diameter (mm)</td>
</tr>
<tr>
<td>20</td>
<td>53.0</td>
</tr>
<tr>
<td>20</td>
<td>59.0</td>
</tr>
<tr>
<td>25</td>
<td>59.0</td>
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<tr>
<td>30</td>
<td>56.0</td>
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<tr>
<td>30</td>
<td>59.0</td>
</tr>
<tr>
<td>35</td>
<td>58.9</td>
</tr>
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<td>61.1</td>
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<td>40</td>
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<td>40</td>
<td>62.0</td>
</tr>
<tr>
<td>45</td>
<td>62.9</td>
</tr>
<tr>
<td>45</td>
<td>63.0</td>
</tr>
</tbody>
</table>

Fig. 4  Self-absorption correction factor for a 1 000 cm$^3$ Marinelli container.

Fig. 5  Peak efficiencies as a function of photon energy for the reference source and void.

was fitted to the measured peak efficiencies of the reference source. The second function was a fit for the case of no material (void) in the 1 000 cm$^3$ Marinelli container (i.e., no self-absorption). This function was obtained on the basis of the same peak efficiencies multiplied by $1/f(\mu_{\text{eff}})$; the fit method was the same as that used for the function for the reference source. The function for the void was used to determine the counting efficiency for the sample together with the self-absorption correction factor for a corresponding linear attenuation coefficient of the sample.

In addition, the constants for 700 and 2 000 cm$^3$ Marinelli containers (provided by the guidelines) were also re-evaluated by the same method as that used for 1 000 cm$^3$ Marinelli container. The results are summarized in Table 3. It was found that both the constants obtained in this study and the original values can be used for the samples with almost the same density as that of the standard source (e.g.,
\(^{137}\text{Cs}\) activity determination in a water, soil, and food sample).

Conversely, a considerable difference was obtained for samples that are either heavier or lighter than the standard source. For the lighter sample with the density of 0.2 g cm\(^{-3}\), such as hay, differences of 6.6% and 2.8% at 662 keV were found for the 700 and the 2,000 cm\(^3\) containers, respectively. Similarly, for the 2 g cm\(^{-3}\) dense soil sample, difference of 2.7% and 3.0% were estimated at 662 keV for the 700 and the 2,000 cm\(^3\) containers, respectively. In this estimation, activities were compared with the corrections for a standard source (\(\text{Al}_2\text{O}_3\), 1 g cm\(^{-3}\)) and these virtual samples (\(\text{SiO}_2\), 0.2 g cm\(^{-3}\) and 2 g cm\(^{-3}\)). These corrections were performed using both original constants and newly obtained constants.

These results indicate that the correction using the coefficients described in the guideline may be biased unless the linear attenuation coefficients of the sample are almost the same as that of the standard source. Therefore, a further evaluation of the self-absorption correction needs to be undertaken over a wide range of \(\mu\) for the 700 and 2,000 cm\(^3\) Marinelli containers.

### Table 3 Coefficients in the self-absorption correction factor for 700, 1,000, and 2,000 cm\(^3\) Marinelli containers with original values described in the guidelines

<table>
<thead>
<tr>
<th>Volume of Marinelli container (cm(^3))</th>
<th>Coefficient values</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(a) (cm)</td>
</tr>
<tr>
<td>700</td>
<td>this work: 1.09440</td>
</tr>
<tr>
<td></td>
<td>original: -0.558435</td>
</tr>
<tr>
<td>1000</td>
<td>this work: 1.36167</td>
</tr>
<tr>
<td></td>
<td>original: -</td>
</tr>
<tr>
<td>2000</td>
<td>this work: 2.23985</td>
</tr>
<tr>
<td></td>
<td>original: 1.71115</td>
</tr>
</tbody>
</table>

Note: Significant digits of the coefficients obtained in this work are underlined.

#### 4.2 Quantified radioactivity

Fig. 6 demonstrates the ratios of the quantified radioactivity to the certified radioactivity for each peak energy from the test sources: panels (A) and (B) are the ratios without and with the self-absorption correction, respectively. The error bars of the plots in the figure denote the combined uncertainty \((k = 2)\) of the standard sources (for calibration and quantification) and counting statistics. Fig. 6 shows that the self-absorption correction factor enables accurate radioactivity determination of the test samples over a wide range of the linear attenuation coefficient \((\mu : 0.05 - 1.0\text{ cm}^{-1})\), whereas the absence of corrections resulted in significant underestimations for \(\mu > 0.2\text{ cm}^{-1}\).

All of the corrected radioactivity values agreed with the certified values within the combined uncertainty. This suggests that the proposed correction factor is versatile and is applicable for almost all cases of environmental sample radioactivity determination.

#### 4.3 Interlaboratory comparison

Fig. 7 shows reported values from the participants in comparison to the certified radioactivities \((^{134}\text{Cs} \text{ and } ^{137}\text{Cs})\) of the test samples. All reported values excluding those obtained by
Fig. 6  Ratios of determined to certified radioactivities for the sources used as test samples.
Panel (A): no correction. Panel (B): correction.

Fig. 7  Reported values from participants in comparison to certified values of the test samples.
Gray zone corresponds to the uncertainty (k = 2) of the certified activity.
detector #4 (a resolution problem was reported) were found to be within the combined uncertainty ($k = 2$) of the CRM denoted by the bands in the figure.

The detector crystals diameters used in this comparison ranged from 50 to 70 mm, and the crystal length ranged from 30 to 80 mm. All housing sizes of the detectors with these crystals were suitable for the re-entrant space of the Marinelli container. In conclusion, the proposed method can be applied for accurate environmental sample radioactivity measurements in the 1 000 cm$^3$ Marinelli container that has been widely used after the FDNPP accident.

5. Summary

The self-absorption correction in large volume samples is an important issue in gamma-ray spectrometry. This study attempted to determine the two coefficients of a semiempirical formula for the self-absorption correction factor used in radioactivity measurements for environmental samples in the 1 000 cm$^3$ Marinelli container with HPGe detectors. Monte Carlo simulations were performed for this task. It was shown that the obtained factor can be used for a wide range of linear attenuation coefficients (0.05 – 1.0 cm$^{-1}$), suggesting that it is valid for almost all environmental samples. The factor obtained in this method was also validated by comparison of test sample radioactivity measurement results obtained by several laboratories with HPGe detectors of different crystal sizes.

Acknowledgments

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References

1) Yamada, T. and Nakamura, Y., Examination of the U8 type polypropylene container used for radioactivity standard volume sources, *Radioisotopes*, 54, 105-110 (2005)
3) Certification report of soil reference material for radioactivity analysis; JSAC 0471, 0472 and 0473, JSAC 2012

11) JP MEXT, 1992, Gamma Ray Spectrometry Using a Germanium Semiconductor Detector (Radiation Measurement Method Series #7), Ministry of Education, Culture, Sports, science and Technology in Japan

12) JP MEXT, 2004, Gamma Ray Spectrometry for an Accident (Radiation Measurement Method Series #29), Ministry of Education, Culture, Sports, science and Technology in Japan


要  旨

HPGe 検出器を用いる 1 000 cm$^3$ マリネリビーカーに充填された
環境試料定量のための実用的自己吸収補正手法

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高純度ゲルマニウム半導体検出器（HPGe 検出器）を用いた、大体積の試料測定では自己吸収の影響が大きい。東京電力福島第一原子力発電所事故以降、充填体積で 1 000 cm$^3$ の試料容器による測定が増加した。本研究では、事故後急速に流通の拡大した充填体積が 1 000 cm$^3$ のマリネリ容器に対する自己吸収補正関数をモンテカルロ・シミュレーション及び半経験式を用いて決定した。得られた自己吸収補正関数は、広範囲な線減衰係数 μ (0.05 ～ 1.0 cm$^{-1}$ ) を持つ標準線源を用いて評価された。その結果、多様な環境試料に対する補正が実施可能であることが示された。併せて、認証標準物質を当該容器に充填した試料を用いた相互比較試験を実施し、様々な結晶寸法を有する検出器に対しても自己吸収補正関数の妥当性を示した。