Development of a self-absorption correction method used for a HPGe detector by means of a Monte Carlo simulation

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Quantitative analysis for food products and natural samples, to determine the activity of each radionuclide, can be made by using a high-purity germanium (HPGe) gamma-ray spectrometer system. The analysis procedure is, in general, based upon the guidelines established by the Nuclear Safety Division of the Ministry of Education, Culture, Sports, Science and Technology in Japan (JP MEXT). In the case of gamma-ray spectrum analysis for large volume samples, re-entrant (marinelli) containers are commonly used. The effect of photon attenuation in a large-volume sample, so-called "self-absorption", should be corrected for precise determination of the activity. As for marinelli containers, two accurate geometries are shown in the JP MEXT guidelines for 700 milliliter and 2 liter volumes. In the document, the functions to obtain the self-absorption coefficients for these specific shapes are also shown. Therefore, self-absorption corrections have been carried out only for these two containers with practical media. However, to measure radioactivity for samples in containers of volumes other than those described in the guidelines, the self-absorption correction functions must be obtained by measuring at least two standard multinuclide volume sources, which consist of different media or different linear attenuation coefficients. In this work, we developed a method to obtain these functions over a wide range of linear attenuation coefficients for self-absorption in various shapes of marinelli containers using a Monte Carlo simulation. This method was applied to a 1-liter marinelli container, which is widely used for the above quantitative analysis, although its self-absorption correction function has not yet been established. The validity of this method was experimentally checked through an analysis of natural samples with known activity levels.

Key Words : *HPGe detector, self-absorption, marinelli, Monte Carlo simulation, peak efficiency*

1. INTRODUCTION

Various radioactive nuclides, originating not only from natural sources, but also from human activity including nuclear weapons testing or nuclear facility accidents, may be found in food or environmental samples. Environmental trends in artificial radioactive nuclides have been continuously monitored. For studies of

nuclides which emit gamma-rays, a high-purity germanium (HPGe) detector is the most suitable instrument, and is used for quantitative analysis in many laboratories.

At the Fukushima Daiichi Nuclear Power Plant accident, a variety of radioactive nuclides were emitted, the most prevalent being cesium-137. Consequently, cesium-137 and other radioactive nuclides were detected in agricultural and marine products harvested from the Kanto and Tohoku areas. As a result, interest in detection of radioactive nuclides in such products has grown significantly among citizens.

For this reason, a screening level and a guideline for screening have been announced officially by the Ministry of Health, Labour and Welfare (JP MHLW). For these screenings, a sodium iodide scintillation detector is the most appropriate instrument because it can be used with a concise and simple procedure. Fourteen types of Sodium Iodide scintillation detectors are commercially available (as of October 2012).

However, different shapes and volumes of the sample container are used for each of these scintillation detectors. While there are a variety of 500 milliliter to 2-liter containers, the marinelli container is the most commonly used.

In the case that the sodium iodide detector yielded a result above the screening level, additional analysis would be done by HPGe spectrometer.

Guidelines for a quantitative radioactive analysis using HPGe detector[1] [2] have been established by JP MEXT. Almost all spectroscopy systems that are commercially available in Japan are designed based on these guidelines. In addition, preliminary treatment and measurement should meet these guidelines.

In order to perform an accurate analysis using a HPGe detector, an accurate peak efficiency should be determined in advance. However, even when using the same detector with the same container, the peak efficiency may be different if the sample density or medium are different, because gamma-rays emitted from a certain part of the sample are absorbed in the sample itself (self-absorption). The effect of self-absorption depends on the energy of the gamma-ray. Therefore, in order to determine the activity of a sample, obtaining an accurate self-absorption correction factor is required for each unique sample condition.

The JP MEXT guidelines illustrate the self-absorption

correction method by using functions for cylindrical containers of 100 milliliters, and marinelli containers of 0.7 and 2 liters with specific shapes. As specific drawings of these shapes are indicated in the guideline document, additional measurements after a screening, should be done using these specific containers.

Thus, to avoid the complications involved in making additional measurements with different containers re-filled, any quantitative analysis should be done using containers where the self-absorption correction function is well understood. However, a self-absorption correction calculated using a Monte Carlo photon transport code[3], for a sample which has a specific matrix and density, has been widely used as an alternative method for deriving the correction function. Although correction using the Monte Carlo method features flexibility with regard to shape and matrix, it is a computationally expensive method (may take minutes to hours depending on the energies and sample condition). Thus, supposing samples have different matrices or different densities, such as food and an environmental sample, running a Monte Carlo simulation for each sample is inadequate and instead deriving a rapid correction using a function (such as those mentioned above) is preferable.

In many cases, the samples prepared for a measurement are 1 or 2 liters in volume. If the sample volume is 2 liters, then the 2-liter marinelli container, which is indicated in the guideline, would be used. Otherwise, such as in the case of a high-priced sample, only a 1-liter sample would be prepared. In this case, a 1-liter marinelli container is the most suitable to measure this sample effectively. The 1-liter marinelli container is widely available in significant quantities unlike the 0.7-liter marinelli.

The self-absorption method for the 1-liter marinelli container is not shown in the JP MEXT guidelines, so most all analysts determine the sample activity without this correction. In order to obtain a self-absorption correction function for a certain container, it is absolutely necessary to measure different standard radioactive volume sources in different media. A correction function derived in this way has not been established for the 1-liter marinelli container.

In this work, we developed a method to obtain self-absorption correction functions for various volume samples, and in particular, for marinelli containers. Furthermore, we derived a self-absorption function for

the widely used 1-liter marinelli container, whose self-absorption correction function has not yet been established, and evaluated the derived function using an actual food sample.

2. CALCULATION

To perform the Monte Carlo simulation, a model, which consists of a HPGe detector and a sample in a typical 1-liter marinelli container, was prepared. The density of the sample was varied step by step to give different linear attenuation coefficients. Peak efficiencies for the HPGe detector which were used in this experiment were obtained for different attenuation coefficients.

MCNP4C2[4] was used for the Monte Carlo photon transport simulation. The cross-sectional geometry of a 1-liter marinelli container is shown in Fig. 1.
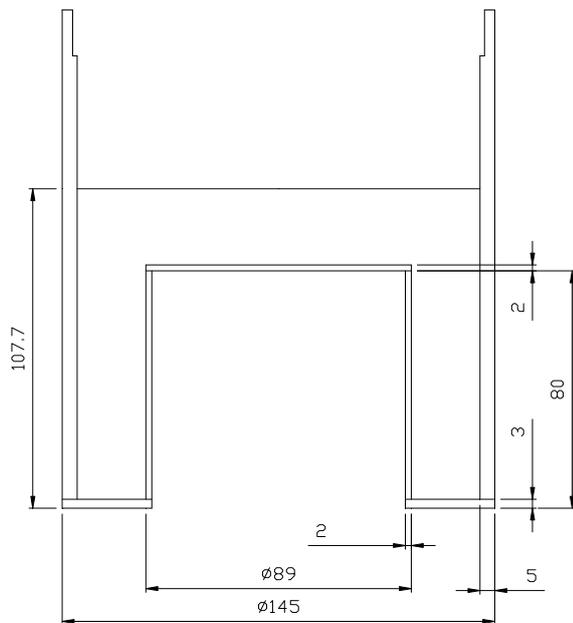

Fig. 1 Cross-sectional dimensions of a 1-liter marinelli container

The equation relating a standard source s and a sample x is written as equation (1) below. In terms of $f(\mu)$, this is defined in the semi-empirical equation (2), where a and b are constants, which depend on the shape of the container. These constants are derived from the peak efficiencies using more than two standard volume sources consisting of different matrices, for which the compositions are known[1,2].

In this work, silicon dioxide was the material used and

densities were varied up to 4 g/cm^3. Photon energies were 88keV to 1836keV corresponding to Cd-109, Co-57, Cr-51, Ce-139, Sr-85, Mn-54, Cs-137, Co-60 and Y-88.

Linear attenuation coefficients were calculated using an approximation[5] that was determined by the composition and photon energies. The linear attenuation coefficient given by this approximation was compared with the XCOM[6] result. The values were found to be in good agreement with a difference of less than one percent.

$$\varepsilon_x(E, \mu_x) = \frac{f(\mu_x)}{f(\mu_s)} \varepsilon_s(E, \mu_s) \qquad (1)$$

$$f(\mu) = \left(1 + a\mu + b\mu^2\right)^{-1} \qquad (2)$$

The constants, a and b for the 1-liter marinelli container were derived using a least-squares fitting method with μ and $f(\mu)$.

3. EXPERIMENT

In order to validate the self-absorption correction function , a mock experiment representing the quantitative analysis was made. For this purpose, a raw rice sample with known activity was used. The detector used was a p-type HPGe detector, ORTEC GEM30-70, which has a relative efficiency of roughly 30%. The crystal size of the HPGe detector was 56.4mm in diameter and 63.7mm in length. The multi-channel analyzer (MCA7600, SEIKO EG&G Co., Ltd.) included a digital PHA module and the conversion gain was set to 4096 channels for 2048keV. The detector was installed into an iron shield with a thickness of 160mm.

Marinelli containers for the sample and the efficiency calibration were made of acryl plate and acryl pipe which were bonded to each other. The dimensions of the marinelli containers were strictly controlled. To control the shape of marinelli container, all dimensions were measured to within a tolerance of less than one millimeter. A 1-liter marinelli container filled with raw rice is shown in Fig.2.

An efficiency calibration was performed by using the standard source (MX033MR; JRIA) consisting of a 1-liter marinelli container with active alumina powders. The density of the alumina was roughly 1g/cm^3 and 9 of the radioactive nuclides mentioned above were added.

The range of linear attenuation coefficients for the

alumina is from 0.046 cm^{-1} to 0.16 cm^{-1}, corresponding to photon energies of 88keV to 1836keV. After the efficiency calibration, measurements of raw rice were performed. The density of the raw rice in the container was 0.862 g/cm^3. This corresponds to a linear attenuation coefficient of 0.082 cm^{-1} at 662keV. The raw rice consisted of both contaminated rice and non-contaminated rice. The total volume of the raw rice was adjusted to be 2000cm^3.

As a check for homogeneity, and to verify radioactive cesium concentration, four 50-gram samples were taken from the raw rice and put into a cylindrical polypropylene pot (Model: 3-100) with a volume of 100ml, in which further quantitative analysis was performed. The 3-100 container filled with raw rice is shown in Fig. 3a.

The cesium-137 concentration was determined by calculating an average activity of these four samples. To confirm the sample homogeneity, the cesium-137 activities were compared among these four samples, where no significant disagreement was found to exceed the measurement uncertainty.

In order to verify the results for the 3-100 container, measurement of a reference material[7)8)] shown in Fig. 3b was also done. There was no significant disagreement between the certificate[7)] and the analyzed result .

The sample from the 1-liter marinelli container was pre-verified with a 3-100 container. Quantitative analysis was done with a self-absorption coefficient that was calculated using the method developed in this work.

4. RESULTS AND DISCUSSION

The Monte Carlo simulation was performed in the combined geometry of a 1-liter marinelli container and a HPGe detector. The calculated self-absorption coefficients corresponding to each linear attenuation coefficient are shown in Fig. 4. The calculated self-absorption function for the detector and this marinelli container are shown in equation (3). The coefficients, a and b, of the function were 1.378 and 0.6867, respectively.

$$f(\mu) = \left(1 + 1.378\mu + 0.6867\mu^2\right)^{-1} \qquad (3)$$
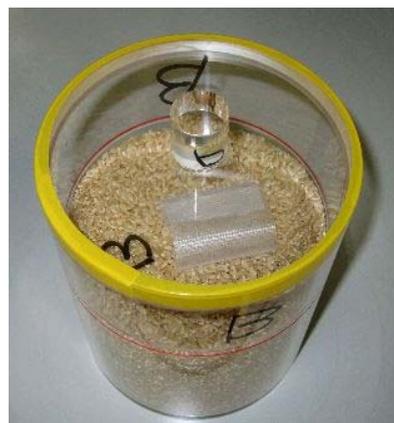

Fig. 2 A sample in a 1L Marinelli container

Fig. 3a A sample in a 3-100 container

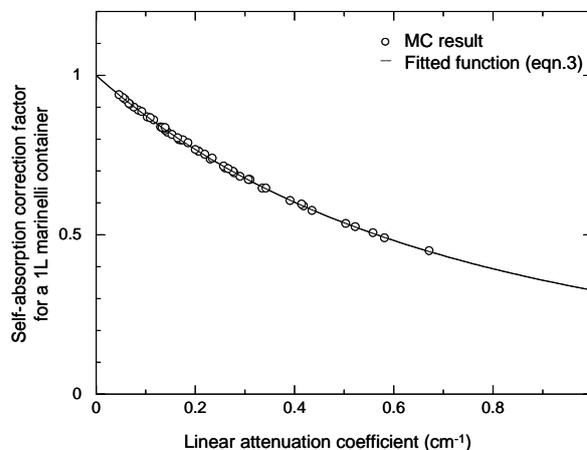Fig. 3b A reference material provided by JSAC

Fig. 4 Self-absorption correction function for a 1L marinelli container

Given the results of the cesium-137 quantitative analysis of the raw rice using a 3-100 container, the cesium-137 concentration was determined to be 102.1 ± 5.2 Bq/kg (95% confidence limit). This sample was put into a 1-liter marinelli container. Quantitative analysis was done by using the measured efficiency for 662keV including the self-attenuation coefficients of both the alumina and the rice sample. As a result of this analysis, the cesium-137 concentration was found to be 103.5 ± 5.9

Bq/kg, in good agreement with the concentration found in the pre-verification of the raw rice sample discussed above.

In this work, we demonstrated the validity of the function derived from a Monte Carlo simulation for a raw rice sample with a limited linear attenuation coefficient of 0.082 cm^{-1}.

However, for the actual measurement of food or environmental samples using a HPGe detector it is expected that for high density samples, such as soil or sludge where the density significantly exceeds 1g/cm^3, a larger linear attenuation coefficient will be required. Thus, it will be necessary to evaluate this method over a wide range of linear attenuation coefficients.

Furthermore, the validity of the self-absorption correction function was shown only for the specific detector used in this work. However, there are different shapes of the detector crystal for different detectors distributed by manufacturers. The applicability of the function derived in this work should be verified for different detector crystal shapes.

5. CONCLUSION

For the first time, a method was developed in order to derive a function for rapid self-absorption correction of versatile marinelli containers by means of a Monte Carlo simulation.

Quantitative analysis of a raw rice sample was done using a 1-liter marinelli container, which is frequently and widely used, but for which the correction for self-absorption has not been determined. For this process, the peak efficiency was used, which was corrected by the self-absorption coefficient with the present method.

The cesium-137 concentration of a raw rice sample was obtained and compared with the pre-verified results. Both results were in good agreement to within their uncertainties. Thus, the present method was verified as a reasonable means of determining the self-absorption correction.

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