Measurement of efficiency calibration and coincidence correction factor for well-type HPGe detector

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Abstract

To calculate the detection efficiency curve and coincidence correction factors for a well-type HPGe detector, the detection efficiency of the HPGe was calibrated using commonly employed single-energy γ radionuclide standard sources, while cascade coincidence correction factors for common radionuclides such as 60Co, 88Y, 131I, 134Cs, 152Eu, and 133Ba were measured. The measurement results indicate that the efficiency value obtained using 139Ce from the commonly used single-energy γ radionuclide standard sources can differ from the fitted value by up to 49%; the coincidence effect of the well-type HPGe significantly influences activity measurement results, with smaller sample heights yielding larger coincidence correction factors. When the sample height is 0.5 cm, the coincidence correction factor for the 563.23 keV γ-ray full-energy peak emitted by 134Cs reaches 8.00, while that for the 604.70 keV γ-ray full-energy peak is 3.01. When the sample activity exceeds approximately 1000 Bq, the contribution of random coincidence to the total coincidence correction factor gradually increases; for instance, when the activity of 131I increases from 1300 Bq to 6300 Bq, the coincidence correction factor for the 80.19 keV γ-ray full-energy peak increases by 10%. The measurement data can provide reference for the selection of efficiency calibration sources for well-type HPGe detectors and the correction of radionuclide activity measurement results.

Full Text

Preamble

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Efficiency Calibration and Coincidence Correction Factor Measurements for Well-Type HPGe

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Abstract

[**Background**]: Well-type HPGe detectors offer high detection efficiency and are commonly used for measuring small-volume samples. While summation correction is necessary in such measurements, relevant data for this detector type is rarely reported in the literature.

[**Purpose**]: To calculate detection efficiency curves and coincidence correction factors for well-type HPGe detectors.

[**Methods**]: The detection efficiency of HPGe was calibrated using commonly available monoenergetic γ-ray standard sources, and cascade coincidence correction factors were measured for common radionuclides including ^{60}Co, ^{88}Y, ^{131}I, ^{134}Cs, ^{152}Eu, and ^{133}Ba.

[**Results**]: Measurements revealed that the efficiency value obtained using ^{139}Ce from standard monoenergetic γ sources deviated from fitted values by up to 49%. The coincidence effect in well-type HPGe significantly impacts activity measurements, with smaller sample heights yielding larger correction factors. At a sample height of 0.5 cm, the coincidence correction factor for the 563.23 keV γ-ray full-energy peak from ^{134}Cs reached 8.00, while that for the 604.70 keV peak was 3.01. When sample activity exceeds approximately 1000 Bq, the contribution of random coincidence to the total correction factor increases progressively. For ^{131}I, increasing the activity from 1300 Bq to 6300 Bq raised the correction factor for the 80.19 keV γ-ray full-energy peak by 10%.

Conclusions: These measurement data provide valuable reference for selecting appropriate calibration sources for well-type HPGe efficiency calibration and for correcting radionuclide activity measurements.

Keywords: Well-type HPGe; Coincidence correction; ^{139}Ce; ^{134}Cs; ^{131}I

Introduction

HPGe detectors are routinely used in laboratories to measure the activity of γ-emitting radionuclides in samples. Well-type HPGe detectors, characterized by their well-shaped crystal geometry, offer large solid angles and high detection efficiency, making them particularly suitable for small sample quantities. For instance, environmental samples such as fallout ash or biological ash typically

weigh only a few grams and are best measured by placing them inside the detector well to achieve higher detection efficiency. However, the high efficiency of well-type HPGe also increases the probability of cascade coincidence effects, which can compromise measurement accuracy. Due to the limited time resolution of γ-spectrometry systems, two or more photons emitted in cascade by a nuclide cannot be distinguished and are recorded as a single event, causing the count rate in the full-energy peak of interest to decrease or increase [1,2]. Cascade coincidence effects can occur between γ-rays or between γ-rays and X-rays [3,4].

The standard method for obtaining cascade coincidence correction factors involves calibrating HPGe detection efficiency using a series of monoenergetic γ-ray standard sources [5-7]. However, the suitability of common monoenergetic γ sources for well-type HPGe and detectors with large sensitive areas requires investigation. Accurate radionuclide activity measurements necessitate corrections for potential cascade coincidence effects. While numerous studies have investigated coincidence corrections for top-mounted HPGe detectors (where samples are placed on the detector crystal surface) [8-11], few have reported measurement data for well-type HPGe. O. Sima et al. [12] used Monte Carlo methods to calculate coincidence correction factors for ^{134}Cs and ^{60}Co in well-type HPGe and validated the results using ^{210}Pb, ^{57}Co, ^{51}Cr, ^{7}Be, ^{137}Cs, ^{54}Mn, ^{65}Zn, and ^{40}K standard sources. Wang Shilian et al. [13] calculated coincidence correction factors for these nuclides in well-type HPGe based on their decay schemes. Neither study considered the influence of X-rays emitted by standard sources, sample geometry characteristics, or random coincidence from high-activity samples on the correction factors.

This study investigates the applicability of common monoenergetic γ standard sources for efficiency calibration of well-type and large-area HPGe detectors. Using a well-type HPGe detector, we measured cascade coincidence correction factors for common environmental radionuclides and examined their relationship with sample height. We also investigated the relationship between activity and random coincidence for certain nuclides. These findings provide valuable reference for accurate radionuclide activity measurements using large-solid-angle detectors such as well-type HPGe.

1. Methods

Two primary methods are commonly employed for coincidence correction: the standard source method and the distance method [14].

1.1 Standard Source Method

This method uses a set of monoenergetic γ-ray standard sources to calibrate the full-energy peak detection efficiency of HPGe, yielding an efficiency curve (E) as a function of energy E without coincidence effects. Under the premise that the sample matrix, geometry, and mass parameters are identical to those

of the monoenergetic standard sources, the full-energy peak detection efficiency $_0(E_0)$ at the energy of interest is obtained using a standard source of the nuclide to be measured. The coincidence correction factor F for the count rate of the full-energy peak of γ-rays with energy E_0 emitted by the target nuclide is given by:

$F = (E_0) / {}_0(E_0)$

The standard source method requires a standard source of the target nuclide with known activity, and demands high accuracy in the source activity.

1.2 Distance Method

Monoenergetic γ standard sources and the sample to be measured, having identical matrix, geometry, and mass parameters, are measured at two positions: a far position F (where coincidence summing effects are negligible) and a near position N close to the detector. The full-energy peak efficiency curves F(E) and N(E) are obtained for both positions, along with the count rates nF and nN for the full-energy peak of interest at energy E_0. The coincidence correction factor F is then calculated as:

$F = (F(E_0) / N(E_0)) \times (nN / nF)$

The distance method does not require knowledge of the target nuclide's activity, but it is not applicable for samples placed inside the well of a well-type HPGe detector.

2. Experimental Equipment

2.1 γ-Spectrometry Detectors

The well-type γ-spectrometer used in this study was a Canberra GCW5021 HPGe detector with a crystal measuring ø70 mm × 60 mm, energy resolution of 2.0 keV (for ^{60}Co at 1332 keV), well inner diameter of 10 mm, and well depth of 40 mm. Additionally, a BE5030 broad-energy HPGe detector with a large sensitive area was employed, featuring a crystal size of ø81 mm × 31 mm and energy resolution of 1.9 keV (for ^{60}Co at 1332 keV).

2.2 Standard Sources

The monoenergetic γ standard sources used were standard solutions produced by LEA (France), containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn, and ^{65}Zn. The specific activity uncertainty for ^{109}Cd was 5% (k=2, 95% confidence level), while uncertainties for other nuclides were $ $3.5%. The ^{40}K solution was prepared using KCl with an uncertainty of approximately 4.0%. Standard sources of ^{133}Ba, ^{134}Cs, ^{152}Eu, and ^{226}Ra were produced by China Isotope & Radiation Corporation, with specific activity uncertainties of $ $2.9%, 3.5%, 3.2%, and 3.6%, respectively.

3. Results and Discussion

3.1 Efficiency Calibration

Monoenergetic γ standard sources are commonly used to obtain the full-energy peak detection efficiency curve for HPGe. The radionuclides in these sources typically emit only one γ-ray energy and do not exhibit cascade coincidence effects. However, some nuclides emit X-rays in addition to a single γ-ray, and coincidences between γ-rays and X-rays may occur. ^{139}Ce is a frequently used nuclide in standard sources [1-7,11], emitting a 166 keV γ-ray along with possible X-rays at 33 keV and 38 keV [15].

The GCW5021 well-type HPGe detector used in our laboratory has a 1 mm aluminum window and 0.1 mm dead layer thickness, while the BE5030 detector features a 0.6 mm carbon fiber window and 0.01 mm dead layer. Conventional P-type coaxial HPGe detectors typically have ~2 mm aluminum windows and average dead layer thicknesses of ~2 mm. Due to these thicker windows and dead layers, almost all low-energy photons below 40 keV are absorbed before reaching the HPGe crystal, preventing cascade coincidence effects between γ-rays and X-rays and allowing ^{139}Ce to be used for efficiency calibration. In contrast, well-type HPGe detectors have thinner windows and dead layers, maintaining relatively high detection efficiency for X-rays despite some absorption, thus enabling cascade coincidences between γ-rays and X-rays.

To investigate the applicability of ^{139}Ce for well-type and broad-energy HPGe detectors with good low-energy photon response and high efficiency, we prepared aqueous standard source samples of various geometries using the monoenergetic sources described in Section 2.2. The full-energy peak detection efficiencies of GCW5021 (well-type) and BE5030 (broad-energy) HPGe detectors were calibrated. The efficiency calibration results are shown in [Figure 1: see original paper] and [Figure 2: see original paper]. The efficiency curves were fitted using measured full-energy peak efficiencies at various γ-ray energies after excluding the 166 keV data point.

Standard source samples were prepared in commonly used geometries for environmental monitoring: ø50 mm × 2 mm, ø50 mm × 20 mm, ø70 mm × 65 mm, ø75 mm × 10 mm, and ø10 mm × 47 mm. During measurement, these containers were placed on the detector surface for both HPGe types, except for the ø10 mm × 47 mm geometry, which was only used for well-type HPGe measurements inside the detector well.

As shown in [Figure 1: see original paper] and [Figure 2: see original paper], the full-energy peak detection efficiency for both HPGe types initially increases then decreases with photon energy, reaching a maximum around 100 keV. For a given crystal volume, higher-energy photons have greater penetration but lower probability of depositing their entire energy in the crystal, resulting in lower full-energy peak efficiency. The detector housing, window, and dead layer absorb some photons, with lower-energy photons being more susceptible to absorption.

This absorption has limited impact on photons above 100 keV but significantly reduces efficiency for photons below 100 keV. Detection efficiency for a given photon energy depends primarily on the average solid angle subtended by the crystal relative to the sample and on sample self-absorption. Larger sample geometries and greater average distances from the crystal yield lower detection efficiency, which explains why the ø70 mm × 65 mm samples show much lower efficiency than ø50 mm × 2 mm samples.

The results in [Figure 1: see original paper] and [Figure 2: see original paper] reveal that for ø10 mm × 47 mm, ø50 mm × 2 mm, and ø50 mm × 20 mm (BE5030) samples, the full-energy peak efficiency at 166 keV from ^{139}Ce deviates from the fitted curve by 38%, 12%, and 6%, respectively. Potential causes include inaccurate activity values in the standard source or fitting deviations. However, for ø50 mm × 2 mm, ø50 mm × 20 mm (GCW5021), ø70 mm × 65 mm (BE5030), and ø75 mm × 10 mm geometries, the deviation is $ $2%, confirming the reliability of the ^{139}Ce source activity.

To verify the consistency between measured efficiency and the fitted curve near 166 keV, we prepared a ø10 mm × 47 mm ^{226}Ra standard source sample, taking advantage of its 186 keV γ-ray being close in energy to 166 keV. The measured efficiency at 186 keV deviated from the fitted curve by $ $1%, confirming the reliability of the efficiency curve obtained after excluding the 166 keV point.

To investigate the cause of large deviations at 166 keV for some samples, [Figure 3: see original paper] shows γ-ray spectra of ^{139}Ce standard sources measured with GCW5021 (ø10 mm × 47 mm and ø50 mm × 20 mm) and BE5030 (ø50 mm × 2 mm and ø50 mm × 20 mm) detectors. When the efficiency deviation from the fitted curve is large, coincidence peaks from 166 keV γ-rays with 33 keV and 38 keV X-rays are prominent; when deviation is small, these coincidence peaks are negligible. This demonstrates that coincidence effects cause the efficiency deviations. To verify this conclusion, we used Canberra's LabSOCS and Genie2000 software to calculate coincidence correction factors for the full-energy peak count rates of ø50 mm × 2 mm and ø50 mm × 20 mm ^{139}Ce aqueous sources measured with the BE5030 detector. After applying these corrections, the deviation at 166 keV reduced to $ $3%.

Detection efficiency is the primary factor affecting coincidence effects. The GCW5021 and BE5030 HPGe detectors used in our laboratory have larger detection solid angles and crystal volumes than conventional P-type HPGe, increasing detection efficiency for both γ-rays and X-rays. Additionally, their housing materials and electrode configurations enhance low-energy photon detection, further increasing the probability of γ-X-ray coincidences. Therefore, when using monoenergetic standard sources containing ^{139}Ce for HPGe efficiency calibration, it is essential to verify whether the detector and sample geometry induce coincidence effects. The higher the HPGe detection efficiency for photons below 40 keV, the more pronounced the coincidence effect and its impact on calibration accuracy. Many studies [4,6] have overlooked ^{139}Ce coincidence effects, potentially leading to inaccurate efficiency calibrations.

The efficiency calibration results in [Figure 1: see original paper] and [Figure 2: see original paper] show that, except for the 166 keV γ-ray, deviations between measured HPGe efficiency and fitted curves are $ $3% for γ-rays between 60 keV and 662 keV. However, deviations increase for γ-rays above 835 keV, reaching a maximum of 5% (ø75 mm × 10 mm, 1460 keV). This occurs because monoenergetic standard sources contain more low-energy γ-rays and fewer high-energy ones, resulting in larger fitting uncertainties at higher energies.

Uncertainty in efficiency calibration results primarily stems from uncertainties in standard source activity, γ-ray emission probabilities, full-energy peak count rates, and source preparation quantities. Following the method in GB/T16145-2022, we calculated relative uncertainties. Standard source activity uncertainties were taken from calibration certificates, γ-ray emission probability uncertainties from nuclear data libraries (all <1%, taken as 1%), full-energy peak count rate uncertainties (<1%, taken as 1%), and sample preparation uncertainties estimated at 2%. Standard source activity uncertainty is the dominant contributor, yielding relative uncertainties of 4%-6% for both GCW5021 and BE5030 efficiency calibrations.

3.2 Influence of Sample Height on Efficiency

To investigate the relationship between detection efficiency and sample height in well-type HPGe, we calibrated the detector using monoenergetic γ standard source aqueous samples with a diameter of 10 mm and heights H of 0.5 mm, 1 mm, 2 mm, 3 mm, 4 mm, and 5 mm. The results are shown in [Figure 4: see original paper]; the efficiency curve was fitted after excluding the 166 keV data point.

The results show that the variation of full-energy peak efficiency with photon energy follows the same pattern as in [Figure 1: see original paper] and [Figure 2: see original paper]. For a given photon energy, smaller sample heights yield higher full-energy peak efficiency because the sample is positioned closer to the well bottom, increasing the detector solid angle and detection probability. Additionally, smaller sample heights mean less sample mass and reduced self-absorption of γ-rays, further increasing efficiency. Yasuhiro Unno et al. [18] used Monte Carlo methods to calculate full-energy peak efficiencies for water samples in an ORTEC GWL-150-15-S HPGe well at heights of 0.8 mm, 1.7 mm, 2.5 mm, 2.8 mm, 3.3 mm, 3.7 mm, and 4.0 mm. Jong-In Byun et al. [19] used MCNPX to calculate efficiencies for a Mirion GSW275L HPGe (275 cm^3 crystal, 28 mm well diameter, 40 mm well depth) at heights of 10 mm, 20 mm, and 30 mm. Both studies confirm the trends observed in our measurements regarding efficiency variation with sample height and γ-ray energy.

The data in [Figure 3: see original paper] show that for aqueous standard sources with H = 0.5 cm, 1 cm, 2 cm, 3 cm, 4 cm, and 5 cm, deviations at 166 keV from the fitted curve are 49%, 48%, 46%, 41%, 40%, and 38%, respectively. Smaller H yields larger deviations, while deviations at other energies remain $ $5%. This

occurs because smaller sample heights increase detection efficiency for both 166 keV γ-rays and X-rays from ^{139}Ce, enhancing coincidence probability and count loss in the 166 keV full-energy peak.

Therefore, ^{139}Ce in common monoenergetic γ standard sources is unsuitable for well-type HPGe detectors with good low-energy photon response and high efficiency. The deviation in efficiency calibration increases as the prepared standard source height decreases.

3.3 Coincidence Correction Factors

We measured full-energy peak coincidence correction factors for ^{60}Co, ^{88}Y, ^{134}Cs, ^{152}Eu, and ^{133}Ba placed inside the well-type HPGe well using the standard source method. For ^{131}I, we combined the distance and standard source methods. Correction factors for samples placed on the detector surface were also measured. Well-placed samples were ø10 mm × 47 mm with activities of 99 Bq (^{60}Co), 83 Bq (^{88}Y), 105 Bq (^{131}I), 127 Bq (^{134}Cs), 213 Bq (^{152}Eu), and 164 Bq (^{133}Ba). Surface-placed samples were ø50 mm × 20 mm with activities five times higher. The results are presented in .

Due to higher detection efficiency for well-placed samples, shows significant differences between correction factors for well-placed versus surface-placed samples of the same nuclide, with maximum differences reaching 5.5 times. Coincidence effects are clearly more probable for well-placed samples. For the same nuclide under identical conditions, correction factors vary substantially among different γ-ray energies. For ^{152}Eu, the correction factor for the 245 keV peak reaches 6.69, while that for the 1086 keV peak is only 1.53, primarily due to differences in nuclear decay processes and detection efficiency at respective energies.

When measuring atmospheric fallout for ^{131}I using well-type HPGe, the 364 keV peak has a correction factor of 0.99, requiring no correction, whereas the 80 keV or 284 keV peaks require correction. For ^{134}Cs in fallout measured with well-placed samples, the 605 keV peak area must be multiplied by 1.94, compared to only 1.09 for surface-placed samples.

To investigate the relationship between correction factor and sample height in the well, we prepared ø10 cm ^{134}Cs aqueous standard sources with heights H = 0.5 cm, 1 cm, 2 cm, 3 cm, 4 cm, and 5 cm. The results are shown in [Figure 5: see original paper].

[Figure 5: see original paper]

The results demonstrate that smaller sample heights yield larger correction factors. At H = 0.5 cm, the correction factor reaches 8.00 for the 563.23 keV peak and 3.01 for the 604.70 keV peak, decreasing to 3.38 and 1.94, respectively, at H = 5 cm. According to the ^{134}Cs decay scheme, the 563.23 keV γ-ray has the highest probability of cascade emission, resulting in a larger correction factor than other energies. Smaller sample heights increase the detector solid angle and the

probability that two or more cascade-emitted photons interact simultaneously with the crystal, thereby increasing the correction factor.

Uncertainty in correction factors measured by the standard source method originates from uncertainties in monoenergetic standard source activity, emission probabilities, peak count rates, sample preparation quantities, efficiency curve fitting, and target nuclide source parameters. The relative uncertainty is approximately 7%-9%. For the distance method, uncertainty sources are similar but exclude target nuclide activity, yielding relative uncertainties of 6%-8%.

3.4 Random Coincidence

When two or more photons from different nuclei enter the sensitive volume within the HPGe time resolution and are recorded as a single event, the count in the peak of interest may decrease or increase. This phenomenon is called random coincidence. The measured correction factor includes contributions from both cascade and random coincidence; cascade coincidence is independent of sample activity, while random coincidence depends on it. Random coincidence is generally neglected in environmental sample measurements [17], but must be considered when measuring high-activity samples with large-solid-angle, high-efficiency well-type HPGe detectors.

To study the relationship between random coincidence and sample activity, we measured correction factors for ø10 mm × 47 mm standard sources of ^{131}I, ^{134}Cs, and ^{133}Ba at various activities. The results are shown in [Figure 6: see original paper], and [Figure 7: see original paper] displays several random coincidence peaks in the ^{133}Ba spectrum.

[Figure 6: see original paper]
[Figure 7: see original paper]

The data in [Figure 6: see original paper] show relative uncertainties of approximately 8% for correction factor measurements. For ^{131}I and ^{133}Ba, correction factors increase with activity above ~1000 Bq, remaining constant below this threshold. For ^{134}Cs, correction factors remain constant even below 1000 Bq. Increasing ^{131}I activity from 1300 Bq to 6300 Bq raises the correction factor for the 80.19 keV peak by 10%, while increasing ^{133}Ba activity from 1600 Bq to 27000 Bq raises the 276.40 keV peak correction factor by 20%. The presence of random coincidence peaks in [Figure 7: see original paper] confirms the influence of random coincidence. Below 1000 Bq, cascade coincidence dominates and random coincidence is negligible; above 1000 Bq, random coincidence contributions increase progressively, as determined by the time resolution capability of the spectrometry system. Current spectrometry technology requires consideration of random coincidence when measuring high-activity samples with well-type HPGe.

Conclusions

1. When calibrating HPGe detection efficiency using monoenergetic γ-ray standard sources, coincidence effects between ^{139}Ce γ-rays and X-rays need not be considered for detectors insensitive to photons below 40 keV. However, for well-type and broad-energy HPGe detectors with good low-energy photon response and high efficiency, the impact of ^{139}Ce standard sources must be verified before use. Larger detector solid angles and higher detection efficiencies for ^{139}Ce γ-rays and X-rays increase coincidence probability and its effect on calibration results.

2. Few nuclides emit monoenergetic γ-rays in the 150-300 keV range, and most have short half-lives. ^{139}Ce, with its relatively long half-life of 137 days, is commonly used for HPGe efficiency calibration. In this energy region, HPGe detection efficiency varies significantly with energy and calibration points are sparse, making deviations from ^{139}Ce coincidence effects difficult to detect during curve fitting.

3. When using well-type HPGe for sample measurement, smaller sample heights yield higher full-energy peak detection efficiency but also increase the impact of coincidence effects on both efficiency calibration and measurement results.

4. Accurate radionuclide activity measurements with large-solid-angle HPGe require coincidence corrections based on the decay scheme of the target nuclide and sample geometry. For high-activity samples, random coincidence effects may also need to be considered.

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Note: Figure translations are in progress. See original paper for figures.