Improved Algorithm for Close Geometry Characterization of Waste using Gamma-Ray Spectroscopy

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ABSTRACT

Proper characterization of waste is important to reduce the costs of disposal. The requirement for short data collection time often means putting the detector as close to the waste container as possible. Nondestructive assay of gamma ray emitting nuclear waste requires modeling because preparing a standard to match the physical and radiological properties of any waste item is not possible. Several assumptions must be made about the waste. Many models use simplified efficiency determinations and attenuation corrections. These models work well for medium sized items when the detector-to-container distance is at least half of the largest dimension of the object. Other models use a hybrid Monte Carlo approach, still using the container assumptions, but using a complete set of measured detector efficiency parameters.

A new algorithm for calculating the detector efficiency has been developed to allow close-in (that is, high efficiency) detector placement without extensive detector characterization, while retaining acceptable accuracy of the analysis results. This algorithm uses a simple mixed-nuclide gamma calibration, the HPGe detector description (crystal diameter and length, crystal type (n-type or p-type), thickness of germanium dead layer, and distance from the top of the end cap to the crystal) to compute the intrinsic detector efficiency. The intrinsic detector efficiency for the front and side of the detector are related to the detector diameter and length. The efficiency and corrections for the gamma rays from the item being measured are calculated in a voxel-by-voxel manner. A new collimator correction algorithm has also been developed for close geometry measurements. These new algorithms have been implemented in the ISOTOPIC software. Results show significant improvement for large boxes (B-25 box or larger) and items counted as close as 10 cm. Results from low-level drum counter measurements show useful improvements.

INTRODUCTION

When the detector is positioned close to a large item containing radioactivity a significant amount of activity enters the HPGe detector from the side. The classic far-field correction[1] for matrix attenuation does not account for this because it assumes all gamma rays enter the detector normal to and only via the front surface of the detector. An algorithm has been developed that utilizes the crystal diameter and length to estimate the efficiency of the detector from the side surface as well

as the front surface. Only one point-source calibration, positioned normal to the front surface of the detector crystal, is needed. This point source, whose activity is traceable to a national standard, must contain gamma rays that span the energy range of the gamma rays emitted by the nuclides present in the measured item.

CHARACTERIZING THE DETECTOR INTRINSIC EFFICIENCY

The algorithms take advantage of point source detector characterization. Initially, full-energy peak intrinsic efficiency[2] (intrinsic efficiency is the fraction of gamma rays recorded in the net full energy peak to the number incident on the detector) is needed for the detector used to make the analysis. To obtain that information, a point-source calibration is needed at a fixed distance from the face of the detector. When the detector-source distance is in the same range as the detector diameter, the solid angle subtended by the detector is give by:

(1)

$$\Omega = 2\pi \cdot \left(1 - \frac{d}{\sqrt{d^2 + R^2}}\right)$$

(2)

$$\varepsilon_p = \frac{n}{N}$$

where:

Ω	=	subtended solid angle (steradian)
R	=	radius of the detector (cm)
d	=	distance from the calibration point source to the detector (cm)
ε_p	=	absolute full energy peak efficiency of the detector at distance d
n	=	number of gamma rays in the net full energy peak
N	=	number of gamma rays emitted at the full energy from the source

All detectors have a dead layer of germanium, a heat shield inside the cryostat, and an aluminum end cap to house the detector. Attenuation by the endcap and dead layer of germanium must be taken into account for realistic detector characterization. The heat shield and other materials inside the cryostat have negligible attenuation. See Figure 1. The intrinsic peak efficiency for a realistic detector can be determined as shown in Equation 5.
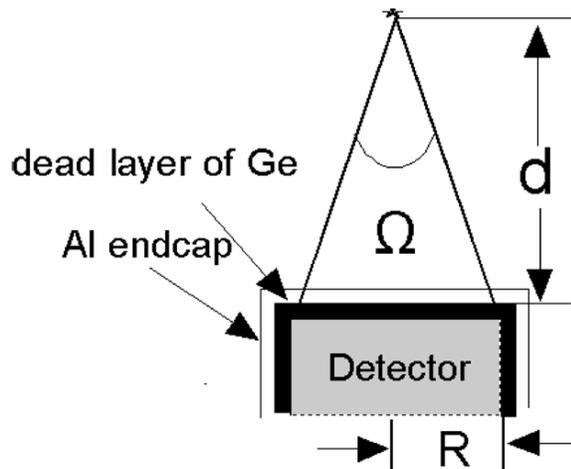

Figure 1 HPGe Detector with a Germanium Dead Layer and an Aluminum Endcap.

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$$(3)$$

$$C_1 = e^{-\mu_e l_e}$$

$$(4)$$

$$C_2 = e^{-\mu_{Ge} l_{dl}}$$

$$(5)$$

$$\varepsilon_{if} = \frac{\varepsilon_p \cdot 4\pi}{C_1 \cdot C_2 \cdot \Omega}$$

where:

C_1 = attenuation correction factor for the aluminum end cap
μ_e = linear attenuation coefficient for the aluminum end cap (cm^{-1})
l_e = aluminum end cap thickness (cm)
C_2 = attenuation correction factor for the dead layer of germanium
μ_{Ge} = linear attenuation coefficient for germanium (cm^{-1})
l_{dl} = thickness of the germanium dead layer (cm)
ε_{if} = full-energy peak intrinsic efficiency for the front of the detector

The side of the detector also detects gamma rays and must be included in the overall full peak intrinsic efficiency. To estimate the average length of gamma-ray cylinder penetration length, *CPL*, assume that, for side detection, the detector is a bar. See Figure 2. The average thickness of the penetration distance is computed using Equation 6.
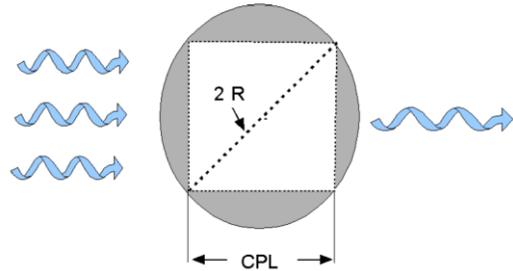

Figure 2 Top View of a Detector to Estimate the Average Penetration Distance.

$$(6)$$

$$CPL = \sqrt{2} \cdot R$$

The relative fraction of gamma-ray activity remaining in the crystal when penetrated from the side and from the front is used to determine the total peak area intrinsic efficiency for the side.

$$(7)$$

$$\varepsilon_{is} = \varepsilon_{if} \frac{1 - e^{-\mu \cdot CPL}}{1 - e^{-\mu \cdot l}}$$

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where:

ε_{is} = side full-energy peak intrinsic efficiency

μ = linear attenuation coefficient for germanium at energy of the gamma ray penetrating the detector (cm^{-1})

l = length of the detector (cm)

COMPUTING ACTIVITY

For a point source positioned with an offset greater than the diameter of the detector, both the side and top of the detector will detect gamma rays from the source. To compute the activity of the source, both detector efficiencies are needed. See Figure 3.

Once the efficiencies of both the front and side of the detector are determined, the activity of the point source can be computed as shown in Equation 8.
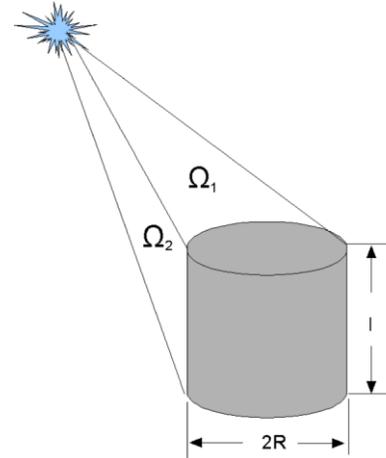

Figure 3 Activity of a Point Source Interacting with the Top and Side Surfaces of a Detector.

(8)

$$Act_{ps} = \frac{CR \cdot 4\pi}{\left(\varepsilon_{if} \cdot C_1 \cdot C_2 \cdot \Omega_1 + \varepsilon_{is} \cdot C_3 \cdot C_4 \cdot \Omega_2\right) \cdot k}$$

where:

Act_{ps} = reported activity (Bq)

Ω_1 = the solid angle subtended on the top surface of the detector (radians)

Ω_2 = the solid angle subtended on the side surface of the detector (radians)

C_3 = attenuation through the side of the end cap and cup holding the detector

C_4 = attenuation through the side dead layer of Ge

k = constant involving the yield (branching ratio), decay corrections, and unit conversion factors (γ emitted/Bq)

When computing the activity of an item, the volume must be subdivided into small voxels, each of which must be corrected individually for matrix and container corrections. The activity of a voxel, Act_i, is shown in Equation 9.

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$$Act_i = \frac{CR \cdot V_i \cdot 4\pi}{\left(\varepsilon_{if} \cdot C_1 \cdot C_2 \cdot \Omega_1 + \varepsilon_{is} \cdot C_3 \cdot C_4 \cdot \Omega_2\right) \cdot V \cdot k}$$

Note that for a box, the volume of each voxel is the same. However, for a cylinder or other items, the voxel volumes are not uniform and the activity must be weighted according to the voxel volume.

(10)

$$Act_{item} = \frac{1}{\sum_{i=1}^{n} \dfrac{D_{1i} \cdot D_{2i} \cdot D_{3i}}{Act_i}}$$

where:

Act_{item} = activity of item being measured (Bq)
V = volume of the item (cm^2)
V_i = volume of the ith voxel(cm^2)
D_{1i} = matrix correction for the ith voxel
D_{2i} = inner container correction factor for the ith voxel
D_{3i} = outer container correction factor for the ith voxel
n = number of voxels

VALIDATING THE ALGORITHM

Sources with known activity were used to validate the algorithm. First, sources were positioned at a standoff of 20 cm and measured at different sideways offsets to validate the improvement using the side efficiency. See Figure 4.

The results were plotted as shown in Figure 5.

Data were collected from many detectors with varying crystal aspect ratios. These calculated results varied from measured results by less than 10% in all measurements. The data shown in Figure 5 vary by less than 2%.
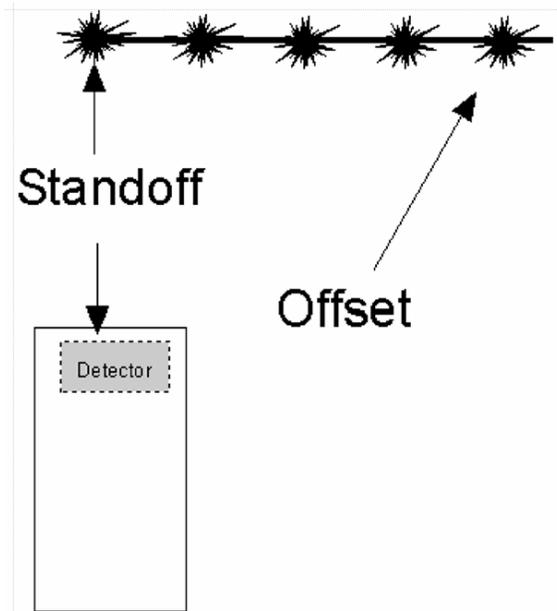

Figure 4 Different Source Positions Used to Validate the Algorithm.

A 50 kg box of KCl was used to validate the algorithm for a homogenous box. Natural ^{40}K was used to provide the known activity. To establish the accuracy for closeup measurements, a detector was positioned at various distances from the box and the results compared to the known ^{40}K content. Natural ^{40}K background was subtracted from all spectra. The standard far-field measurement analysis was compared to the analysis obtained from the close geometry algorithm. The results are shown in Figure 6.
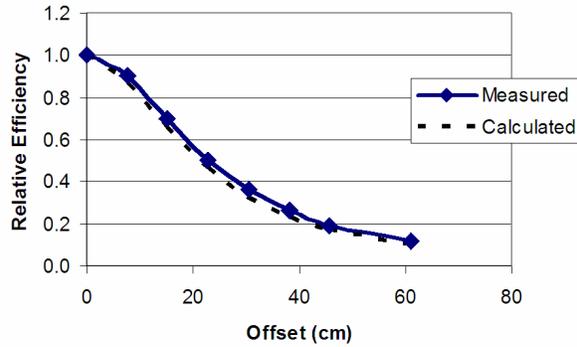
Analyzing ^{40}K spectra using the close-geometry algorithm gives results in good agreement with the accepted value of ^{40}K for standoff distances of 10 cm to 100 cm. As would be expected, the far-field method results are high because the extra gamma ray flux entering the detector from the side is not taken in to consideration in the calibration.
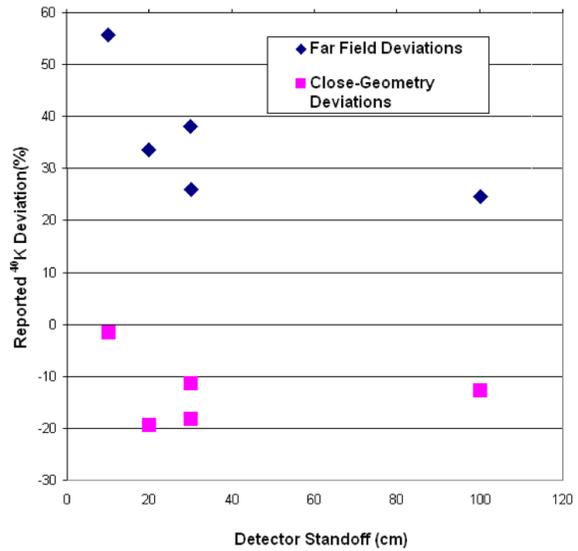
Typically, containers of nuclear waste are not as homogeneous as the ^{40}K content in a box of KCl. For very low level waste measurements, multiple detectors must be positioned very close to 208-liter drums for maximum sensitivity. These drums are frequently rotated to simulate a more homogeneous container as most models initially assume item homogeneity. To simulate a nuclear waste drum, a 208-liter drum was filled with very non-homogeneous, medium-density matrix and spiked with ^{134}Cs, ^{137}Cs, and ^{60}Co. Three measurements were made at three drum locations, as shown in Figure 7, and averaged. The detector was positioned only 10 cm from the surface of the drum at all three locations. The drum was rotated, measured, and analyzed using the close-geometry algorithm and the far-field algorithm.

Figure 5 Change of Efficiency with Offset at Standoff of 20 cm.

Figure 6 Comparison of Close-in Geometry and Far-Field Algorithms for the KCl Box.

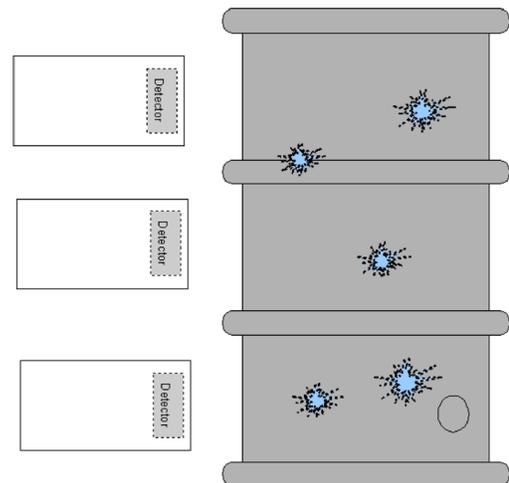**Figure 7 Using the Close Geometry Algorithm to Measure Drums with Non-homogeneous Matrices.**

A comparison of the two methods is shown in Table 1.

Table 1. Comparison of Close Geometry with Far-Field Geometry Algorithm for a Non-homogeneous Drum of Mixed Waste					
Nuclide	Far Field (Bq)	Close Geo. (Bq)	Accepted (Bq)	Difference: FF to Accepted	Difference: CG to Accepted
^{134}Cs	8399	5402	4978	69%	8.5%
^{137}Cs	14911	9805	9850	51%	-0.5%

The close-geometry results are in good agreement with the accepted activities. As expected, the far-field results are high because a large fraction of the gamma rays enters the side of the detector and this activity is not accounted for in that model.

SUMMARY

The close-geometry algorithm show greatly improved accuracy for measurement of radioactive items when the detector is positioned close to the item being measured and no collimator is used. By using the detector crystal diameter and length in addition to other detector parameters, improved accuracy can be obtained without additional calibration effort. This algorithm has been incorporated into the new version of the ISOTOPIC software.

REFERENCES

1. D. Reilly, N. Ensslin, and H. Smith Jr., "Passive Nondestructive Assay of Nuclear Material," United States Regulatory Commission, NUREG/CR 5550, 1991.

2. G.F. Knoll, "Radiation Detection and Measurement," (3rd ed.); Wiley: New York, (1999).