



LAWRENCE  
LIVERMORE  
NATIONAL  
LABORATORY

# A Comparison of Measured and Calculated Gamma Ray Attenuation for a Common Counting Geometry

*R.F. Gaylord*

*Chemistry and Materials Sciences  
Environmental Services (CES)*

*Chemical Biology and Nuclear Science Division*

**March 5, 2004**

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

## INTRODUCTION

In order to perform quantitative gamma spectroscopy, it is necessary to know the sample specific detection efficiency for photons as a function of energy. The detection efficiency, along with the branching ratio for the isotope and gamma ray of interest, is used to convert observed counts/second to actual disintegrations/second, and, hence, has a large effect on the accuracy of the measurement. In cases where the geometry of the source is simple and reproducible, such as a point source, small vial of solid, or jar of liquid, geometry-specific standards may be counted to determine the detection efficiency. In cases where the samples are large, irregular, or unique, this method generally cannot be used. For example, it is impossible to obtain a NIST-traceable standard glove box or 55-gallon drum. In these cases, a combination of measured absolute detector efficiency and calculated sample-specific correction factors is commonly used. The correction factors may be calculated via Monte Carlo simulation of the item (the method used by Canberra's ISOCSS system), or via semi-empirical calculation of matrix and container attenuations based on the thickness and composition of the container and radioactive matrix (ISOTOPIC by EG&G Ortec uses this method). The accuracy of these correction factors for specific geometries is often of vital interest when assessing the quality of gamma spectroscopy data.

During the Building 251 Risk Reduction Project, over 100 samples of high activity actinides will be characterized via gamma spectroscopy, typically without removing the material from the current storage containers. Most of the radioactive materials in Building 251 are stored in cylindrical stainless steel canisters (called USV containers, after the Underground Storage Vault they are commonly stored in), 13 cm in diameter, by 28 cm high, with walls that are 1.8 mm thick. While the actual samples have a variety of configurations inside the USV container, a very common configuration is the material (usually as an oxide powder pellet of approximately 2 cm diameter by ~2 mm thick) in a squat glass jar, with the jar placed in a thin steel food-pack can, which is then placed in the bottom of the USV canister. During data acquisition, the USV containers are typically rotated at approximately 4 rpm on a turntable to eliminate errors due to the material not being centered in the can, or attenuation not being isotropic. An aluminum plate is placed over the container, secured by three vertical rods, to securely hold the container. Pictures of both the containers, and this typical counting configuration are shown below.



The purpose of this experiment was to measure gamma ray attenuation as a function of energy for this standard configuration, and to compare the measured correction factors to those calculated using ISOTOPIC, a commonly used matrix and geometry correction code from EG&GOrtec. ISOTOPIC divides the correction factors for a given sample into 3 categories; matrix attenuation, container attenuation, and geometry. In this experiment, only the container attenuation factors were measured.

## METHOD

A mixed  $\gamma$ -ray point source containing nine different nuclides was used in this experiment (Analytics Inc.). The source consisted of a small spot of dried solution between layers of mylar tape, centered in an aluminum ring, and had an activity of approximately 13  $\mu\text{Ci}$  total. The source contained the nuclides and gamma rays shown below.

Nuclide	Energy (keV)
Am-241	59.5
Cd-109	88.0
Co-57	122.1
Ce-139	165.9
Hg-203	279.2
Sn-113	391.7
Cs-137	661.6
Y-88	898.0
Co-60	1173.2
Co-60	1332.5
Y-88	1836.0

A 45% relative efficiency HPGe detector (EG&G Ortec) was used for these measurements. The detector was energy calibrated before use with a NIST  $\text{Am-241}/\text{Eu-152}$  standard (Analytics Inc.).

The source was counted "free in air" for 28 hours at a distance of 18.55" from the detector face. This count time was sufficient to give greater than  $3\text{E}+05$  counts in the peaks of interest. Next, the source was placed, with the disk standing vertically, in the center of a squat glass jar, 4.2 cm in diameter by 10 cm high, and 3.3 mm thick. The jar was placed in a thin steel food-pack can, 10.4 cm in diameter by 11.8 cm high, with a thickness of 0.33 mm, which was then placed in the center of a USV can. The can was placed so that the exact center (the location of the source) was at 18.55" from the detector face. This configuration was counted for 5.2 hours. The USV can configuration was then rotated at the same distance, at a speed of 4 rpm, and a 2.5 hour count was taken.

All spectra were reanalyzed by Gamma Visions software (EG&G Ortec), and the peak count rate (uncorrected for efficiency) was determined for the peaks of interest.

## RESULTS

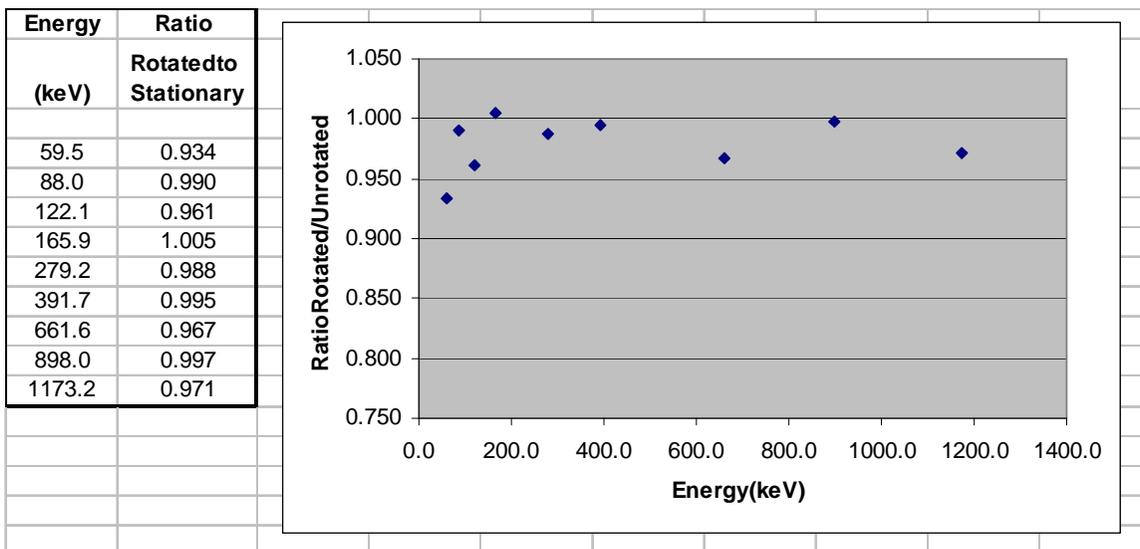
The peak count rate was determined for nine gamma rays, from 59.5 keV (Am -241) to 1173.2 keV (Co -60) for both the “source in air” configuration, and the USV canister configuration. These values are shown below, along with both the ratio [counts/sec (USV configuration)/counts/sec (unshielded configuration)], and the inverse of this ratio, which is defined as the container correction factor for this geometry. With the exception of Am -241, each line had greater than 4.6E+04 total counts, so the error in the calculated container correction factor below is less than 0.5%. For the Am -241 value, there were 1.1E+04 counts, so the error for this correction factor is 1%.

Energy (keV)	Unshielded Cts/sec	USV Configuration Cts/sec	USV Configuration Ratio to unshielded	Container Correction Factor
59.5	5.382	0.640	0.1189	8.41
88.0	9.914	4.411	0.4450	2.25
122.1	5.759	3.576	0.6210	1.61
165.9	6.142	4.308	0.7015	1.43
279.2	4.375	3.402	0.7775	1.29
391.7	4.468	3.600	0.8058	1.24
661.6	3.143	2.718	0.8646	1.16
898.0	5.587	4.952	0.8864	1.13
1173.2	3.756	3.412	0.9083	1.10
1332.5	3.327	3.143	0.9446	1.06

This geometry was modeled using ISOTOPIC, as a point source with a curved steel plate attenuator between the detector and the source. The "best fit" correction factors were generated with a container thickness of 2.3 mm steel.

Energy (keV)	Container Correction Factor	ISOTOPIC Correction Factor	%difference Isotopic to Measured
59.5	8.41	8.39	-0.26
88.0	2.25	2.20	-2.11
122.1	1.61	1.59	-1.26
165.9	1.43	1.36	-4.60
279.2	1.29	1.21	-5.92
391.7	1.24	1.17	-5.72
661.6	1.16	1.14	-1.78
898.0	1.13	1.12	-0.72
1173.2	1.10	1.10	-0.09

Next, the count rate for this geometry was determined with the source rotating and stationary to determine the effect of the rotation. The results are shown below. The error of this ratio is 1.4% for 59 keV, and less than 0.7% for values other than 59 keV.



## CONCLUSION

Gamma ray attenuation was measured as a function of energy for a commonly used sample counting geometry used in the B-251 Risk Reduction Project. As expected, the attenuation is strongly energy dependent, ranging from approximately 10% at 1 MeV, to greater than a factor of 8 at 60 keV. The program ISOTOPIC was found to calculate

gamma ray container correction factors to better than 6% accuracy for gamma rays in the energy range 60 – 1173 keV. Rotation of the sample caused additional attenuation, averaging 3%, possibly with a minor energy dependence. This measured attenuation may be explained by a simple calculation of the area of the detected field of view that is covered by the rods that hold the sample, which is ~3%.

In real B-251 samples, which typically contain milligram -to-gram quantities of actinide oxides at densities of 3 – 6 grams/cm<sup>3</sup>, the gamma ray attenuation due to the sample matrix itself will often be far larger than the container attenuation. As an example, 2 cm thickness of UO<sub>2</sub>, at a density of 5 gm/cm<sup>3</sup> will attenuate a 150 keV gamma ray by a factor of over 20, and will attenuate a 280 keV gamma ray by a factor of over 5. It can be seen that the matrix attenuation factors are more than an order of magnitude larger than the container attenuation factors. In addition, since the true composition and density of the matrix cannot be determined without opening the containers, the uncertainty in matrix attenuation will dominate measurement error, and is not easily reduced.