

COMPARISON OF A VARIETY OF GAMMA ATTENUATION CORRECTION TECHNIQUES FOR DIFFERENT WASTE MATRICES

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ABSTRACT

There are three widely used matrix attenuation correction techniques which are presently used in gamma waste assay systems today. These are the traditional transmission correction techniques from the safeguards Segmented Gamma Scanning Systems, interpolation between several efficiency calibrations of uniform density matrices (as used in Canberra's Q² system), and calculation of the matrix or sample density from comparison of peak ratios from nuclides which emit several gamma lines of differing energies. Each of these techniques has advantages and disadvantages, depending on the source uniformity and density, matrix uniformity and density, container size, required system sensitivity, and typical activity range per container. Each of these techniques is defined and the accuracy of its response when assaying uranium and plutonium contaminated waste is compared. Due to being a commercial establishment without access to the special nuclear materials the measurements are simulated using commonly available fission product nuclides.

INTRODUCTION

The accurate measurement of fissile materials, primarily uranium and plutonium, has been an ongoing project at many facilities worldwide for the past 20 years. During most of this time the primary effort has been in the measurements of process and product materials for the purpose of safeguards and accountability of the material. In recent years these measurements have expanded into the measurement of waste

containers which may contain this material or other radioactive material which needs to be quantified before storage or disposal.

There are two primary measurement techniques which have been used for the measurement of these materials. These are passive/active and passive neutron counting systems and passive gamma assay systems. This paper focuses only on techniques which relate to the passive gamma assay measurement techniques.

The primary problem in accurately assaying gamma emitting radionuclides in waste materials is to correct for the typically unknown waste matrix and source configuration. For small containers with low density waste these corrections are minor and easily handled. But, as the volume and types of waste which must be assayed have increased, the typical container size and density have increased dramatically. At the same time regulatory requirements are demanding more accurate and lower level measurements.

The three widely used matrix attenuation correction techniques which are currently used in gamma waste assay systems today are the traditional transmission correction technique (used in the safeguards Segmented Gamma Scanning Systems), interpolation between several energy-vs-efficiency calibrations of uniform density matrices (used in Canberra's Q² system), and calculation of the matrix or sample density from comparison of sample peak ratios (used in a variety of forms in different software packages). These are described in detail in the following sections.

DESCRIPTION OF MATRIX CORRECTION TECHNIQUES

Each of the following matrix correction techniques is in use in commercially available systems today. This section briefly describes the technique and the algorithm without going into great detail regarding the development of the algorithm. A more complete derivation of these techniques can be found in the associated references.

Transmission Correction

The transmission correction technique for passive gamma assays was developed by Los Alamos National Laboratory.¹ This technique uses a highly collimated transmission source which is beamed through the sample container towards the germanium detector. Correction algorithms can be used to calculate the attenuation of the gamma radiation from the sample based on the attenuation of the transmission source beam. To optimize this correction the transmission source gamma line was typically chosen to be very close in energy to the gamma line for the assay nuclide.

When this technique is applied to assaying general fission product waste, the ratios of the mass absorption coefficients between the transmission energy and the assay peak energy must be used to correct for the differences in absorption over large energy ranges.

The basic assumptions which have been traditionally applied to this technique are that the matrix is uniform over the vertical segment being measured, and that the sample activity is uniformly distributed in the segment and does not contain lumps. A recent paper "Non-destructive Assay Techniques and Associated Measurement Uncertainties"² has assigned typical errors associated with these parameters.

Multiple Efficiency Calibrations

This technique as demonstrated in Canberra's Q² system utilizes several energy-vs-efficiency calibrations covering the typical sample density range which will be measured.

When container assays are performed, the container density is either automatically calculated based on the container volume and the measured weight, or can be manually entered if known. The software then interpolates between the two efficiency calibrations which bracket the container density for the particular energies of interest.

As with the transmission source technique, this technique assumes that both the matrix and the sample distribution are uniform. However, if the efficiency calibrations are performed with representative matrices, no particular knowledge of the mass attenuation coefficients for the sample matrix need to be known.

Peak Ratioing

The final technique requires that the nuclide of interest (or ones with the same distribution in the sample) contain several gamma energy lines. The basis of this technique is that the changes in the ratios of these peaks directly characterizes the attenuation matrix. To obtain a matrix correction with this technique, the ratios of two or more peaks from different gamma lines of a single nuclide are initially measured with no attenuating material present. During an assay, the ratios of the peaks of the same nuclides are again measured. Correction algorithms are then used to determine the matrix density based on the variation in the peak ratios. This technique may average the results for several nuclides to improve the results.

As opposed to the first two techniques, this technique makes no assumptions concerning the uniformity of the sample matrix or activity distribution. This technique does however require adequate count rates for the peaks used in the ratioing to be able to calculate a precise matrix correction factor.

Experimental Measurement Techniques

Experimental measurements were made in the factory at Canberra Industries to compare the accuracy of each of these techniques under various conditions. To simulate ²³⁵U or ²³⁹Pu sources, measurements were made using a ¹⁵²Eu source, which covers a wide range of gamma energies. For transmission correction measurements the gamma energy which was used was the 1332 keV line from ⁶⁰Co.

Table 1. Comparison of uranium and plutonium lines simulated with the Eu source for peak ratioing

ASSAY NUCLIDES	ENERGY (keV)	ABUNDANCE	¹⁵² Eu GAMMA USED	
			(keV)	ABUNDANCE
²³⁵ U	143	0.11	122	0.37
²³⁵ U	204	0.05	244	0.08
²³⁹ Pu	129	0.00005	122	0.37
²³⁹ Pu	414	0.00001	412	0.02

Table 1 indicates which lines were used from the ¹⁵²Eu to simulate lines for peak ratioing for the uranium and plutonium. The results shown in Table 2 for the peak ratioing were corrected using the two lines to simulate the uranium. Corrections using the plutonium lines for ratioing gave similar results.

Initial measurements were made using Canberra's four Q² calibration drums. These drums, standard 200 liter (55 gallon), each contain a uniform density matrix. The matrices used were:

Foam	0.1 gm/cc
Insulation material	0.3 gm/cc
Wood	0.7 gm/cc
Sand	1.7 gm/cc

Each drum has 9 hollow tubes placed vertically through the matrix in equal volume sections of the drum. These are used for loading measurements and calibration sources into the drums. Position 1 represents the center of the drum, position 5 represents a point approximately equal to a uniform distribution, and position 9 represents the tube at the outer edge of the drum.

A final drum was loaded with a non-uniform mixture of materials ranging from paper and plastic to wood and steel to simulate a non-uniform matrix.

Measurements were made on each of the drums and the various correction techniques were applied to the same data. Some of the measurements for the multiple efficiency correction technique were not able to be performed at this time. Therefore, data with the non-uniform drum was not available for this technique. Mass absorption coefficients were not totally optimized. This is partially why some energy ranges tend to be biased from other results in all measurements.

RESULTS

A compilation of the results is shown in Table 2. The results indicate the deviation from a normalized value of 1. It can be seen from the results that each technique has advantages in some cases and disadvantages in others. These advantages and disadvantages are summarized in Table 3.

The transmission source correction technique has advantages in that it accurately measures the average density of each segment of the container. Therefore, the results are excellent when the source distribution is uniform. However, for high density drums or large containers the measured count rate from the transmission source can be reduced to levels where the error caused by counting statistics can become significant. This does not tend to show up in our data due to the high energy of the transmission source (1332 keV)

Table 2. Corrected results with different correction techniques

.1 GM/CC DRUM									
ENERGY	POS 5			POS 1			POS 9		
keV	TRANS	PEAK	Q2	TRANS	PEAK	Q2	TRANS	PEAK	Q2
122	0.88	0.62	0.99	0.78	0.96	0.85	1	0.95	1.31
244	0.89	0.62	0.99	0.92	0.96	0.92	1.11	0.95	1.29
412	0.88	XXX	XXX	1.07	0.95	XXX	1.5	1.24	XXX
779	1.06	0.71	0.95	0.95	0.79	0.9	1.1	0.81	1.25
965	1.09	0.7	1.07	1.06	0.91	0.91	1.2	0.9	1.27
1408	1	0.66	0.95	1.18	0.78	0.93	1.09	0.8	1.27
.3 GM/CC DRUM									
keV	TRANS	PEAK	Q2	TRANS	PEAK	Q2	TRANS	PEAK	Q2
122	0.9	1.19	1.01	0.65	0.6	0.62	1.11	1.6	1.64
244	0.96	1.19	1.08	0.72	0.6	0.71	1.23	1.6	1.66
412	1.22	1.62	XXX	0.86	0.73	XXX	1.2	2.12	XXX
779	1	0.87	0.95	0.91	0.71	0.77	1.17	1.04	1.41
965	1.08	0.97	0.96	0.95	0.76	0.81	1.2	1.13	1.42
1408	1.03	0.89	0.98	0.93	0.73	0.81	1.09	0.97	1.37
.8 GM/CC DRUM									
keV	TRANS	PEAK	Q2	TRANS	PEAK	Q2	TRANS	PEAK	Q2
122	0.72	0.37	0.96	0.2	0.21	0.11	1.96	1.66	2.24
244	0.78	0.37	0.99	0.26	0.21	0.2	1.98	1.66	2.14
412	0.96	0.5	XXX	0.4	0.33	XXX	1.76	2.47	XXX
779	0.9	0.44	0.98	0.55	0.36	0.36	1.65	0.88	1.66
965	0.93	0.5	0.89	0.61	0.45	0.39	1.61	0.97	1.67
1408	0.9	0.5	1.05	0.67	0.47	0.46	1.43	0.86	1.52
1.7 GM/CC DRUM									
keV	TRANS	PEAK	Q2	TRANS	PEAK	Q2	TRANS	PEAK	Q2
122	0.8	0.73	0.99	0.006	0.38	0	3.8	1.03	XXX
244	1	0.73	1.04	0.022	0.38	0	3.5	1.03	XXX
412	1.1	0.74	XXX	0.02	0.23	XXX	3	0.93	XXX
779	1.17	0.65	0.97	XXX	XXX	0.09	2.8	0.88	XXX
965	1.31	0.71	1.07	0.22	0.73	0.14	2.8	0.89	XXX
1408	1.17	0.69	0.99	0.27	0.7	0.19	2.2	0.86	XXX
NON-UNIFORM DENSITY DRUM									
keV	TRANS	PEAK	Q2	TRANS	PEAK	Q2	TRANS	PEAK	Q2
122	0.97	1.21	XXX	0.67	1.66	XXX	1.12	1.28	XXX
244	1.12	1.21	XXX	0.9	1.65	XXX	1.33	1.28	XXX
412	1.12	1.18	XXX	0.86	1.47	XXX	1.12	1.05	XXX
779	1.48	0.92	XXX	0.96	1.19	XXX	1.27	1.12	XXX
965	1.23	1.16	XXX	1.07	1.34	XXX	1.28	1.16	XXX
1408	1.15	1.04	XXX	0.87	1.12	XXX	1.13	0.99	XXX

and the long count times on the higher density drums to improve statistics. High activities in the drums can also tend to reduce the statistical accuracy of the transmission count, and can even mask it totally. As the density of the matrix increases, non-uniform source distributions become the dominant source of error.

Measuring the sample weight and calculating the sample density based on the weight and volume as is used in the Q² technique also provides good results for uniform sample distributions. In addition, this technique is not affected by counting statistics. This keeps the correction errors from becoming significant on high density or high activity samples. This technique has the same disadvantage as the transmission correc-

tion technique when the source activity distributions become non-uniform. This is seen in the measurements with the sample in the center or the edge of the drum.

The peak ratioing correction technique is extremely sensitive to counting statistics. This is due to the fact that in most cases small changes in the peak ratios will cause large correction factors. Some of the values shown in Table 2 for the peak ratioing would have been closer to unity if statistical limitations had been imposed. In low density containers the attenuation correction is minimal. On these containers the peak ratioing technique is typically less accurate than the transmission or container weight technique. The real

Table 3. Advantages and disadvantages of each technique

<p>TRANSMISSION CORRECTION TECHNIQUE</p> <p>ADVANTAGES:</p> <ul style="list-style-type: none"> • Accurately measures the average density through the drum for each segment • Technique can be used when sample activities are low <p>DISADVANTAGES:</p> <ul style="list-style-type: none"> • Large errors for high density containers • Significant errors with radial non-uniformities in the source distribution with moderate to high density containers • Large errors if transmission count rate is masked by high sample activity • When measuring fission product waste, changes in mass absorption coefficients can increase errors
<p>MULTIPLE EFFICIENCY CALIBRATIONS BASED ON DENSITY</p> <p>ADVANTAGES:</p> <ul style="list-style-type: none"> • Technique is not sensitive to counting statistics • Efficiency calibrations cover the complete energy range therefore not sensitive to mass absorption coefficients <p>DISADVANTAGES:</p> <ul style="list-style-type: none"> • Significant errors with radial non-uniformities in the source distribution with moderate to high density containers
<p>PEAK RATIOING TECHNIQUE</p> <p>ADVANTAGES:</p> <ul style="list-style-type: none"> • Corrects for non-uniform source distributions • Corrects for self absorption in the source <p>DISADVANTAGES:</p> <ul style="list-style-type: none"> • Extremely sensitive to counting statistics • Cannot be used for small quantities of source material • Less accurate than other techniques on low density containers • Sensitive to changes in atomic number of matrix material

advantage to the peak ratioing technique is that the measurement is actually looking at the effect of the sample self attenuation or the attenuation through the matrix. Therefore, non-uniform distributions of activity are more accurately corrected with this technique as long as there are reasonable counting statistics for the nuclides of interest. This technique also improves the correction when lumps of uranium or plutonium are in the container.

All of these techniques can be somewhat sensitive to significant changes in the atomic number of the matrix material. This is not a problem with the transmission technique if the transmission source activity is close to the energy of the nuclides being measured, but if general fission product waste is being measured, many gamma lines will not be close to the transmission source energy. Therefore, for best results it is important to know the types of matrices which are in the sample.

CONCLUSIONS

As seen in the results, each technique has its advantages and disadvantages. Therefore, the measurement requirements and knowledge or lack of knowledge concerning the source and matrix may define the best technique in most applications.

Ultimately a combined system which would utilize several of these techniques would provide the best results in general measurement situations. In Canberra's waste assay system which uses the peak ratioing technique, the sample weight and volume are automatically used when the peak ratioing counting statistics are inadequate.

Another possible implementation might use the transmission correction technique in low density containers. When transmission source count rates could not provide adequate counting statistics the correction could be based on the container weight and volume. After the basic matrix correction was performed using the technique above, a secondary correction could be made based on the peak ratioing technique, which would reduce the effects of non-uniform source distributions. Several other combinations of these techniques could also be considered.

In all situations, limitations on the use of the transmission source and peak ratioing techniques must be based on the counting statistics. Otherwise the correction can potentially increase the error of the measurement.

REFERENCES

1. Martin, E.R., Jones, D.F., and Parker, J.L. (1977). *Gamma-Ray Measurements with the Segmented Gamma Scan*. Report LA-7059-M. Los Alamos National Laboratory.
2. Hsue, S.T. and Sampson, T.E. (1992). "Non-destructive Assay Techniques and Associated Measurement Uncertainties". *Journal of Nuclear Materials Management*. 20, 2: 17.
3. Gillespie, B. and Zebarth, M. (April 1993). *Comparison of a Variety of Gamma Attenuation Correction Techniques for Different Waste Matrices*. Presented at ESARDA: Salamanca, Spain.

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