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QUANTITATIVE PORTABLE GAMMA SPECTROSCOPY SAMPLE ANALYSIS FOR NON-STANDARD SAMPLE GEOMETRIES**

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Utilizing a portable spectroscopy system, a quantitative method for analysis of samples containing a mixture of fission and activation products in nonstandard geometries was developed. The method can be used with various sample and shielding configurations where analysis on a laboratory based gamma spectroscopy system is impractical. The portable gamma spectroscopy method involves calibration of the detector and modeling of the sample and shielding to identify and quantify the radionuclides present in the sample. The method utilizes the intrinsic efficiency of the detector and the unattenuated gamma fluence rate at the detector surface per unit activity from the sample to calculate the nuclide activity and Minimum Detectable Activity (MDA). For a complex geometry, a computer code written for shielding applications (MICROSHIELD) is utilized to determine the unattenuated gamma fluence rate per unit activity at the detector surface. Lastly, the method is only applicable to nuclides which emit gamma rays and cannot be used for pure beta emitters. In addition, if sample self absorption and shielding is significant, the attenuation will result in high MDA's for nuclides which solely emit low energy gamma rays. The following presents the analysis technique and presents verification results demonstrating the accuracy of the method.

INTRODUCTION

Gamma spectroscopy is the most commonly used method for qualitative determination of gamma emitting isotopes for various samples. The dominant characteristic of germanium detectors is their excellent energy resolution. The superiority of the germanium system in energy resolution

allows the separation of many closely spaced gamma-ray energies which remain unresolved in the NaI(Tl) spectrum. For typical quantitative analysis performed on a laboratory based system, an absolute efficiency calibration is performed using a traceable source similar in geometry and gamma emission energy characteristics as the samples requiring analysis. Unfortunately the

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absolute efficiency is highly dependent on sample geometry which requires separate calibrations to be performed for each sample geometry of interest. In addition, many samples are physically too large for calibration with typical sources.

By determining the intrinsic efficiency of the detector and modeling the attenuation associated with the sample and shielding, quantitative analysis on nonstandard geometries can be performed. Quantitative analysis is possible since the intrinsic efficiency of the detector is relatively independent of the sample geometry and depends primarily on the gamma emission energy and physical thickness of the detector in the direction of the incident radiation.

METHODS

Energy Calibration

An accurate energy calibration should involve using a source with gamma ray energies spanning those to be measured in the unknown spectrum. For this method a mixed gamma source ranging from 60 keV (Am^{241}) to 1836 keV (Y^{88}) was used to identify nuclides from 50 keV to 2.5 MeV.

The goal in gamma spectroscopy is to identify and quantify the radionuclides in a sample. By performing an energy calibration, a relationship between channel number and gamma ray energy can be determined. During the energy calibration, data is fit to the following equation.

$$E = m * \text{channel} + b + c * \text{channel}^2 \quad (1)$$

where

E = Energy of photopeak in keV

m = slope

channel = channel number

b = intercept

c = non-linear coefficient

Point Source Detector Calibration

As discussed previously, the detector efficiencies are typically presented as intrinsic or absolute. The absolute efficiency is defined as the number of pulses recorded divided by the number of gamma rays emitted by the source. Similarly, the intrinsic efficiency is defined as the number of pulses recorded divided by the number of gamma rays incident on the detector. Given the following arrangement for an area source and a detector,

Figure 1. Solid Angle Arrangement

the solid angle seen by the source is given by:

$$\omega = 2\pi \left(1 - \cos \alpha \left(1 + \frac{3r^2}{2d^2} * \frac{(\sin 2\alpha)^2}{4} \right) \right) \quad (2)$$

where:

d = source to detector distance

a = detector radius

r = source radius

alpha = arctan(a/d)

For source to detector distances much greater than the detector radius Equation 2 reduces to:

$$\omega = 2\pi[1 - \cos\alpha]. \quad (3)$$

In order to determine the intrinsic efficiency of the detector it is necessary to place a source at an ample distance from the detector surface so that the source will be seen as a point source. If the source is not seen as a point source, an area to area correction factor will have to be determined.

The calibration distance will be dependent on the area of the calibration source, and also on the detector radius. Using Equation 2 and 3 for a detector radius of 2.75 cm, and a source radius of 1.27 cm, it is shown that using a distance of 9.8 cm provides enough distance for the calibration source to be considered a point source.

To ensure that the gamma rays entered through the front surface of the detector, during calibration and sample characterization, the sides of the detector were shielded with lead. Calibration of the portable gamma spectroscopy system was done by counting a one inch diameter mixed nuclide source approximately 9.8 cm from the detector surface.

$$\log(Eff_{9.8}) = a_1 E^1 + a_2 + \frac{a_3}{E} + \frac{a_4}{E^2} + \frac{a_5}{E^3} + \frac{a_6}{E^4} \quad (4)$$

where:

- $Eff_{9.8}$ = Gamma ray efficiency for point source at 9.8 cm (cps/γps)
- E = Gamma ray emission energy (keV)
- a_1 - a_6 = Fit parameters

The intrinsic efficiency of the detector usually depends primarily on the detector

material, the radiation energy, and the physical thickness of the detector in the direction of the incident radiation. It was assumed that the source emits radiation isotropically, and that no attenuation takes place between the source and detector. From the point source calibration, it was possible to determine the intrinsic efficiency by the following:

$$E_{int} = \frac{Eff_{9.8}}{\left(\frac{E}{4\pi r^2}\right)} \quad (5)$$

where:

- E_{int} = Intrinsic efficiency of detector [cps/(MeV/cm²sec)]
- $Eff_{9.8}$ = Efficiency at 9.8 cm calibration (cps/γps)
- r = Source to detector distance (cm)
- E = Gamma ray emission (MeV/γ).

ACTIVITY DETERMINATION

Since the geometry of the samples differed greatly from the geometry of the actual calibration it was necessary to determine the actual unattenuated gamma energy fluence from the sample that was reaching the detector surface. However, to determine the actual energy fluence reaching the detector it was necessary to model the sample geometries using the shielding code MICROSIELD 4.2. To determine the energy fluence rate reaching the detector surface, the source was assumed to be 1 Curie. Once the energy fluence rate reaching the detector per unit activity is calculated, the intrinsic efficiency can be applied to determine an activity value of the samples.

$$A_{sam} = \frac{R}{E_{int} * F} \quad (6)$$

where:

A_{sam} = Activity of sample [Curies]
 R = Observed net counts in peak / sample count time [counts/sec]
 E_{int} = Intrinsic efficiency of detector [cps / (MeV/cm²sec)]
 F = Fluence reaching detector from sample [(MeV/cm²sec) / Ci].

MINIMUM DETECTABLE ACTIVITY (MDA) FOR AM²⁴¹

For the particular application of this method for Sandia National Laboratories (SNL) it was necessary to attempt to identify the minimum detectable concentration of Pu²³⁹ (i.e. through the use of the associated Am²⁴¹ gamma ray) in the presence of relatively high levels of fission and activation products. SNL used this method to perform waste characterization analysis on radioactive waste containing experimental reactor fuels of various burn up. There was a need to obtain information related to the highest level of Am²⁴¹ (and the associated Pu²³⁹) that could be contained in the waste and still remain undetected. This information was used to assess the likelihood that the waste was TRU waste and to assess each package's impact on SNL's nuclear facility status. A similar treatment could be used to assess the MDA of any other gamma emitting nuclide of interest.

If sample self absorption and shielding is significant, the attenuation will result in high MDA's for nuclides which solely emit low energy gamma rays. To estimate the MDA of (e.g. Am²⁴¹) in the sample under consideration, an efficiency [Eff_{sam} (c/γ)] must be determined for the sample matrix.

Eff_{sam} is determined by the use of the equation listed below.

$$Eff_{sam} = \frac{E_{int} * F}{3.7 \times 10^{10} * Y} \quad (7)$$

where:

Eff_{sam} = Sample matrix efficiency (c/γ)
 E_{int} = Intrinsic efficiency [cps/(MeV/cm²sec)]
 F = Fluence rate reaching detector from sample [(MeV/cm²sec)/Ci]
 3.7×10^{10} = Conversion factor Bq to Curies [3.7 x 10¹⁰ Bq/Ci]
 Y = Number of γ's per disintegration [=0.359 for Am²⁴¹]

Since the MDA is inversely proportional to the efficiency [c/γ] the following relationship can be determined.

$$Eff_{sam} * MDA_{sam} = Eff_{9.8} * MDA_{9.8} \quad (8)$$

This equation can then be solved for the estimated Am-241 MDA for the sample.

$$MDA_{sam} = \frac{Eff_{9.8} * MDA_{9.8}}{Eff_{sam}} \quad (9)$$

where:

MDA_{sam} = Estimated Am-241 MDA for Sample [nCi]
 Eff_{sam} = Efficiency of sample matrix [counts/γ]
 $MDA_{9.8}$ = MDA using mixed nuclide calibration at 9.89 cm [nCi]
 $Eff_{9.8}$ = Gamma ray efficiency for point source at 9.8 cm [counts/γ].

VERIFICATION MEASUREMENTS

Sample and Material Description

Several geometries were utilized for the verification/validation measurements. The first geometry chosen was an Analytics 125 mL Loral jar that contained an epoxy source matrix ($\rho=1.75 \text{ g/cm}^3$). The jar was modeled as a right circular cylinder with a height of 3.3 cm, and a diameter of 6.0 cm. An aluminum ($\rho=2.70 \text{ g/cm}^3$) and plastic ($\rho=1.135 \text{ g/cm}^3$) attenuator was placed between the jar and the detector. For modeling purposes the epoxy was modeled as hexane (CH_2) which is the typical constituent of rubber cement.

Figure 2. Jar Verification Geometry

Since waste is frequently packaged in 55 Gallon drums an NIST traceable, calibrated line source was used in conjunction with Canberra Industries, Inc. Q² 55 gallon calibration drums. The Canberra calibration drums consist of a set of four drums, each containing a different density material. The line source consists of uniformly distributed Am^{241} and Eu^{152} in an epoxy matrix ($\rho=1.07 \text{ g/cm}^3$) cast in a 0.375" outside diameter by 30.0" long aluminum tube with 0.035" thick walls. At both ends of the aluminum tube, 0.080" end plugs are used giving the source an active length of 29.84". Within the

calibration drum, there are nine individual locations for the source tube. For the verification measurements, the source tube was placed directly in the center of the calibration drum that was used.

Table 1. 55 Gallon Drum Densities

Material	Canberra Density (g/cm^3)
Foam	0.014
Fiberboard	0.470
Pressed Wood	0.766
Sand	1.67

The calibration drums are Department of Transportation (DOT) specification 17H 55 gallon steel drums. The 55 gallon drums were modeled as a right circular cylinder with a height of 35 inches and an inside diameter of 22.5 inches. The body of the drum is constructed from 18 gauge steel with thickness range of 0.0428 inches to 0.0478 inches. For modeling purposes, the average thickness of 0.0453 inches was used.

Figure 3. 55 Gallon Drum Verification Geometry - Side View

Figure 4. 55 Gallon Drum Verification
Geometry - Top View

The drum walls were modeled as stainless steel. For the shielding calculations the composition of steel was assumed to consist of 60% iron, 20% manganese, and 20% nickel by mass, and having a density of 7.9 g/cm³. The foam material was assumed to primarily consist of polyethylene ($\rho = 0.014$ g/cm³). The fiberboard drum and pressed wood were modeled as cellulose ($C_6H_{10}O_5$)_n with a density equivalent to the Canberra supplied densities of 0.470 g/cm³, and 0.766 g/cm³, respectively. The fourth drum (sand) was modeled as SiO₂, the primary constituent of sand, with a density of 1.67 g/cm³. The following table lists the materials used in the calibration drums in addition to Canberra Industries published density values from the Q² gamma spectroscopy system applications systems document.

Table 2. 55 Gallon Drum and Source
Material Descriptions

Material	Modeled Material (% by Mass)	Chemical Formula	Density (g/cm ³)
Steel	60% Fe, 20% Mn, 20% Ni	Fe, Mn, Ni	7.9
Aluminum	100% Aluminum	Al	2.7
Epoxy	50% Epichlorohydrin 50% Bisphenol A (Typical components of epoxy)	CH ₂ OCHCH ₂ Cl (CH ₃) ₂ C(C ₆ H ₄ OH) ₂	1.07
Foam	Polyethylene (typical component)	(CH ₂) _n	0.015
Fiberboard	Cellulose	(C ₆ H ₁₀ O ₅) _n	0.441
Pressed Wood	Cellulose	(C ₆ H ₁₀ O ₅) _n	0.740
Sand	Sand	SiO ₂	1.67

SUMMARY OF VERIFICATION RESULTS

Tables 3, 4, 5, 6, and 7 present the predicted activities and compares the results with the certificate activities for the five verification samples. MDA values for the 55 Gallon Drum geometries are also presented in Table 8.

Table 3. Comparison of Results for Jar Geometry ($\rho=1.75 \text{ g/cm}^3$)

Nuclide	Energy (keV)	Estimated Activity (Bq)	*2 σ Error (Bq)	Certificate Activity (Bq)	2 σ Error (Bq)	% Bias
Co-57	122	543	82	458	11	19
Ce-144	166	478	71	438	11	9
Sn-113	392	1862	280	1654	41	13
Sr-85	514	2447	447	2292	56	7
Cs-137	662	1633	212	1437	36	14
Y-88	898	4241	555	3816	93	11
Co-60	1173	2445	317	2191	55	12
Co-60	1332	2498	323	2191	55	14
Y-88	1836	4320	621	3816	93	13

* Note the 2 σ error reported is only the error associated with the unprocessed gamma spectroscopy results. This error is only associated with the raw count data and encompasses only the counting statistics which involves peak area error, gamma ray abundance error, efficiency error, a 2% non-random sample error, and a 10% random sample error. The actual sample error with the activity value is much larger than the counting statistical error due to the uncertainties in the geometry and materials used for modeling.

Table 4. Comparison of Results for 55 Gallon Foam Drum ($\rho=0.014 \text{ g/cm}^3$)

Nuclide	Energy (keV)	Estimated Activity (kBq)	*2 σ Error (kBq)	Certificate Activity (kBq)	2 σ Error (kBq)	% Bias
Am-241	59	308	50	267.5	5.3	15
Eu-152	122	572	82	582.6	16.7	-2
Eu-152	245	571	101	582.6	16.7	-2
Eu-152	344	592	99	582.6	16.7	2
Eu-152	779	640	86	582.6	16.7	10
Eu-152	964	663	91	582.6	16.7	14
Eu-152	1112	671	90	582.6	16.7	15
Eu-152	1408	682	89	582.6	16.7	17

* Note the 2 σ error reported is only the error associated with the unprocessed gamma spectroscopy results. This error is only associated with the raw count data and encompasses only the counting statistics which involves peak area error, gamma ray abundance error, efficiency error, a 2% non-random sample error, and a 10% random sample error. The actual sample error with the activity value is much larger than the counting statistical error due to the uncertainties in the geometry and materials used for modeling.

Table 5. Comparison of Results for 55 Gallon Fiberboard Drum ($\rho=0.470 \text{ g/cm}^3$)

Nuclide	Energy (keV)	Estimated Activity (kBq)	*2 σ Error (kBq)	Certificate Activity (kBq)	2 σ Error (kBq)	% Bias
Am-241	59	505	165	267.5	5.3	89
Eu-152	122	816	117	582.2	16.7	40
Eu-152	245	727	140	582.2	16.7	25
Eu-152	344	769	132	582.2	16.7	32
Eu-152	779	784	109	582.2	16.7	35
Eu-152	964	792	108	582.2	16.7	36
Eu-152	1112	804	110	582.2	16.7	38
Eu-152	1408	791	104	582.2	16.7	36

* Note the 2 σ error reported is only the error associated with the unprocessed gamma spectroscopy results. This error is only associated with the raw count data and encompasses only the counting statistics which involves peak area error, gamma ray abundance error, efficiency error, a 2% non-random sample error, and a 10% random sample error. The actual sample error with the activity value is much larger than the counting statistical error due to the uncertainties in the geometry and materials used for modeling.

Table 6. Comparison of Results for 55 Gallon Wood Drum ($\rho=0.766 \text{ g/cm}^3$)

Nuclide	Energy (keV)	Estimated Activity (kBq)	*2 σ Error (kBq)	Certificate Activity (kBq)	2 σ Error (kBq)	% Bias
Am-241	59	ND	---	267.5	5.3	---
Eu-152	122	459	81	581.6	16.6	-21
Eu-152	245	489	91	581.6	16.6	-16
Eu-152	344	451	76	581.6	16.6	-22
Eu-152	779	453	63	581.6	16.6	-22
Eu-152	964	470	65	581.6	16.6	-19
Eu-152	1112	469	65	581.6	16.6	-19
Eu-152	1408	469	62	581.6	16.6	-19

* Note the 2 σ error reported is only the error associated with the unprocessed gamma spectroscopy results. This error is only associated with the raw count data and encompasses only the counting statistics which involves peak area error, gamma ray abundance error, efficiency error, a 2% non-random sample error, and a 10% random sample error. The actual sample error with the activity value is much larger than the counting statistical error due to the uncertainties in the geometry and materials used for modeling.

Table 7. Comparison of Results for 55 Gallon Sand Drum ($\rho=1.67 \text{ g/cm}^3$)

Nuclide	Energy (keV)	Estimated Activity (kBq)	*2 σ Error (kBq)	Certificate Activity (kBq)	2 σ Error (kBq)	% Bias
Am-241	59	ND	---	267.5	5.3	---
Eu-152	122	ND	---	582.0	16.6	---
Eu-152	245	ND	---	582.0	16.6	---
Eu-152	344	448	96	582.0	16.6	-23
Eu-152	779	573	87	582.0	16.6	-2
Eu-152	964	567	87	582.0	16.6	-3
Eu-152	1112	613	92	582.0	16.6	5
Eu-152	1408	600	82	582.0	16.6	3

*Note the 2 σ error reported is only the error associated with the unprocessed gamma spectroscopy results. This error is only associated with the raw count data and encompasses only the counting statistics which involves peak area error, gamma ray abundance error, efficiency error, a 2% non-random sample error, and a 10% random sample error. The actual sample error with the activity value is much larger than the counting statistical error due to the uncertainties in the geometry and materials used for modeling.

Table 8. Minimum Detectable Activities for 55 Gallon Drums

Drum Material	Nuclide	MDA (kBq)
Foam	Am ²⁴¹	10
Fiberboard	Am ²⁴¹	130
Wood	Am ²⁴¹	406
Sand	Am ²⁴¹	350673

This method is highly dependent on the modeling parameters. There will be less geometry dependence for samples that are located at large distances from the detector. At larger distances there will be less dependence on the solid angle seen by the detector surface and also on the attenuation properties of the sample materials.

Although mass attenuation coefficients are

material and energy dependent, the dependence of the mass attenuation coefficient on the material is significantly reduced at higher energies. Therefore, less uncertainty is associated with the activity calculations involving high energy gamma ray emissions. Therefore, the nuclide activity results should be based on the higher energy gamma ray emission with the highest yield when possible.

In addition, samples that experience deadtimes typically above 10% results in random summing losses which produces a non-linear response in the gamma spectroscopy system.

CONCLUSION

This method proves useful when quantitative results are needed for samples that are physically too large and/or too highly

radioactive for analysis on a laboratory based system. Reasonable results can be achieved when sample self absorption and shielding can be modeled accurately. However, lower energy gamma emissions as well as MDA values may not be quantified if sample self absorption and shielding is significant.

Two additional enhancements to resolve problems resulting from random summing losses in high count rate samples and with an unknown sample source density is to place a transmission source behind the sample to correct for measurement losses encountered during sample counting. Also, the transmission source can also be utilized to determine source density based on attenuation through the sample.

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