SANDIA REPORT
SAND 2013-6475
Unlimited Release
Printed Month August 2013

MICROSHIELD / ISOCS
Gamma Modeling Comparison

[1]Sansone, Kenneth R

[1]Sandia National Laboratories, RPSD Laboratory, P.O. Box 5800, Albuquerque, NM 87185-1103

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico 87185 and Livermore, California 94550

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under Contract DE-AC04-94AL85000..

Approved for public release; further dissemination unlimited.

Sandia National Laboratories

Issued by Sandia National Laboratories, operated for the United States Department of Energy by Sandia Corporation.

NOTICE: This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government, nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, make any warranty, express or implied, or assume any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represent 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, any agency thereof, or any of their contractors or subcontractors. The views and opinions expressed herein do not necessarily state or reflect those of the United States Government, any agency thereof, or any of their contractors.

Printed in the United States of America. This report has been reproduced directly from the best available copy.

Available to DOE and DOE contractors from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831

Telephone: (865) 576-8401
Facsimile: (865) 576-5728
E-Mail: reports@adonis.osti.gov
Online ordering: http://www.osti.gov/bridge

Available to the public from
U.S. Department of Commerce
National Technical Information Service
5285 Port Royal Rd.
Springfield, VA 22161

Telephone: (800) 553-6847
Facsimile: (703) 605-6900
E-Mail: orders@ntis.fedworld.gov
Online order: http://www.ntis.gov/help/ordermethods.asp?loc=7-4-0#online

SANDIA REPORT
SAND 2013-6475
Unlimited Release
Printed Month August 2013

MICROSHIELD / ISOCS
Gamma Modeling Comparison

Kenneth R. Sansone
Radiation Protection Sample Diagnostics Program
Sandia National Laboratories
P.O. Box 5800
Albuquerque, NM 87185-1103

Abstract

Quantitative radiological analysis attempts to determine the quantity of activity or concentration of specific radionuclide(s) in a sample. Based upon the certified standards that are used to calibrate gamma spectral detectors, geometric similarities between sample shape and the calibration standards determine if the analysis results developed are qualitative or quantitative. A sample analyzed that does not mimic a calibrated sample geometry must be reported as a non-standard geometry and thus the results are considered qualitative and not quantitative.

MicroShield^R or ISOCS^R calibration software can be used to model non-standard geometric sample shapes in an effort to obtain a quantitative analytical result.

MicroShield^R and Canberra's ISOCS^R software contain several geometry templates that can provide accurate quantitative modeling for a variety of sample configurations. Included in the software are computational algorithms that are used to develop and calculate energy efficiency values for the modeled sample geometry which can then be used with conventional analysis methodology to calculate the result. The response of the analytical method and the sensitivity of the mechanical and electronic equipment to the radionuclide of interest must be calibrated, or standardized, using a calibrated radiological source that contains a known and certified amount of activity.

Table of Contents

Table of Contents 4
Nomenclature 4
Introduction 5
Hypothesis 6
Experiment 6
Results and Discussion 7
Errors Related to Modeling Codes 9
Conclusions 12
References 12
Distribution 13

Nomenclature

RPDP	Radiation Protection Dosimetry Program
RPSD	Radiation Protection Sample Diagnostics
Lc	Critical Level
MDA	Minimum Detectable Activity
RMWMF	Radiological and Mixed Waste Management Facility
ROI	Region Of Interest
HPGe	High Purity Germanium

Introduction

The Sandia National Laboratories Radiation Protection Sample Diagnostics (RPSD) laboratory is routinely requested to perform portable-field gamma spectral analysis on items that either contain or are suspected of containing radioactive material. The determination to perform a portable or field gamma spectral analysis over a gamma spectral analysis that is normally performed in the laboratory is usually due to either or both of two criteria:

1) The sample is too large to be accommodated by the sample staging area of standard laboratory gamma spectral analysis equipment;

2) The sample contains levels of activity that result in dead time percentages that exceed the analytical capability of the laboratory detector.

A sample analyzed with a portable gamma spectral system can vary in mass from several grams to hundreds of kilograms and the size can vary from a hot particle to the size of a transportainer. These samples can vary in both elemental and dimensional composition and usually exhibit non-standard geometric shapes. The source material itself can be of high or low density or completely exposed or contained. Attenuation of nuclide emissions from containment material may vary from less than 0.1 millimeter of polyethylene to several centimeters of lead. Variables such as distance between the gamma detector and the sample, source composition and density, and attenuation must be taken into account in conjunction with identifying the radionuclides(s) of interest to provide quantitative analytical results.

The majority of portable gamma spectral analysis performed are requested with qualitative results. Qualitative analysis are typically useful for isotopic determination (identification of radionuclides present) with estimated activity levels. There are situations however where precise quantitative results are requested by the customer. To provide quantitative analytical results from the analysis of non-standard geometries and to factor in all aforementioned variables such as shielding that may attenuate nuclide energy emissions between the source and detector, programs such as MicroShield^{R} and ISOCS^{R} can be used.

Hypothesis

MicroShield^R or ISOCS^R modeling codes can both provide quantitative activity values from modeling non-standard geometry samples with an acceptable degree of confidence.

Experiment

A 208 liter steel drum of radiological waste identified as D9001701was analyzed with a high purity germanium (HPGe) detector. The drum exhibited a high dose rate that resulted in dead time values that prevented an accurate analysis at the standard drum count distance of 91.44 cm. The drum was therefore positioned 526 cm from the detector face. The height center of the detector relative to the sample was 44.3 cm.

The spectrum from the analysis was subtracted from a background count (B1970913) performed in the identical location as the sample. The drum was modeled as 60% full, containing material that was a combination of 5% iron and 95% polyethylene. The net mass of the sample material was 4.2 kilograms.

The sample density (0.0308 g/cm^3) was calculated fractionally relating to the net mass of the sample. The activity was assumed to be uniformly distributed with a density equivalent to the calculated sample density.

The wall thickness of the drum was 0.1199 cm with an elemental composition of 60% iron, 20% manganese and 20% nickel.

Results and Discussion

The quantified activity of both modeled results is shown overlapped in Table 1.0. The activities developed from the MicroShield^R program were converted into picocuries / gram concentration so that a direct comparison could be made to the ISOCS^R results. Modeled results utilizing the MicroShield^R code resulted in slightly higher activity for most nuclides. Be-7 and Eu-155, which resulted with the lowest quantified activity of both codes, also exhibited the largest difference in activity. Activity for Cs-137 and Eu-154 were almost identical for both modeling codes. It is apparent that the nuclides with higher activity concentrations such as Cs-137 and Eu-154 resulted in a closer correlation with less error. Refer to Table1.0 and Chart A for a comparison of the activities calculated by both codes.
Refer to Table 2.0, which documents the percent difference and order of magnitude in activity between the two modeling codes.

Table 1.0 – Activity of Nuclides from ISOCS and MicroShield Modeling

Modeling Code	Nuclide	Activity (pCi / gram)
MicroShieldR / ISOCS	Am-241	5.12E5 / 4.90E5
MicroShieldR / ISOCS	Be-7	9.81E3 / 1.04E4
MicroShieldR / ISOCS	Eu-154	8.21E5 / 8.24E5
MicroShieldR / ISOCS	Eu-155	1.00E5 / 8.40E4
MicroShieldR / ISOCS	Cs-137	1.46E6 / 1.45E6

Chart A. Plotted Activity of Nuclides from ISOCS and MicroShield Modeling

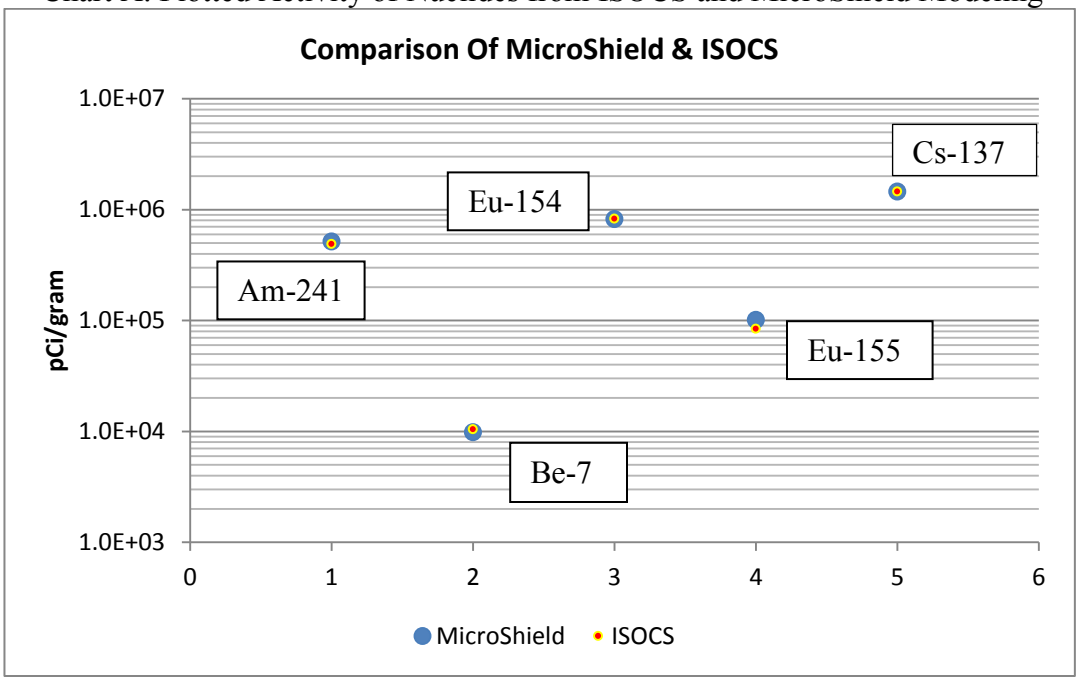

Table 2.0 – Difference & Magnitude of Nuclide Activity from ISOCS and MicroShield Modeling

Nuclide	Difference between models	Activity Magnitude (pCi/g)
Cs-137	0.685%	~1.5E6
Eu-154	0.364%	~8E5
Am-241	4.297%	~5E5
Be-7	5.673%	~1E4
Eu-155	16.000%	~1E5

Errors Related to Modeling Codes

MicroShield

For the method utilizing the MicroShield modeling code, the degree of uncertainty is expressed as an Accuracy Factor. The accuracy factor compares the unattenuated gamma fluence rate per unit activity between a uniform activity distribution ("reasonable attenuation" model) and an assumed activity distribution represented by a line source in the center of the sample ("high attenuation" model). A line source activity distribution was selected as the "high attenuation" model since this geometry provides significant gamma ray attenuation through the sample matrix to provide reasonably conservative estimates of system accuracy. Using this method, larger accuracy factors indicate greater uncertainty. Please note that the accuracy factor only considers the uncertainty associated with the activity distribution and does not consider the uncertainty associated with matrix composition, container fill height, or counting statistics.

Chart B, below, illustrates the generated report using the MicroShield [R] program. The program calculates and documents the uniform and line source estimated activity.

The MicroShield accuracy factor is displayed in the far right column of the generated report.

Chart B. MicroShield Results

Parent Nuclide	Nuclide	Energy (keV)	Yield γps/dps	Net Counts	Continuum Counts	Case 1 Uniform Source Unattenuated Gamma Fluence Rate per Ci at Aperature (γ/cm²-sec)/Ci	Case 2 Line Source Unattenuated Gamma Fluence Rate per Ci at Aperature (γ/cm²-sec)/Ci	Case 3 User Specified Unattenuated Gamma Fluence Rate per Ci at Aperature (γ/cm²-sec)/Ci	Efficiency (cps per (γ/cm²-sec)	Case 1 Uniform Source Ci Estimated Activity	Case 1 Uniform Source Ci Estimated Critical Level	Case 2 Line Source Ci Estimated Activity	Case 2 Line Source Ci Estimated Critical Level	Accuracy Factor
Am-241	Am-241	59.536	3.570E-01	158421		7.15E+02	8.47E+02		2.86E+01	2.15E-03		1.81E-03		0.84
Be-7	Be-7	477.593	1.042E-01	1532		7.91E+02	7.95E+02		1.30E+01	4.12E-05		4.11E-05		1.00
Cs-137	Cs-137	667.660	8.521E-01	1478320		6.68E+03	6.71E+03		1.00E+01	6.14E-03		6.12E-03		1.00
Eu-154	Eu-154	873.230	1.145E-01	93466		9.19E+02	9.22E+02		8.19E+00	3.45E-03		3.44E-03		1.00
E-155	Eu-155	86.545	3.090E-01	70340		1.36E+03	1.45E+03		3.42E+01	4.21E-04		3.93E-04		0.93

ISOCS

Computational errors developed from ISOCS[R] are displayed as a 95.5% confidence level or 2 standard deviations (2-sigma error) in the reanalyzed report (Chart C) utilizing Genie 2k Algorithms ignoring geometry and activity distribution.

Chart C. ISOCS Results

```
*************************************************************
*                    Sandia National Laboratories
*    Radiation Protection Sample Diagnostics Program
*                      11/19/08  9:57:11 AM
*************************************************************
*
* Analyzed by:                          Reviewed by:
*************************************************************
Customer                  : LONGLEY J
Customer Sample ID        : C200340 (P2000939)
Lab Sample ID             : D9001701
Sample Description        : 55 GALLON DRUM CONTAINING SOLIDS
Sample Quantity           :  4200.000  gram
Sample Date/Time          : 9/13/07   10:55:09 AM
Acquire Start Date/Time   : 9/13/07   10:55:09 AM
Detector Name             : PGE01
Procedure Number          : RPSD-09-01
Elapsed Live/Real Time    :   3600 /   3990 seconds

Comments:
UNCALIBRATED GEOMETRY. ALL VALUES ESTIMATED. FOR ID ONLY.
*************************************************************
```

Nuclide Name	Activity (pCi/gram)	2-sigma Error	MDA (pCi/gram)
U-238	Not Detected	---------	3.48E+004
RA-226	Not Detected	---------	4.23E+004
PB-214	Not Detected	---------	3.44E+003
BI-214	Not Detected	---------	2.41E+003
PB-210	Not Detected	---------	6.11E+005
TH-232	Not Detected	---------	1.12E+004
RA-228	Not Detected	---------	4.95E+003
AC-228	Not Detected	---------	2.96E+003
TH-228	Not Detected	---------	3.71E+004
RA-224	Not Detected	---------	6.97E+002
PB-212	Not Detected	---------	2.83E+003
BI-212	Not Detected	---------	3.07E+004
TL-208	2.33E+004	3.88E+003	3.22E+003
U-235	Not Detected	---------	1.37E+004
TH-231	Not Detected	---------	5.28E+005
PA-231	Not Detected	---------	7.39E+004
TH-227	Not Detected	---------	1.11E+004
RA-223	Not Detected	---------	8.74E+003
RN-219	Not Detected	---------	2.07E+004
PB-211	Not Detected	---------	4.66E+004
TL-207	Not Detected	---------	3.77E+005
AM-241	4.90E+005	1.18E+005	9.75E+003
PU-239	Not Detected	---------	2.37E+007
NP-237	Not Detected	---------	1.52E+005
PA-233	Not Detected	---------	3.28E+003
TH-229	Not Detected	---------	1.43E+004

The ISOCS error is displayed in the third column of the generated report.

Calculations for the derived 2-sigma error in the Genie 2K program are as follows:

The random uncertainty of the activity, C, is calculated as

$$\sigma_C = C \cdot \sqrt{\left(\frac{\sigma_R}{100}\right)^2 + \left(\frac{\sigma_S}{S}\right)^2 + \left(\frac{\sigma_V}{V}\right)^2 + \left(\frac{\sigma_{\varepsilon'}}{\varepsilon'}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2 + \left(\frac{\sigma_K}{K}\right)^2}$$

where

σ_R is the user defined random uncertainty (%),

σ_S is the uncertainty of the net peak area S,

σ_V is the uncertainty of the sample quantity V,

$\sigma_{\varepsilon'}$ is the uncertainty of the effective efficiency, which is defined a3

$$\sigma_{\varepsilon'} = \varepsilon' \cdot \sqrt{(\sigma_\varepsilon / \varepsilon)^2 + (\rho t \cdot \sigma_{\mu(E)})^2 + (\mu(E) \cdot \sigma_{\rho t})^2}$$

where ε is the non-attenuation corrected detection efficiency at the peak energy in question, σ_ε is its uncertainty, $\mu(E)$ is the mass attenuation (in units of cm^2/g) at gamma energy E, $\sigma_{\mu(E)}$ is its uncertainty, $\rho\tau$ is the average sample mass per unit area, and $\sigma_{\rho t}$ is its uncertainty.

σ_y is the uncertainty of the branching ratio y, and

σ_K is the uncertainty of the composite decay correction factor K,

And finally, the *total* uncertainty of the activity, C, is calculate

$$\sigma_{C(T)} = \sigma_C + \frac{\sigma_{sys} \cdot C}{100}$$

where σ_{sys} is the user defined systematic uncertainty (%).

Conclusion

The developed activities involve a number of assumptions. In particular, the activity was assumed to be uniform with a density equivalent to the calculated effective sample density of 95% Polyethylene and 5% Iron. Additionally, low energy emissions are sensitive to the accuracy of the modeled geometry. 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 normally associated with the activity calculations involving high-energy gamma ray emissions. Accordingly, the nuclide activity results are presented for a high-energy gamma ray emission with a high yield when possible. Also, daughter and/or parents will also be present in equilibrium depending on the half-life of the nuclides involved.

There was very good correlation between the activity concentration values developed from both codes. In conclusion it is to be assumed that using either of the two modeling codes in conjunction with proper counting methodology to quantify gamma spectrums obtained from the analysis of non-standardized or non-calibrated geometry samples can provide similar results.

References

- Shleien, Bernard, The Health Physics and Radiological Health Handbook, Revised Edition, 1992.
- Gosselin, Smith, and Hodge, Clinical Toxicology of Commercial Products, Fifth Edition.
- Kocher, David C., Radioactive Decay Data Tables A Handbook of Decay Data for Application to Radiation Dosimetry and Radiological Assessments, DOE-TIC-11026, 1981.
- Brown, E., Firestone, R.B. Table of Radioactive Isotopes 1986, John Wiley & Sons, Inc.
- Frame, Gleason, Worthington, Nuclide Identification Catalog for Gamma Emitters and Alpha Emitters, ORAU 88/K-32, First Edition, Revised November 1989.
- Lewis, Richard J. Sr., Sax, N.Irving, Hawley's Condensed Chemical Dictionary, Eleventh Edition, 1987.
- Ebara, S.B.; Enghauser, M.W. Quantitative portable gamma spectroscopy sample analysis for non-standard sample geometries, Journal of Radioanalytical and Nuclear Chemistry, Vol. 233, Nos 1-2 (1998) 273-279.
- MicroShieldR 5.02. Grove Engineering, Inc. Rockville, MD.
- Genie 2k Spectroscopy Software Customization Tool Manual Copyright 2009, Canberra Industries

Distribution

1	MS 1103	K. Sansone, 4121
1	MS 1103	S. Shanks, 4121
1	MS 1103	B. Hanson, 4121
1	MS 1103	A. Bendure, 4121
1	MS 0729	T. Cooper, 4120
1	MS 0725	S. Gutierrez, 4100
1	MS 0899	Technical Library, 9536 (electronic copy)

This page is intentionally left blank

Sandia National Laboratories