

Practical Solutions to Radioactive Waste Characterization

Ronald Keyser, Timothy Twomey
EG&G ORTEC

Richard Hagenauer
Lockheed Martin Energy Systems, Oak Ridge, TN

Abstract

The movement or storage of containers of radioactive waste requires characterization of the contents for radioactivity to verify conformance with transportation and waste acceptance controls. Spectroscopy of the emitted gamma rays is often the only viable method of assay of the contents. The spectrum analysis must include suitable efficiency, attenuation and geometry corrections for bulk containers. Program **ISOTOPIC** performs this analysis in a straightforward and simple manner without using complex modeling of the HPGe detector. The operator interface for Windows 95/98/NT will be described and recent results presented.

Introduction

The analysis of poorly characterized material such as found in nuclear facility waste streams or in "holdup" measurements of special nuclear material by gamma spectroscopy is often difficult. The reason is that it is often impossible to find a standard which accurately represents the geometrical and absorption effects (container and matrix) which modify the spectrum actually seen by the detector.

One solution is to use calibrations derived from Monte-Carlo modeling of the entire container-detector configuration. However, while mathematically elegant, this approach may restrict the user to a specifically pre-calibrated detector and does not address the problems associated with the variable matrix in the container. The pre-calibration procedure is computationally intensive and thus is costly, and has the disadvantage that the system must be used only with pre-calibrated detectors, which may become unavailable or non-functional.

Program **ISOTOPIC** has been developed at the Oak Ridge East Tennessee Technology Park (ETTP) facility (formerly called K-25 site), to provide a more practical solution to this class of problem which greatly simplifies the calibration problem. Using only a single point source for a primary calibration, any needed secondary calibration may be generated for a HPGe detector, accounting for geometry, container wall and matrix absorption via an internal data-base of materials and geometry coefficients. Collimator effects are also taken into account.

Correction for Matrix Gamma Ray attenuation

Correction for gamma-ray attenuation is the major problem which is encountered in the assay of waste containers. If there are lumps in waste or holdup deposits, it is difficult to determine the distance the gamma rays travel through (1) low-density material, (2) high-density material, and (3) empty space, before leaving the sample.

Two techniques utilizing gamma-ray spectroscopy can be used to measure inhomogeneous material or waste including (1) the use of differential attenuation of gamma rays and (2) the use of a transmission source to determine and correct for attenuation. Program **ISOTOPIC** employs the first approach.

By looking at the differential attenuation of the gamma rays of different energies being emitted from the same nuclide, Program **ISOTOPIC** can estimate the attenuation of the gamma rays. Only the material between the source and the detector is considered, because this is the only absorbing material. The advantage is that the same gamma rays are being used to estimate the attenuation and quantify the nuclides. The program starts with a report file from a peak-fitting program, models the geometry, corrects for matrix, container, and self-attenuation, and computes the activity. The analyst may interactively adjust density and thickness parameters to most closely match the matrix that the gamma rays actually penetrate.

When calculating the density of the contents of a container by using the weight and volume of the container, the container is assumed by **ISOTOPIC** to be full. If it is not completely full, then the actual material density will be higher than the calculated density. If a nuclide that emits several gamma rays at different energies is present, then the analyst will be able to correct for the different density using Program **ISOTOPIC**. Good examples are (1) uranium with several relatively intense gamma rays from ^{235}U ranging in energy from 84 to 205 keV and (2) several other gamma rays from ^{238}U daughters ranging from 92 to 1001 keV.

For large containers, the matrix attenuation corrections are usually the major source of error. If only a few gamma rays of different energies are present, it becomes difficult to get an accurate estimate of the correction factors. If the distribution of nuclides is not uniform, one nuclide may be near the detector, penetrate a minimum matrix, and exhibit a small correction. Another nuclide may be located farther from the detector and penetrate a larger thickness of the matrix. Since the attenuations of the gamma rays of the two nuclides are different, the analyst will have to choose which nuclide to use for the overall matrix correction. Usually, the nuclide with the more penetrating gamma rays is the best one to select for overall matrix attenuation.

Correction for GEOMETRY

Program **ISOTOPIC** takes the point-source approach and generalizes it over a variety of geometries. The container is assumed to be uniformly filled with point sources, and the measured activity is assumed to come uniformly from each of these point sources. The program effectively integrates over all the point sources while correcting each one for matrix-attenuation, self-attenuation, container wall thickness, and distance from the detector (detector offset). The analyst may choose from three different geometries. The point source geometry is for localized deposits in process equipment, small sources, hot spots in a waste drum or box, or boxes and cylinders counted a large distance away. The cylinder geometry can be used for containers ranging from juice cans to 416-liter (110-gal) or larger drums. The cylinder geometry also can be used to model pipes or tanks that are part of process equipment. The box geometry can be used for rectangular boxes up to the 1.2 x 1.2 x 1.8 m (4 x 4 x 6 ft) ST-5 box (B-25 box). Any item of an approximately rectangular shape can be modeled with the box geometry. A long small-diameter pipe is a long small-diameter cylinder, and contaminated walls or floors can be modeled as very thin boxes. The main distinguishing feature separating the cylinder and the box geometries is the curved versus flat shape of the surface facing the detector.

Container Rotation

Containers often do not have the radioactive material distributed uniformly throughout the volume. If this is true and the higher concentration of radioactive material is near the detector, the measured results will be biased high. If the higher concentration is away from the detector, the results will be biased low. Rotation of the container will help reduce the bias resulting from varying concentrations of radioactive material. The cylinder geometry portion of Program **ISOTOPIC** has a box to be checked to reduce the estimated uncertainty if the container is rotated.

Other Sources of Error

Other important sources of error include random errors (e.g., poor counting statistics and changing gamma-ray background) and systematic errors (e.g., in the peak fitting routine, published branching ratios, and the assigned ^{235}U enrichment). In most cases, except very low-level waste, counting statistics are not a significant source of error for the primary gamma ray used for quantification. However, if the attenuation corrections are determined using weaker gamma-rays from a nuclide, the attenuation error will be reduced with additional counting time, because additional peaks are helpful in determining the best matrix correction. If only a few peaks are available to adjust the attenuation correction, the density calculated from knowledge of the weight and volume of the container should be used. Usually, the largest sources of error are attenuation corrections, geometry corrections, counting statistics, and lack of secular equilibrium.

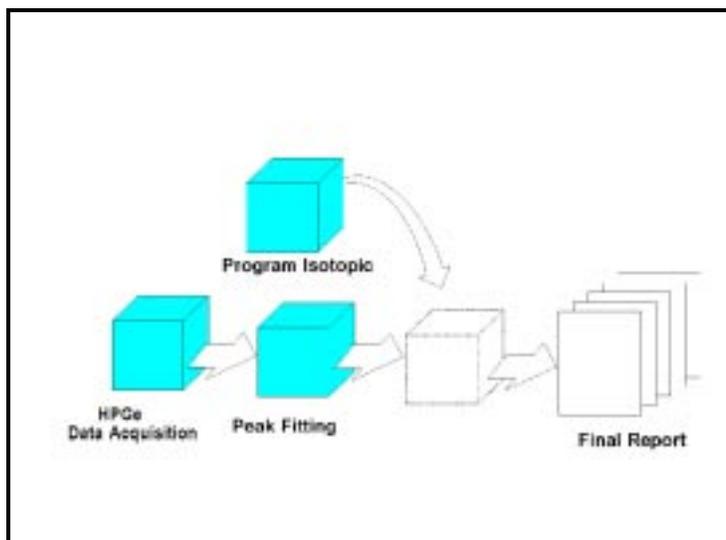
English and metric units can be chosen for the mass and length parameters. A wide variety of activity units can be chosen for reporting including: Ci, bq, bq/kg and many others. Densities are always reported in units of g/cc. Results are always reported in units of grams plus the user selected activity units.

ISOTOPIC in Use

Program **ISOTOPIC** is a 32-bit application, which runs in conjunction with the EG&G ORTEC GammaVision-32 program for HPGe detector Gamma-Ray analysis. GammaVision-32 itself is a member of the EG&G ORTEC **CONNECTIONS-32** suite of software products.

CONNECTIONS is a Y2K compliant, truly multi-tasking, 32-bit spectroscopy platform based on Windows 95/98/NT. The **CONNECTIONS** concept is “see anything, do anything, be anywhere, using any hardware.” **CONNECTIONS** is secure, standard and open. Multiple **CONNECTIONS** applications may co-exist on the network, and interface simply with custom report writers and industry standard data bases; controlling and viewing all attached spectroscopy hardware activity from any workstation. Programmer toolkits facilitate custom development. **CONNECTIONS** hardware support includes all current and past EG&G ORTEC MCA devices and also an increasing number of non-ORTEC types.

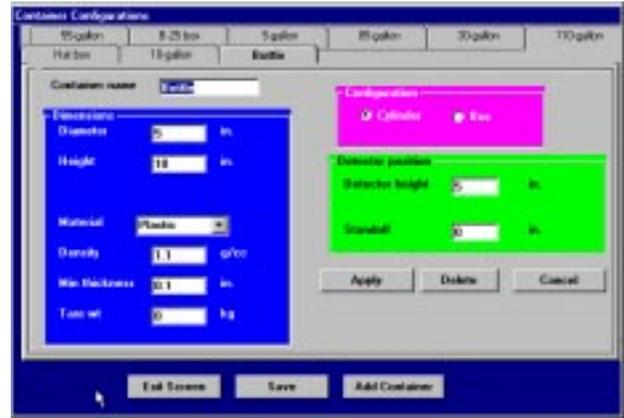
GammaVision-32 controls the acquisition, saves the spectrum and performs the peak analysis. Program **ISOTOPIC**, acting as a post processor then corrects for the matrix, container, collimator and geometry effects using the operator settings and internal databases.



Program **ISOTOPIC** is the missing piece between peak fitting and waste analyses.

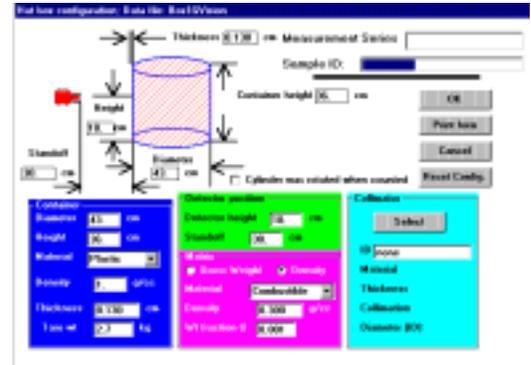
User Interface

The **ISOTOPIC** user interface conforms to the usual norms of modern Windows programming. It may be configured to handle a large number of standard pre-specified Geometry and Matrix scenarios. If a series of measurements will be made in the same configuration, these settings can be saved for more analyses. Up to 12 standard configurations may be saved to simplify repetitive analyses. Objects such as metal plates, and surfaces such as walls, can be handled as well as boxes and drums.

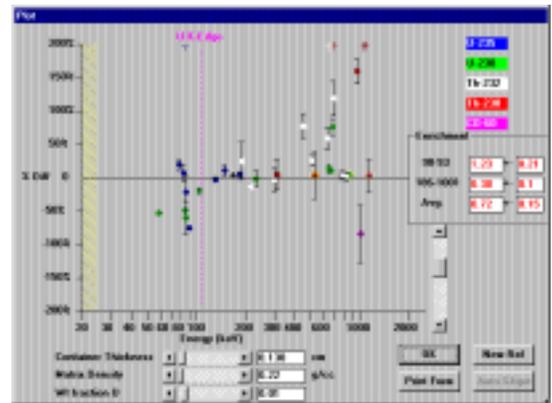


After the spectrum acquisition and peak analysis is completed, the operator interactively specifies the measurement configuration. Dimensions and materials are entered.

After the operator has specified the container parameters, **ISOTOPIC** produces the Isotope plot shown at the right. This shows the percentage difference between the corrected measured activity and the activity calculated for the user-specified “reference peak” for that nuclide. The reference gamma ray is positioned on the base line (% Dif 0).



If uranium is present, the “auto slope” feature can calculate the effects of the container and matrix by fitting the U-238 lines to Zero slope. The operator now optimizes the analysis interactively: adjusting the container, matrix and weight fraction uranium, to refine the results. The visual evidence of a good result is immediately seen when the points from a multi-peaked isotope are distributed normally about the “zero-line.” If the U-235 enrichment is known, then the U-238 and U-234 values can be computed more accurately for samples containing weak uranium activity.

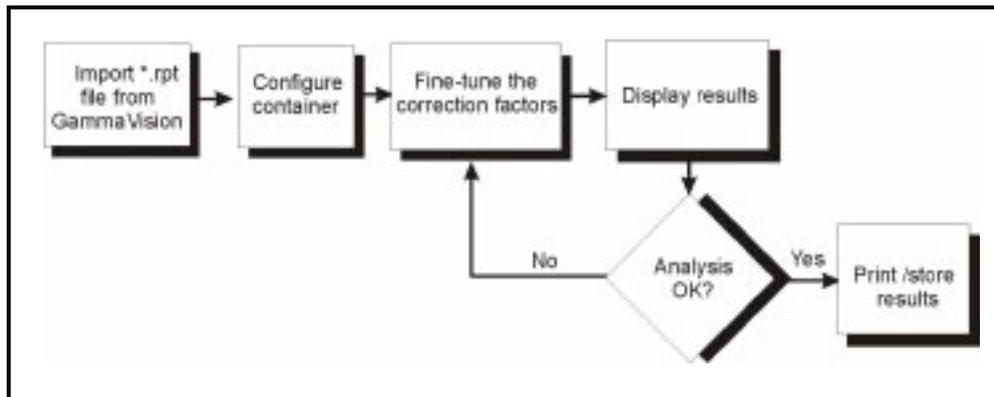


The Isotope plot can provide valuable other information. For example, that the isotopes may not be uniformly distributed within the sample. Homogenous and inhomogeneous samples may be analyzed by this method. A good indication that a package contains an inhomogeneous distribution of materials may be the fact that the user can obtain a combination of parameters which render the activity plot flat for some but not all nuclides. If a situation of “infinite thickness” exists which would cause results below a certain energy to be systematically low, the operator is alerted by “hatching” on the isotope plot.

When the fine tuning is finished the operator may select a report for each isotope showing the activity and weight. These results may then be printed and archived. The report files are written in either a database summary or as complete reports showing all input and correction information.



Investigation of the correction factors can help the user decide whether counting longer is likely to significantly improve the precision of the result. The user is also warned if any of the corrections appear to be excessively large.



Analysis flow chart for Program ISOTOPIC

Modeling with ISOTOPIC

ISOTOPIC allows the operator to simulate a wide range of geometry, matrix, and container situations. By using the background spectrum and treating it as if it was sample, the user can compute the minimum detectable activity (MDA) over any requested density for the given detector and chosen measurement geometry. The nuclide of interest is put into the gamma ray library so the MDA can be computed from the peak-fitting routines. Program **ISOTOPIC** will then provide the proper correction factors for geometry, matrix, and container and report the corrected MDA's.

Expected analysis uncertainties

Due to the often diverse characteristics of samples to be measured, large uncertainties (precision and bias) are not uncommon. Uncertainties due to small-scale matrix inhomogeneties, and consequential local geometrical effects can entirely overwhelm the uncertainty in the primary detector calibration. (This point serves only to emphasize the fact that expensive and detector-specific computer-modeling of detector primary efficiency calibrations only complicate the measurement process, with no “payback” in improved precision.) Generally, the uncertainty increases with density or container size. Table 1 lists some expected uncertainties for two container sizes. These are only approximations. Other waste sample measurements may vary.

Table 1. Typical Uncertainties Expected for Nondestructive Measurements			Sensitivity (MDA) 30 Minute count 60% relative efficiency HPGe detector at 18" distance		
Container	Density (g/cc)	Overall Uncertainty	U-235 (g)	U-238 (g)	Cs-137 (nCi)
B-25 box	< 0.1	30%	8.0E-3	5.0E-1	6.0E+00
	0.1 - 0.8	50%	4.0E-02	1.5E+00	2.0E+01
	> 0.8	70%	5.0E-02	2.0E+00	4.0E+01
Cylinders	< 0.1- <0.3	30%	1.8E-03	2.5E-01	1.5E+00
	0.3 - 1.0	50%	6.0E-03	5.0E-01	4.0E+00
	> 1.0	70%	1.0E-02	1.0E+00	7.0E+00

Measurement comparisons

Generally, it is difficult to obtain measurement comparisons between destructive and nondestructive methods, because it is prohibitively expensive to destructively analyze a sample. For low-density samples the sampling error used to select samples for destructive analyses is very large. The chemical concentration differences among subsamples usually dominate the differences between destructive and nondestructive results.

At ETTP there has been some chemical sampling for containers holding larger concentrations of uranium. These 5-gallon buckets contained U occluded on Al_2O_3 obtained from trap material used in the gaseous diffusion process. The comparisons between destructive and nondestructive results are shown in Table 2 for ^{235}U enrichment total uranium. A report of intercomparisons between NDA techniques using passive-gamma spectroscopy and K-edge densitometry for piping show good agreement between the techniques.

Sample ID	NDA Results		Chemistry Results		% Difference	
	U(g)	^{235}U (wt%)	U(g)	^{235}U (wt%)	U(g)	^{235}U (wt%)
4094	880.	0.42	751.	0.53	17.2	-26.2
4095	330.	0.55	333.	0.30	-0.9	45.5
4096	130.	1.97	163.	1.84	-20.2	6.6
4097	273.	0.33	409.	0.24	-33.3	27.3
5718	231.	0.89	193.	1.04	19.7	-16.9
5720	2870.	1.80	2797.	1.75	2.6	2.8
Avg.					-9.9	6.6
Avg. (abs)					15.6	20.8

New Enhancements for Version 2

Changes from Version 1 to Version 2 have provided additional convenience and scientific information.

Additional scientific information:

1. A breakdown of the estimate of errors for each correction component gives users a chance to improve their results by giving more attention to the largest problem area. Error estimates for geometry are reduced when the sample is rotated.
2. U-235/U-234 ratios can be used when these ratios are known. The U-234 activity is usually the dominate isotope activity in uranium enriched in U-235.
3. Units of Bq and Bq/kg have been added for the convenience of the European community.
4. Warning messages are now given when (1) the reference peak area is missing, (2) the measurement is below the computed MDA, or (3) the reference peak computed the nuclide mass when "infinitely thick" at the reference energy. The user then decides if the determined nuclide activity should be deleted or some other adjustment made.
5. Peaks areas that are apparently "spurious" can be automatically accumulated with "spurious" peaks from other spectra. Later, the peaks are collated. Peaks that reoccur are most likely peaks from an unidentified nuclide.

Additional convenience features:

- 1 Program ISOTOPIC is now a 32-bit program with support for long file names.
- 2 Dimensions for 12 different user box or cylinder configurations can be stored and recalled.
- 3 Weight information can be added within GammaVision and automatically utilized by Program ISOTOPIC.
6. The GammaVision gamma-ray library can now be read directly into Program ISOTOPIC. No ASCII conversion is necessary.
7. A browse feature is available to facilitate file path entries.

Conclusion

Program **ISOTOPIC** is a flexible analysis tool for use in a wide variety of container/object counting situations. It may be used with any available HPGe detector, is easy to use, and by the use of interactive graphics screens, permits the user to simply optimize the analysis results. In many measurement situations, **ISOTOPIC** represents a simple and effective analysis approach which can yield high sample throughput and operational cost savings.