

## The Whys and Wherefores of Measuring Drum Activity with ISO-CART™ and the program ISOTOPIC

Decommissioning, decontamination, and normal processing of nuclear waste generates quantities of radioactive material in the form of many different matrices stored in many different sized containers. These containers must be measured to assay their radioactive content before moving, shipping offsite, burying, or placing in a storage area. Within their wide range of spectroscopy systems and products, ORTEC offers two products of great utility in this area, which specifically provide simple and cost-effective solutions to these gamma-ray measurement problems.

The first product is a software program ISOTOPIC which solves the problems associated with sample matrices and geometries to determine activities. ISOTOPIC is added to existing gamma-ray spectroscopy systems using the ORTEC GammaVision® analysis package<sup>1</sup>.

The second product is the ORTEC ISO-CART mobile assay system. It provides a “turn-key,” cart-mounted, easy-to-use solution. All necessary hardware, including detector, shielding, collimator, MCA, computer, CART and software are included. The ISO-CART is supplied from the ORTEC factory ready to go to work. User training is available on the use of the ISOTOPIC software or the complete ISO-CART system.

### Intended Audience

This application note is intended for those who already own or are contemplating deploying either of these products for the first time. It provides a simplified procedure and “helpful hints” on the practicalities of these measurements. It is applicable to users of both the ISO-CART system or with “home built” gamma-spectroscopy systems incorporating ISOTOPIC and GammaVision software. The hints refer to “typical situations” based on extensive experience in measuring actual samples of this type. Before describing specific steps required to make the measurement, some consideration is given to the choice of equipment which will result in good quality, reliable results from such a system. The note concludes with a summary section of “frequently asked questions” relating to the application of ISOTOPIC to measurement problems.

### Choice of Equipment

The quality of the results can be strongly influenced by the choice of equipment, both in terms of hardware and of software. Even when samples are in difficult measurement situations, with high attenuation, and nonhomogeneity, a poor choice of equipment will emphasize the situation and make results imprecise and unreliable. Equipment should be chosen with care.

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<sup>1</sup>The ISOTOPIC Program runs in conjunction with ORTEC GammaVision-32 for HPGe detector gamma-ray analysis. GammaVision controls the acquisition, saves the spectrum and does the peak analysis. ISOTOPIC then “post processes” the results further to correct for matrix, container, collimator and geometry effects using operator-supplied information about dimensions, weights, distances and materials, along with internal databases of geometry and matrix correction information.

## HPGe Detector

A HPGe detector with an efficiency of at least 50% relative<sup>2</sup> is needed for low-level waste items. This detector is needed in order to separate the many gamma-rays and to have the sensitivity for the required detection limits. Larger efficiency HPGe detectors have higher peak-to-Compton ratios than smaller ones. A detector with a high peak-to-Compton ratio will provide a higher peak to background ratio at lower energies, improving lower limits of detection (Ref. 1). If most of the gamma-rays of interest are below 1.0 MeV, a detector with a larger diameter (for the same relative efficiency) will be more sensitive (Ref. 2). Most often, p-type (GEM) detectors are used for the gamma-ray detection. These detectors are useful for the measurement of gamma-ray energies down to about 60 keV. Most gamma-rays below this energy are absorbed in the matrix or wall of the drum.

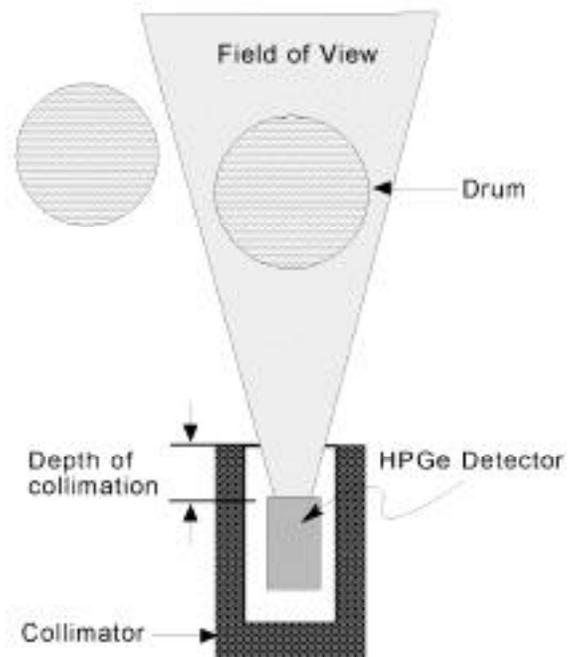
## Signal Processing Electronics

In many cases the drums are measured in an unheated building where the temperature can change significantly during the measurement period. Stability of measurement electronics is clearly important, and the use of digital stabilizers should be considered in extreme cases.

The ORTEC DART<sup>®</sup> portable MCA has been deployed with a high degree of success in a range of mobile applications, and is utilized as a standard component in the ISO-CART Mobile Assay System. DART is in use in mobile NDA measurements world-wide. For in-plant applications, where mains power is available, the ORTEC DSPEC<sup>®</sup> and DSPEC<sup>PLUS</sup><sup>®</sup> offers unsurpassed stability and count-rate performance. (Separate data sheets and technical papers on these products are available on request.)

## Collimator Considerations

Usually, waste drums are stored in a central location to be measured. The radioactivity in these drums may increase the background radiation field in which an individual drum is to be measured. To minimize the effect of background radiation, use a collimator to shield the detector and limit the field-of-view to a single drum. The wall of the collimator should be at least 1.6 cm (5/8") thick lead or equivalent stopping power. If additional activity is present in the measurement area, the collimator should extend from 2.5 cm (1") behind the detector and 2.5 cm (1") or greater in front of the detector. See Figure 1. The collimator can be made of tungsten or steel instead



**Figure 1. Use a collimator to establish a Field of View.**

<sup>2</sup>Relative efficiency is defined relative to a 3" by 3" NaI detector, according to ANSI/IEEE 325-1996.

**Table 1. Stopping Power for Common Materials Used for Collimators**

Material	Fluorescent X Rays (keV)	Percent of Gamma-Rays Absorbed by 2 cm of Material		
		100 keV	500 keV	1000 keV
Lead	73-75	~100	97.4	79.0
Tungsten	58-59	~100	99.3	91.5
Steel	6.4	99.5	72.5	60.6

of lead. Table 1 shows the popular collimator materials and their stopping power at selected energies. If the fluorescence x rays from the collimator material overlap other nuclide reference gamma-ray peaks and interfere with the analysis of the container nuclides, the collimator should be lined with cadmium, tin, or copper to adsorb these x rays. Because tungsten is so dense, it is the best material for fabricating collimators. However, when cost is taken into account, lead may be an attractive alternative. A practical collimator (for stopping power and weight) is 2 cm of lead with 2 mm of tin or cadmium and 1 mm of copper on the inside.

### **The ORTEC ISO-CART**

The ORTEC ISO-CART Mobile Assay System (Fig. 2) is an ideal solution when the measuring instruments have to be frequently moved. It provides all necessary hardware and software, with a choice of detectors and collimators in a highly mobile format. ISO-CART features the latest ORTEC HPGe detector technology with a choice of efficiencies and collimators. The highly popular DART high-performance portable MCA, is configured with GammaVision® and ISOTOPIC software. ISO-CART's large wheels make it easy to maneuver on uneven surfaces. A separate data sheet is available on request.



**Figure 2. The ORTEC ISO-CART™ is an ideal solution for portable measurement instruments.**

## **Calibration of Equipment and Validation of Method**

The HPGe detector system is calibrated for energy and efficiency (necessary for nuclide quantification) using a NIST-traceable (or equivalent) mixed nuclide point source. The ISOTOPIC software uses this point source calibration to model the efficiency for the item being measured. This calibration provides the relationship between measured count-rate and the actual activity (becquerels or microcuries). It should be noted that ANY system of this type will require at least an energy calibration with a point source. The advantage of the ISOTOPIC approach is its simplicity. The detector may be calibrated for energy and efficiency at the same time using the same point source. This can be an entirely automated procedure because of the capabilities of GammaVision. Internally, ISOTOPIC contains all of the data needed to extrapolate from the point source to the actual measurement situation. (The procedure is effective for any HPGe detector. It is not necessary to employ special "vendor precalibrated" detectors.) The calibration source should contain gamma-rays that span the energy region of interest. For most applications, suitable standards are easily obtained.

The overall system (including hardware and analysis software) should be validated by preparing standards containing: (1) activity traceable to a national standard and (2) matrices (either simulated or real) similar to that being analyzed. These simulated samples should be analyzed as if real and results compared with the known activity values of the standards. The overall accuracy of measurements can be established by this means.

# Data Taking

Below is a step by step procedure covering the process of data taking.

## 1. Record the background

Record a background measurement. If the background does not change due to movement of radioactivity nearby, one background measurement should suffice for the entire measurement period. Take a background measurement with a "blank" container in front of the detector. (A blank container contains material that is not radioactive but has similar weight to a typical sample.) If no container or an empty container is used to measure the background, then the recorded background will tend to be artificially high. The sample material and container reduce the background by absorbing many of the background gamma-rays. The thickness of the collimator may need to be increased if there are high energy gamma-rays from a nearby source that will interfere with the analysis. Some nuclear facilities use a portable shield behind the detector when backgrounds change in that direction.

## 2. Record the sample and container weights

Record the weight of the sample and tare (empty container) for the estimate of matrix density. The use of transmission sources to estimate the density at only one or two locations along the height of the container has no advantage over the use of sample weight to compute density, because there may be an unusually large air pocket or localized high attenuation in the path of the transmission source beam. The activity within the sample may see different attenuation properties than the transmission source.

## 3. Look for "hot spots"

If it is possible or likely that the container includes "hot spots," scan it with a 3" x 3" or larger hand-held NaI detector to locate them. If this procedure indicates that a "hot spot" is indeed present such that ~75% of the activity is there, then proceed as in step 6 below. If no hot spot was located, proceed with step 4.

## 4. Position your detector

Position the detector approximately one drum diameter from the sidewall so the entire drum is within the collimator field-of-view. For a 208-liter (55 U.S.-gallon) drum a distance of 46 cm (18 inches) is recommended. Closer distances will reduce counting times but increase geometrical errors. If the drum is only half full the detector should be positioned at the center of mass of the waste. ISOTOPIC "scales" the activity which is "seen" to the full size of the object as specified on the parameter entry screens. Therefore, if the container is known to be half-full, it should be described as half the actual height.

## 5. Start the count

Start out by counting the item for 30 minutes (typical). Items that indicate high activity may be counted as quickly as 5 minutes. Items with high density and very low activity may require additional counting time. Shorter counting times are possible if a higher efficiency detector (70% to 100% relative) is used. The ORTEC GammaVision software has a useful "count to MDA" feature. "Count to MDA" minimizes the count time necessary to achieve the desired detection limit for a specified nuclide. If the material in the drum is nonhomogeneous, then use a turntable to rotate it. Rotating the drum helps simulate an average matrix attenuation. Most solid items that need measurements are nonhomogeneous. The sample should be rotated several times during the counting period. Inexpensive turntables for 208-liter drums are available from ORTEC. Containers that are smaller than a 208-liter drum can be positioned closer to the detector. If the entire drum is assumed to contain radioactivity, then the entire container should be within the detector field-of-view.

## 6. Special considerations for hot spots

There are some special considerations for "hot spots" to improve the results. If step 3 above indicates that a "hot spot" is present such that 75% of the activity is represented by that "hot spot," then position the detector close to the "hot spot," but not less than 30 cm away from the side of the drum. Measure and record the distance from the "hot spot" to the

detector and treat the item as a point source for analysis purposes. Do not rotate the sample. A second measurement may be made at a distance of the drum diameter, in the usual way. After analysis, the two results are compared or averaged to give the best estimate of the activity in this non-homogeneously filled drum.

## Analysis

Before we describe the simple step by step analysis procedure, we will explain how GammaVision and ISOTOPIC interact:

GammaVision is the ORTEC peak-fitting software for general purpose gamma-ray analysis of spectra taken with a HPGe detector. GammaVision is packed full of analysis features and options which provide a great deal of flexibility. ISOTOPIC is a “post processor” to GammaVision. GammaVision “hands over” to ISOTOPIC a list of peak-by-peak activities computed for each isotope in turn, uncorrected for geometry or absorption, and determined using a calibration which the user makes with a simple TRACEABLE multi-nuclide point source. ISOTOPIC uses the peak results from GammaVision along the source-to-detector distance employed for calibration. Sample physical data (size, distance, weight, material) are entered into ISOTOPIC via easy to use data entry screens. ISOTOPIC then performs the necessary attenuation corrections to the peak-by-peak activities and geometry corrections to the reference activity.

### 1. Setting Up the GammaVision Analysis Library

By reviewing both spectrum and library, select a reference gamma-ray for the nuclide(s) being measured that is high energy, intense, and interference free. The high-energy gamma-rays are not as subject to localized attenuation as much as lower-energy gamma-rays, but, with the exception of  $^{60}\text{Co}$ , most nuclides of interest for measurement do not have an intense high-energy gamma-ray that can be used for reference. A compromise usually has to be made between the reference gamma-ray branching ratio and its energy. It is worth remembering that the error due to counting statistics is usually only a small contribution to the overall measurement uncertainty. Setup the gamma-ray library by using only one gamma-ray for a reference. Position this gamma-ray first in the library for that nuclide. Within the GammaVision library, ONLY the chosen reference peak should be set to be included in the activity averaging process, because ISOTOPIC must perform the individual peak attenuation corrections after the GammaVision analysis. Set this gamma-ray as a key gamma-ray.

### 2. Calibration

Calibrate GammaVision in the usual way, with a point source at a “reasonable distance” (several detector diameters are recommended, say 25 cm). Positioning the source closer will introduce a larger percentage error due to uncertainty in the distance to the active volume of the detector. Use the (patented) autocalibrate for the energy and FWHM calibration. Delete any peaks returned by the autocalibration that obviously lie well away from the FWHM curve. A good FWHM calibration is critical to good peak fitting.

### 3. GammaVision Analysis Option Choices

Use the following GammaVision analysis options:

- (a) Use WAN32 analysis engine.
- (b) Use a five point background average option so that weak peaks will have enough data to establish a good Compton background.
- (c) Use a peak cutoff for the data analysis of 30%. During the peak calibrate phase the peak cutoff may need to be set to 100%.
- (d) Use NUREG 4.16 (Method 12) as the convention for determination of MDA.
- (e) DO NOT use geometry or attenuation corrections within GammaVision. These are handled within ISOTOPIC.

#### 4. Entering Configuration Description

Enter a “best guess” for matrix characteristics as a starting point. The sample weight is very important. If it is not possible to weigh the sample then assume a nominal density. See Figure 3. As an alternative starting point, select a plausible matrix material from the drop down list.

Adjust the correction factor parameters on the isotope plot screen iteratively until all the gamma-ray peaks of the specified nuclide have the same (or as close as possible to the same) value (within uncertainty) as the reference peak of that nuclide. A horizontal plot results when all of the peaks for the given nuclide have been correctly accounted for in terms of container, matrix and uranium (if present) attenuation effects. When there is very little activity within the drum, there may not be enough gamma-ray peaks with good counting statistics to obtain a good slope. When this happens, use the weight of the sample to determine matrix density and do not adjust the matrix density using the plot screen. Figure 4 is an example of a configuration with an inadequate density assignment. Figure 5 is an example of a properly corrected configuration.

#### 5. Display the results before reporting.

Review and check the results. Do these seem reasonable? Do the ratios of the various nuclides seem reasonable based on experience or expectation? When uranium is present and the weight percent  $^{235}\text{U}$  is known, results can be normalized on either the  $^{235}\text{U}$  or  $^{238}\text{U}$  isotope for improved accuracy. Activity can be normalized to another nuclide or uranium isotope on this screen. See Figure 6.

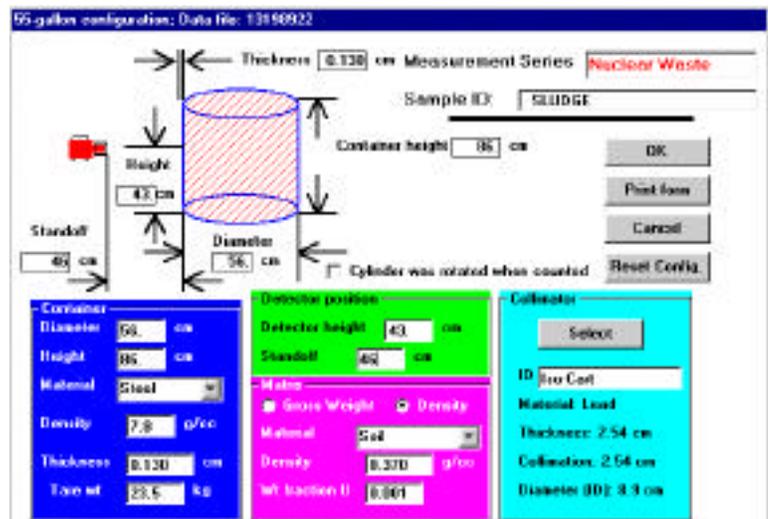


Figure 3. Enter as much information about the sample as possible.

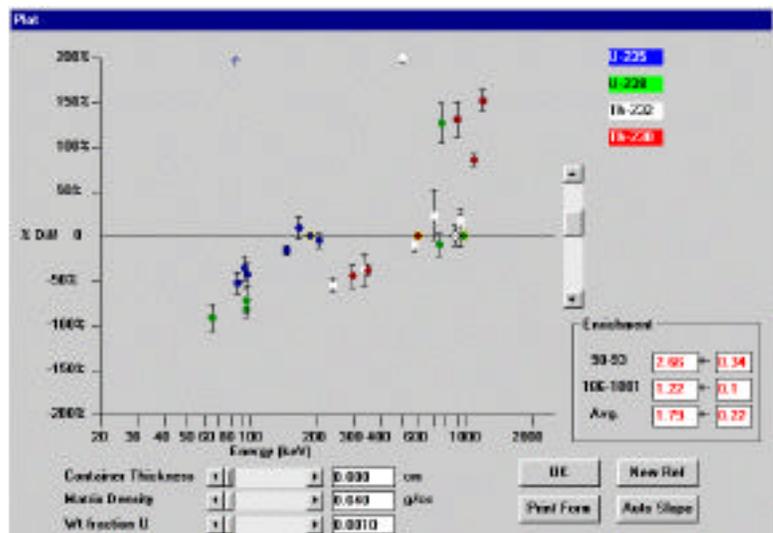


Figure 4. An example of under-corrected configuration.



Figure 5. An example of a properly corrected screen.

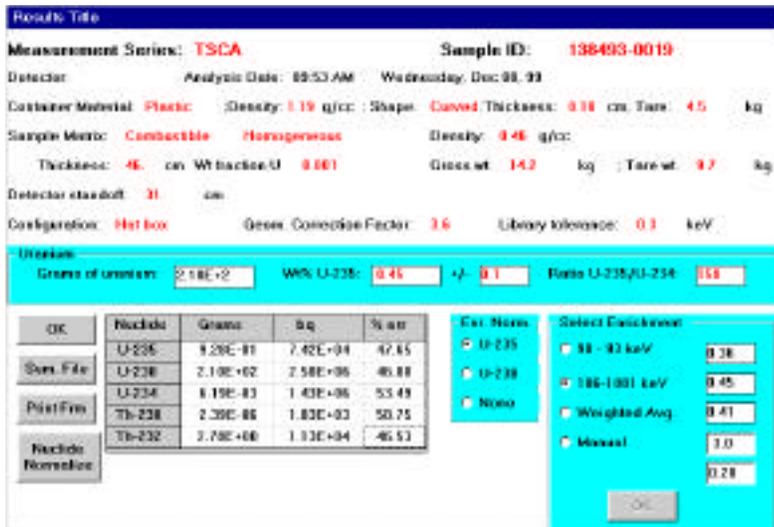


Figure 6. A visual summary of results.

## 6. REPORT Stage

ISOTOPIC provides flexibility in reporting. The user may choose to store the results as hard copy, disk file or data base for further processing.

## 7. Review of unidentified peaks

GammaVision and ISOTOPIC produce a list of all unidentified peaks found in the spectrum. These peaks are not included in the analysis. You should examine all the unidentified peaks to see if they represent gamma-rays from a nuclide already identified, gamma-rays from a new nuclide, or are spurious peaks. If the peaks are from a nuclide they should be added to the library.

## Prediction of System Performance

Estimation of likely minimum detectable activity (MDA) for a new type of sample or a new detector is possible, even before you have either one.

It is very easy to model new sample configurations and to determine the MDA to be expected for a given range of nuclides. What is needed is the point source energy calibration and a background at least similar to what would be measured by that detector in that facility. Data from previous measurements or measurements in other facilities can often be adequate to make these estimates. It is not necessary to have an actual sample, but the matrix must be reasonably known as well as the list of nuclides of interest. The background is analyzed as if it is a sample. The MDA activity obtained from GammaVision is then corrected for geometry, matrix and container attenuation by ISOTOPIC. If counting times will be employed which are different to those employed for the background analyzed, (which is usually the case) the following equation can be used to correct the MDA to the new counting time:

$$MDA_{rt} = MDA_{bk} \times \sqrt{CT_{bk}/CT_{rt}}$$

where:

- $MDA_{rt}$  = MDA activity at the new counting time,
- $MDA_{bk}$  = MDA activity at the counting time for the background,
- $CT_{rt}$  = new counting time,
- $CT_{bk}$  = counting time of the background spectrum .

Some typical measurement sensitivities are shown in Table 2.

**Table 2. Detector sensitivities for a 208-liter Drum with a combustible matrix**

Nuclide	Density = 0.3 g/cc			Density = 0.5 g/cc			Density = 1.0 g/cc		
	g	μCi	kbq	g	μCi	kbq	g	μCi	kbq
<sup>60</sup> Co	-	0.010	0.38	-	0.013	0.48	-	0.021	0.74
<sup>137</sup> Cs	-	0.015	0.56	-	0.020	0.75	-	0.035	1.1
<sup>226</sup> Ra	-	0.038	1.4	-	0.051	1.9	-	0.09	3.3
<sup>235</sup> U	0.019	0.040	1.5	0.029	0.062	2.31	0.056	0.12	3.7
<sup>238</sup> U	4.8	1.61	59.6	6.3	2.08	77.1	10.5	3.49	126
<sup>239</sup> Pu	0.008	530	2.0E4	0.013	811	3.0E4	0.056	1590	5.8E4

Measurement distance from the detector to the surface of the drum = 46 cm (18 in.)

Detector volume = 80% relative

Measurement counting time = 15 minutes

Typical dimensions for a 208-liter drum are: 86 cm high, 86 cm height, and 0.13 cm thick

## ISOTOPIC Measurement Uncertainty

For measured items containing dense material, measurement uncertainties of 30% can be expected. It is difficult to estimate uncertainty accurately. The major sources of error are usually in the determination of the matrix correction factor. Usually, the higher the matrix density the larger the uncertainty in the matrix correction. When using ISOTOPIC, this uncertainty can be reduced by counting longer or using a more efficient detector so that weaker gamma-rays from a nuclide can be used to correct for the matrix. There will be smaller random errors due to uncertainty in positioning the item in front of the detector and counting statistics. If the item is not rotated during the measurements and the detector is close to the container, the uncertainty in the position of the activity (called geometry error) can become significant. ("Close," in this context is less than one drum diameter.) This error can be reduced by positioning the detector further away and rotating the container. In the cylinder configuration, there is a user-set option to identify when an item is rotated. The data is analyzed in the same way if the option is selected or not checked. However, the geometry error and the overall uncertainty will be reduced if the item is rotated during analysis.

## Other Approaches to this Measurement Problem

Monte Carlo calculations, which are used in competing systems to compute activities, must assume a well-characterized matrix. This is seldom the case in the measurement of nuclear waste, which totally negates the apparent sophistication of the approach. In contrast, program ISOTOPIC can be used for a wide variety of container configurations and matrices; analyses are presented in units of activity and grams (necessary for accountable nuclides) for all nuclides.

## Conclusion

ISOTOPIC and the ISO-CART mobile assay system provide a straightforward, good solution in many application situations involving ill-defined matrices and geometries. This application note has provided "getting started" information and answers to many of the more common questions. If you have further questions do not hesitate to ask us. Please send your query to Rich\_Hagenauer@perkinelmer.com, cc info\_ortec@perkinelmer.com or call 800-251-9750 for technical support.

# Frequently Asked Questions

## **Do I need process knowledge of the nuclear facility to analyze samples?**

You don't need process knowledge, but the more you know about the waste, the better the analysis results. Process knowledge is very helpful for the analysis. For poorly characterized material it is helpful to know if a chemical separation was done on the nuclide in question and has equilibrium been established between the daughter and parent nuclides. This is very important when the analyst uses a daughter nuclide to quantify the parent. Thorium and uranium frequently have isotopes that are in a fixed and known ratio. In addition, it is helpful to know if fission product or neutron activation products will be present when trying to identify nuclides present in the sample.

## **Should the container exceed the field-of-view of the collimator so that I am looking at part of it, or should I be able to see the whole container?**

You should be able to see the whole container through the collimator. If the detector/collimator can only view part of the container, then it will still make a calculation for the whole container, but if the mix of radioactive material is different in the unseen part, then the result will not be as good as if the whole container is viewed.

## **May I combine or average the results obtained by viewing the container from different sides as an alternative to rotating it?**

Yes, it is good practice to average results of several measurements for large, dense containers.

## **What if I collect data without a collimator?**

The collimator provides two functions: (1) It provides directional field-of-view of the detector and (2) shields the detector from ambient background. Usually, there are several samples near by that effect the background. To minimize this effect, shielding is needed and is provided by the collimator. Data can be collected without the need of directional field-of-view in many cases, but to operate in an unshielded environment is not recommended.

## **If one peak is an "outlier" on the plot what should I do?**

You should investigate why the peak is an outlier. ISOTOPIC and GammaVision provide the tools and details to do this. The GammaVision interactive reanalysis mode allows you to inspect the fitting of individual peaks by the software. Examination of the residuals of the fit will help determine if there is a poor fit, due for example, to an unexpected interference.

## **On the plot screen sometimes one nuclide will have a negative slope and one nuclide will have a positive slope. Which nuclide do I adjust to fine tune the corrections?**

Use the nuclide with the widest energy span of usable points on the screen. If possible, disregard points representing gamma-ray peaks that are below 65 keV. This condition is normally due to different densities of material shielding different activities in the drum.

## **When using a daughter nuclide to quantify the parent, which half-life do I use in the gamma-ray library?**

If the daughter is in equilibrium with the parent, use the parent half-life and include the gamma-ray from the daughter in the parent library. For example, the 911-keV gamma-ray from  $^{228}\text{Ac}$  is frequently used to quantify  $^{232}\text{Th}$ . The 911-keV gamma-ray should be placed in the  $^{232}\text{Th}$  library along with gamma-ray energies from other  $^{232}\text{Th}$  daughters.

### **How do I quantify the daughter activity if I place references to their gamma-rays in the parent library?**

If equilibrium is established, the activity results of the daughter will be exactly the same as the parent. If parent and daughter nuclides are not in equilibrium, then a more complicated calculation is needed to quantify the parent activity. This calculation is not supported in ISOTOPIC.

### **I suspect that there are going to be inhomogeneties in the drum and that rotating it will reduce the systematic error, but how fast should the drum rotate?**

The drum needs to rotate enough times during the acquisition so that the detector “sees” a good averaged sample. The sample should be rotated at least 20 times during the measurement period. Additional rotations will not reduce the systematic error significantly.

### **ISOTOPIC is easy and economical to use. How does it compare in drum analysis with other techniques such as segmented gamma scanning (SGS) and Tomographic Gamma Scanning (TGS)?**

SGS and more recently, TGS systems are designed to help with nonhomogeneous distributions of attenuation and radioactivity, by “dividing up” the sample under assay into subsamples. In the case of the SGS, the sample is rotated around a vertical axis and “segmented” into horizontal slices. The activity is then the sum of the activities computed for each segment, and the rotation may be combined with the use of a “shine through” source to compute average attenuation for each segment. (Alternatively the “differential absorption” technique on which ISOTOPIC is based may be used.) The TGS uses a transmission source and computes corrected activities “voxel by voxel” (A voxel is a volume element), building up a 3-D mapping of activity and attenuation for the whole drum. In order of accuracy for “difficult” measurements the TGS is most accurate (and costly), the SGS next, and ISOTOPIC and ISOCART least accurate and least costly. “Difficult” is taken to mean “high density” and/or “inhomogeneous matrix or radionuclide distribution.” For light waste (e.g., combustibles) where attenuation is light and contamination is reasonably uniform, the ISOTOPIC result can be as good as the result from either of the other two. For dense waste in which the absorber and the radionuclides are distributed in a highly non-uniform way, the TGS is the best solution

## **References**

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2 R. M. Keyser, T. R. Twomey and P. Sangsingkeow “Advances in HPGe Detectors for Real World Applications,” *The Journal of Radioanalytical and Nuclear Chemistry*, Vol. 224, No. 3, (2000)