

Establishing an ISO-CART Measurement Station to Meet Government Burial Regulations for Radioactive Material

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Meeting Government Regulations on Burial Requirements

Burial regulations vary from country to country, but universally the cost of burial of radioactive material is very expensive. An accurate method is needed to classify the wastes to minimize burial costs. In the United States one of the most expensive wastes to process for disposal is items containing transuranic material. If these items exceed a government-specified limit the items must be buried in a transuranic landfill. Today, the measurement limitation is 3700 Bq (100 nCi) of alpha-emitting material with half-lives greater than 20 years per gram of waste. The most common elements that fall into this category include plutonium, neptunium, and americium. ^{237}Np and ^{241}Am are easily detected but the many of the plutonium isotopes have very low gamma-ray yields. In other words, when those plutonium isotopes decay many alpha particles but relatively few gamma rays are emitted, and gamma rays are the basis of isotope detection and quantification through nondestructive assay. Usually transuranic material is measured and stored in 208-liter (55-gallon) drums. The appendix shows the sensitivity of these nuclides to detection in this type of container. The ^{239}Pu isotope is easily detected and measured, but accurate relative plutonium isotopic values are needed to quantify the other isotopes. These are usually determined from mass spectrometry or alpha spectrometry, or with specialized software (PC/FRAM or MGA++) analysis of gamma-ray spectra obtained from very-high-resolution HPGe detectors.

The unrestricted (free) release limits of slightly contaminated items vary dramatically from country to country. The European Union (EU) [1] and the International Atomic Energy Agency (IAEA) [2] have given countries guidelines for unrestricted release to the public. In the United States the regulations are more strict [3]. See appendix. The standards for the United States are based on the requirement that less than 10 μSv (1 mRem)/yr be added to the public exposure. In the United States the Nuclear Regulatory Commission (NRC) must be shown that release to the public will not increase an exposed individual's radioactive intake by more than this level. However, measurements using nondestructive assay require special permits from the NRC before the measured contaminated items can be released to the public. Normally, these "free release" limits are met by scanning the surface of the contaminated item with sensitive alpha/beta meters. This is a very costly effort for bulk material, and nondestructive assay is much preferred if permits will allow.

Scope

This application note is designed to assist the user of an ORTEC ISO-CART/ISOTOPIC assay system to perform accurate nondestructive measurement of radioactive items in 208-liter drums. This application note addresses choice of equipment, collimator settings, detector positioning, background measurements, counting times, and other good practice methods useful for the development of a measurement station. It includes some helpful hints on the use of the ISOTOPIC software itself. The limits related to unrestricted release of slightly contaminated material and transuranic limits are discussed.

Note the regulations may vary from country to country, facility to facility, and year to year. The user can refer to the appendix as a guide, but the limits in the appendix should not be used as definitive for your facility. The analyses from a measurement station can easily meet radioactive standards for burial at facilities with less restrictive limits.

Definitions

Field-of-View - The entire detection range encompassed by the collimated detector when it is trained in a particular direction.

Blank - A prepared sample with no significant radioactivity whose size, density and average atomic number properties are similar to items being measured.

Cross talk – activity from a nearby item that is detected by the counting system.

Detector/collimator assembly – a detector surrounded by a shield that imposes a directional response on the collimated detector.

Detector standoff – Distance from the face of the collimator to the surface of the item being measured.

K-edge – regions of the spectrum where mass attenuation coefficients are rapidly changing with energy. For this note the K-edge is important for matrices containing large fractions of uranium or plutonium.

Staging areas – locations for storing items prior to/after measurements.

MCB- A term used to mean the signal processing electronics associated with a HPGe detector.

Choice of Equipment

General hardware needs

Below is a summary of items needed for a measurement station. Figure 1 illustrates such a station.

- (1) Detector/and acquisition electronics combination
- (2) Collimator
- (3) ISO-CART
- (4) ISO-turntable
- (5) ISOTOPIC software
- (6) Helpful options:
 - X-COOLER/ISO-CART bracket
 - Back-shield
 - Laser meter
 - Factory calibration
 - Heavy duty collimator
 - Scale

- Wireless communications (for new trans-SPECs)



Figure 1. A measurement station using a trans-SPEC.

Selecting a detector and acquisition electronics

An important first decision in implementing measuring station is the selection of a detector. Large-volume detectors potentially offer better efficiency at both higher and lower energies, by virtue of their crystal length and crystal surface area, respectively. High-energy efficiency depends on both area and depth whereas, at lower energies, crystal surface area dominates crystal length parameters and the issue of contact thickness or dead layer can become a significant factor. Large-volume detectors have better peak-to-Compton ratios, which reduces the Compton continuum and improves detection limit.

P-type (GEM) detectors generally have a dead layer of germanium that attenuates low-energy gamma rays to an extent. This can reduce the efficiency for 59-keV gamma rays from ^{241}Am by a factor ranging from 4 to 10. If ^{241}Am or gamma rays of similar energies are of interest, this effect should not be ignored. The ORTEC FX-GEM and GMX detectors have ultra-thin entrance windows and consequentially good low-energy efficiency. See reference 4 for a good overview of the considerations to be used in choosing an HPGe detector. Note also that P-type detectors are more susceptible to neutron damage. Neutrons from items containing plutonium waste can damage a P-type detector much more quickly than an N-type detector. However, remember that most gamma-rays below 59-keV will be absorbed by the steel container and matrix of the 208-liter drum. Guidance on the thresholds for neutron damage thresholds can be found in Reference 5.

The detector-acquisition electronics combination from ORTEC can come in several different formats, a HPGe detector and an associated portable digiDART MCA is common. It can operate on AC power or battery. For fixed installed use, A HPGe detector and the DSPEC jr. 2.0 , a multichannel buffer, used in conjunction with a PC, is a good alternative when AC power is available.

More recently available, the ORTEC trans-SPEC-family instruments are completely integrated and portable HPGe spectrometers which include detector, acquisition electronics, and battery-powered cooling system in a small package. The trans-SPECs are light and readily portable from location to location. Two standard detector sizes are available. In a measurement station environment continuous power would likely be needed. Wireless communication can be implemented between the control computer and the trans-SPEC.

Cooling methods for HPGe detectors

Most HPGe detectors are LN₂ cooled. ORTEC ISO-CART systems are usually configured with a MOD-7 Dewar that will keep an HPGe detector cool the detector from 3 to 4 days.

In some facilities it is not convenient to keep LN₂ on hand. The ORTEC X-COOLERS is designed as a static cooler system, powered from the mains. It is capable of cooling any size of ORTEC HPGe detector in a PopTop configuration. As long as electricity is available the X-COOLER will keep a detector at operating temperature. A special bracket (ISO-DCC-X) is need for the X-COOLER to keep the detector horizontal within the collimators.

The ISO-CART with choice of collimators

The ISO-CART provides a portable platform for the measurement station. This cart allows the user to raise, lower, and change the angle of the detector/collimator assembly. See Figure 1. Ask for the ISO-CART brochure or visit the ORTEC website at:

<http://www.ortec-online.com/pdf/isocart.pdf>

The heavy-duty ORTEC shield/collimator (SHD-3) attenuates a larger fraction of the ambient background than the lighter models and so is recommended unless weight is a major issue. With a thicker shield, lower sensitivities can be reached. Additional back-shields (2 cm of lead) are available that fit at the back of the preamplifier section of a PopTop detector.

Note that, in the case of an LN₂-cooled portable detector, the MOD-7 type of internal cryostat detector mount and the liquid nitrogen in the Dewar are equivalent to 5 cm (2") of lead for back-shielding purposes.

For trans-SPEC models, additional shielding and collimation can be added.

Depth of Collimation

The normal settings for the detector/collimator assembly are to position the detector within the collimator for a 2.5 cm (1 in.) recess. The standoff for the corresponding configuration will vary with crystal size. Use the *Field of View* subroutine located in the **Services** menu of the Operator Program to help to establish the detector standoff. Note that the detector crystal diameter will be needed. See Figure 2.

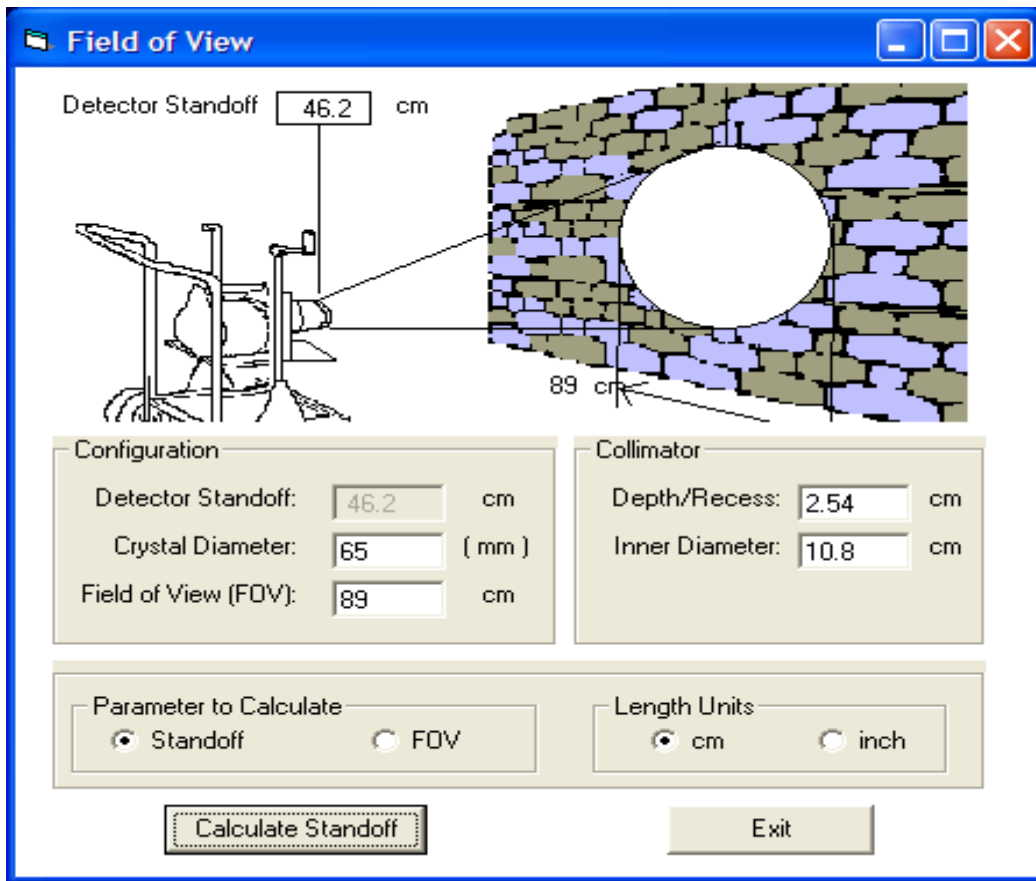


Figure 2. ISOTOPIC subroutine used to help establish detector standoff.

Additional options for a measurement station

- ISO-SCALE: a digital floor scale with a capacity of 1000 lb and a 0.2% accuracy.
- ISO-LASERMETER: a laser distance meter used to establish an accurate (within 3 mm) distance from the collimator face to the inner container.
- ISO-TURNTABLE: rotates the sample waste drum to reduce matrix attenuation error.

Software Use Considerations

In this section follows hints and suggestions to get the best performance for the ORTEC ISOTOPIC-32 (ISO-PLUS) software when carrying out drum assays.

Hints for setting up the library

- (1) Determine if the progeny activity needs to be reported. This is especially complicated with the actinides. Note that when a decay series is in equilibrium the progeny activity is identical to the parent activity.

- (2) For each radionuclide use only one reference gamma ray for each radionuclide, and position it at the top of the list within the library. This gamma ray should be well resolved from neighboring gamma rays and, if possible, of high energy. Higher-energy gamma rays penetrate the matrix better than lower energy gamma rays and therefore make excellent reference gamma rays.
- (3) Minimize the primary library to gamma rays that are likely to be seen in the spectrum.
- (4) Investigate unidentified peaks. Often gamma rays from this list are emitted from nuclides already in the library. To minimize the unidentified peaks reported in the analysis report and to keep the library uncluttered, include in the library every gamma ray ever (by radionuclide) seen at the facility, and leave out gamma rays that have never been seen. Note this implies library development for a particular measurement site will be somewhat continuous.
- (5) The automatic energy calibration option works well if the library is simple and does not contain many low-energy gamma rays. If necessary, you can prepare an additional simple library containing at least five gamma rays found in each sample, spaced throughout the energy range likely to be encountered. Use this library for automatic energy calibration when needed.
- (6) Add “key gamma” flags as necessary to reduce the reporting of false positive radionuclides. The first gamma ray listed in the library is automatically a “key gamma.”

Developing the configurations

Three templates are used to develop a configuration. An example 208-liter drum configuration is included in the default ISOTOPIC database. The only changes needed for the first template are the estimated matrix density and matrix material. For the next template, only the counting times is needed. The third template is very important. See Figure 3. Make the following changes.

- (1) Enter the detector crystal parameters. These are important to establish the efficiency and the detector field of view.
- (2) Enter the proper collimator information. Detector recess is very important.
- (3) Enter the proper standoff and detector height position. Note that standoff is determined by measuring the distance from the collimator front to the outer container.
- (4) When possible, the detector calibration parameters should be entered into each detector through the MCB. If it has been done properly, then each spectrum should have the calibration information “pasted” onto the spectrum and no entry is needed in the calibration file field.
- (5) Enter the proper PBC table.
- (6) Verify that the detector orientation option selection is correct.

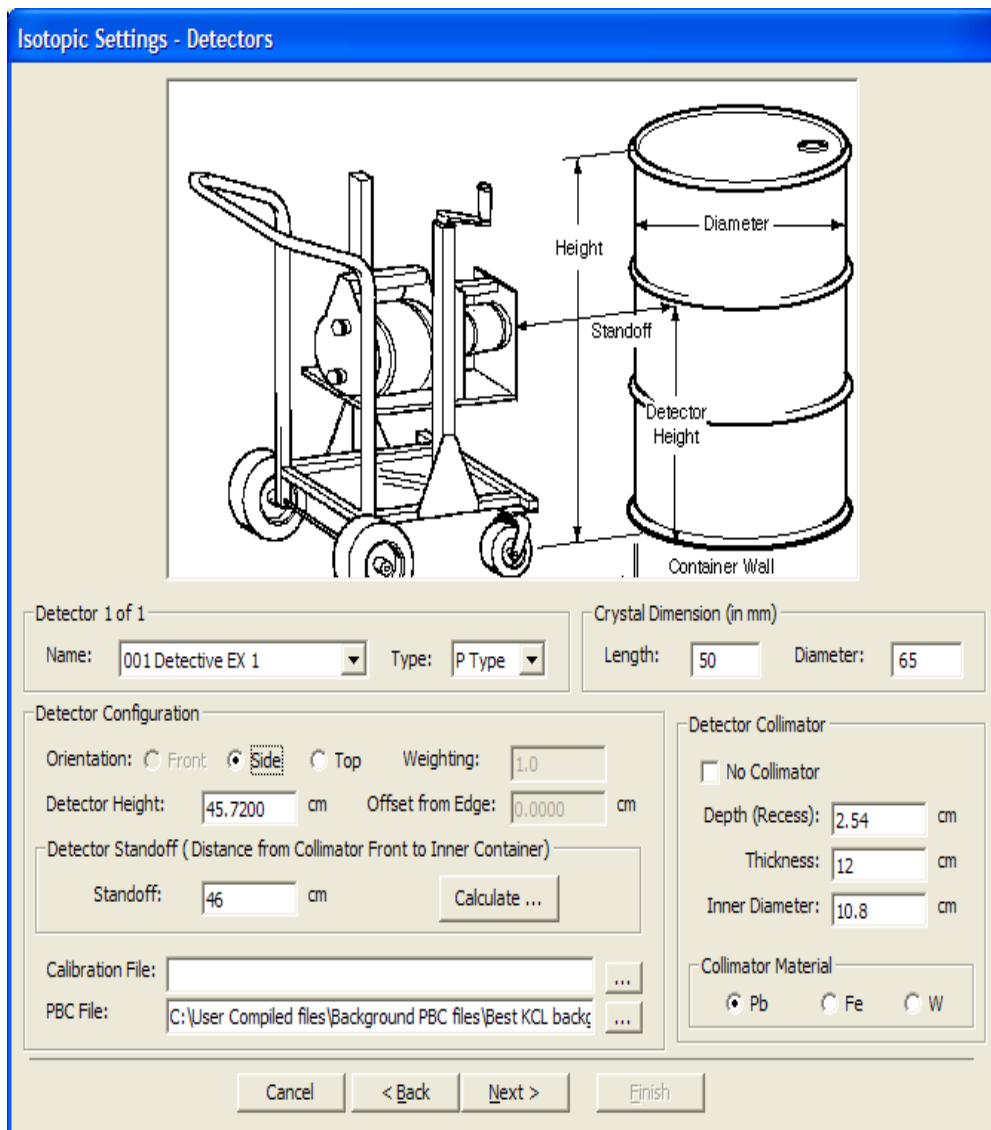


Figure 3. Template used to establish the detector parameter and standoff information.

Working with the fine-tune adjustments

When an item being measured is initially modeled, it is assumed that the activity within the matrix is homogeneous. In practice, however, this is seldom the case. The analysis will be affected in several ways.

- (1) Frequently, activity in a waste item is located in a few hot spots or near the bottom of the drum; or the item may have many air pockets where gamma activity attenuation is much less than the attenuation the average density would predict.
- (2) Note also that gamma rays will likely penetrate the container at an angle that is not normal to the detector. Thus, the container attenuation will be greater than is generally predicted based on the wall thickness only.

- (3) Heavy elements such as uranium and plutonium often include self attenuation. The matrix mass attenuation coefficients sometimes need to include contributions from uranium or plutonium.

The ISOTOPIC software has a fine-tune adjustment feature that will permit the user to vary the density, container thickness and heavy element contributions. The user must then decide how much of these adjustments are necessary from each of the contributions. It is difficult to determine how much of each contribution is necessary. However, the sum of all the corrections should allow gamma rays from the same radionuclide to show the same activity if the modeling estimates are the correct. This is graphically presented in a graph of % deviation from the reference gamma-ray activity vs. energy as shown in Figure 4.

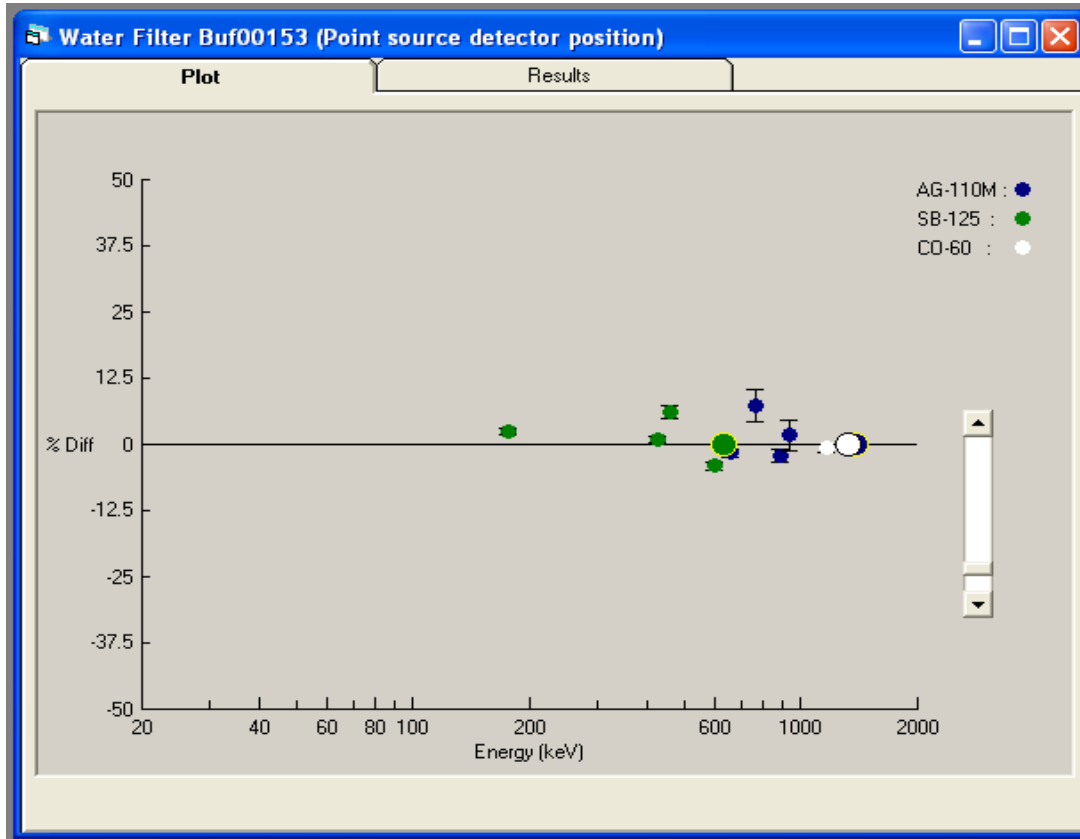


Figure 4. Fine-tune model adjustments.

Be careful with your adjustments. Do not increase the density of the sample to more than twice the average density of the sample or decrease it to less than half the average density. Usually, container thickness is the last fine-tune adjustments to make. Note also that the effective container thickness will always be greater than the measured thickness because most gamma rays processed by a detector enter from an angle. However do not increase the thickness by more than 50% of the actual thickness.

Special helps with uranium analysis

Frequently the wt% ^{235}U is known by the facility with high accuracy. This information can be used to obtain more accurate uranium results by normalizing the activity to ^{235}U results. For

example, if it is known to high accuracy that the wt% ^{235}U is 5.0% and the analysis results report 5.0 grams of ^{235}U . The ^{238}U value can be forced to 95.0 grams. See Figure 5.

| Analysis | Uranium/Plutonium | Report |
|---|-------------------|----------------|
| Uranium Isotopic Weight Fractions | | |
| <input checked="" type="radio"/> Use Entered Value | U-234 | .001 |
| <input type="radio"/> Determined from Spectrum | U-235 | .05 (+/-) .002 |
| | U-236 | 0 |
| | U-238 | 0.949 |
| All uranium isotopes with weight fractions entered will be referenced to U235 | | |
| | Total | 1 |
| Pu Isotopic Weight fractions | | |
| <input type="radio"/> Use Entered Value | Pu-238 | 0.0019 |
| <input checked="" type="radio"/> Determined from Spectrum | Pu-239 | 0.7095 |
| | Pu-240 | 0.2458 |
| | Pu-241 | 0.0328 |
| | Pu-242 | 0.00975 |
| | Total | 0.99975 |

Figure 5. Entering the wt fraction uranium and plutonium isotopes.

To use this feature, click on the Uranium/Plutonium tab in the analysis template and enter the weight fraction ^{235}U . The same principle applies to plutonium. ^{239}Pu is easily determined, however, the other plutonium isotopes are much more difficult to analyze. If the weight fraction of plutonium isotopes is known, this feature should be used.

Common Measurement Mistakes

Background measurements

Taking a background measurement is not as simple as it seems. One of the common mistakes is to measure a background with the detector in place but without a sample. This will give an artificially high background because the item being measured will mask some of the background activity. The best method is to position and measure a blank, and to develop a peaked background correction (PBC) table. The matrix of the blank should be as free as possible of radioactivity yet contain the same gamma absorbing material and density that a sample item is likely to have. If the PBC table is to be used for many samples, take a long (overnight) count for your PBC table so there is no significant bias introduced to all the samples.

If the radioactive items in the staging area are contributing significantly to the background, then no items should be moved into or out of that area while measurements are being taken. Try to position the staging area as far as possible from the measurement station area.

Caution should be used when deciding the location of a staging area. If one hot item in the staging area is contributing significantly to the background, a special background should be taken to account for this contribution. Ambient radiation monitors may alert the user that an especially radioactive item is located in the staging area and a special background should be taken.

Likewise, if counting stations are positioned close together, crosstalk could occur between the detectors of those stations. Thicker collimators are recommended to minimize this effect.

Measuring on the opposite side of the container

Nuclear waste is very nonhomogeneous and, for large containers, matrix attenuation corrections dominate the correction factors. If drums can be rotated during measurement the matrix “appears” to be more homogeneous to the detector. This does not solve the geometry error of different activity levels within a drum, however, the geometry correction factor error is usually less than the matrix correction factor error.

It is very important to measure the opposite side of the drum if it cannot be rotated. When modifying the configuration in ISOTOPIC, configure this instance as two detectors (positions).

Minimizing uncertainty

There are several measurement techniques that can be used to minimize the measurement uncertainty. These can be characterized by minimizing the correction factors. These hints will help in making decisions:

- (1) Counting statistics are seldom a major contributing factor to the overall uncertainty. Increasing the item-to-detector distance will improve the geometry correction but reduce the sensitivity.
- (2) Avoid collimator corrections if possible. Position the item at the distance shown in Table 1 or greater so that these corrections do not vary from 1.00 (no correction).

Sample fill height

Uncertainty in effective matrix density is one of the largest sources of measurement uncertainty. Measured results from containers improperly assumed to be full will be biased low because the average density is computed based on the volume of a full drum. To minimize this effect, it is good practice for facilities to fill the containers to be measured prior to measurements. To determine the height of the contents without opening the drum, perform simple transmission measurements using a low-energy transmission source. Position the source on one side of the container and use a collimated NaI detector on the other side. When both are moved down the container, such that the collimated detector can always “see” the transmission source, a drop in count rate will reflect the top level of the matrix. Note that activity of the item being measured will contribute to the transmission source measurement, thus the transmission source should be hotter than the sample.

System Performance

What system performance can be expected? This is not a trivial question. System performance depends on:

- (1) Gamma-ray yield and detectability of the nuclides of interest
- (2) Homogeneity of the matrix
- (3) Matrix density
- (4) Container uniformity
- (5) Properly identified fill height
- (6) Detector specifications
- (7) Shielding
- (8) Count time
- (9) Environmental background

The first five items are generally beyond the control of the operator while the last four can be, with care, improved or optimized.

One often overlooked consideration is that of specific activity detection limits, such as Bq/g. For higher density items attenuation is increased, but the increase in mass of the matrix may help the specific activity detection limit.

Typical Results

The appendix provides some indicative results in terms of Minimum Detectable Activity (MDA) (see column 3) in comparison to some common release limits. These were made with a 77% HPGe detector and, in general, “bigger is better” in terms of detector efficiency. The MDA can be improved and systematic errors due to nonhomogeneity reduced by making multiple measurements of a single sample either sequentially with a single detector or simultaneously with multiple detectors and using the ISOTOPIC average MDA feature.

Conclusion

A collimated ISO-CART with a large HPGe detector can meet or exceed the measurement requirements for burial of radioactive material. However, because the slightly contaminated material is normally nonhomogeneous, special permits must be granted for unrestricted release of slightly contaminated material. For separation of transuranic waste into the proper waste category, measurements performed with the ISO-CART/ISOTOPIC system exceed the required specifications.

Additional helpful information can be found the *Getting Started in ISOTOPIC 4.x* installation and startup guide.

References

1. European Communities, *Laying Down Basic Safety Standards for the Protection of the Health of Workers and the General Public Against the Danger Arising from Radiation*, Luxembourg, Council Directive 96/29/EURATOM (MAY 1996).
2. International Atomic Energy Agency, *Clearance Levels for Radionuclides in Solid Materials-Application of Exemption Principles*, Vienna, IAEA-TECDOC-855 (1996).
3. Nuclear Regulatory Commission, *Radiological Assessments for Clearance of Equipment and Materials from Nuclear Facilities*, Washington, D.C., Draft, NUREG-1640 (1990).
4. *The Best Choice of High Purity Germanium (HPGe) Detector*, ORTEC application note, http://www.ortec-online.com/pdf/best_choice_det.pdf.
5. *Neutron Radiation Damage in Germanium (HPGe) Detectors*, ORTEC application note, <http://www.ortec-online.com/detectors/photon/a8.htm>.

Appendix

Measurement Station Sensitivity (MDA) (Bq/g) for 208-liter drums*

| Nuclide | Ref. gamma ray(keV) | Measurement station sensitivity | Unrestricted release limits (Bq/g) | | | US Transuranic limit |
|---------|---------------------|---------------------------------|------------------------------------|------|--------|----------------------|
| | | | EU (RP89) | IAEA | US NRC | |
| Mn-54 | 833.82 | 0.0061 | 1 | 0.3 | 0.1 | |
| Co-58 | 810.78 | 0.0075 | | | 0.1 | |
| Fe-59 | 1098.85 | 0.0109 | 10 | 3 | 0.09 | |
| Co-60 | 1332.5 | 0.0050 | 1 | 0.3 | 0.04 | |
| Zn-65 | 1115.48 | 0.0169 | | | 0.05 | |
| Nb-94 | 871.09 | 0.0076 | | | 0.06 | |
| Nb-95 | 765.81 | 0.0084 | 10 | 1.3 | 0.1 | |
| Ru-103 | 496.71 | 0.0064 | | | 0.02 | |
| Rh-106 | 621.93 | 0.0717 | | | 0.05 | |
| Ag-110M | 657.91 | 0.0075 | 10 | 0.3 | 0.04 | |
| Sb-124 | 1690.84 | 0.0017 | | | 0.06 | |
| Sb-125 | 600.6 | 0.0304 | | | 0.2 | |
| Cs-134 | 604.72 | 0.0079 | | | 0.02 | |
| Cs-137 | 661.81 | 0.0077 | 10 | 0.3 | 0.04 | |
| Ce-144 | 134.96 | 0.1329 | | | 4 | |
| Pr-144 | 696.39 | 0.4923 | | | 3 | |
| Eu-152 | 1408.01 | 0.0347 | | | 0.09 | |
| Eu-154 | 1274.44 | 0.0163 | | | 0.08 | |
| Ce-141 | 145.49 | 0.0213 | | | 0.06 | |
| Ra-226 | 609.3 | 0.0193 | 1 | | | |
| U-235 | 185.65 | 0.0166 | 1 | | | |
| U-238 | 1001.03 | 0.9097 | 1 | | | |
| Pu-238 | 152.65 | 508.17 | | | | 3700 |
| Pu-239 | 413.71 | 501.37 | | | | 3700 |
| Pu-240 | 160.64 | 1876.15 | | | | 3700 |
| Pu-241 | 208.04 | 1783.14 | | | | |

*Drums were measured for 15 minutes at a standoff of 46 cm with a collimated 77% detector. Density was 0.3 g/cc. MDA was computed using the Reg. Guide 4.16 method.