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Definition of Energy-Calibrated Spectra for National Reachback

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Abstract

Accurate energy calibration is critical for the timeliness and accuracy of analysis results of spectra submitted to National Reachback, particularly for the detection of threat items. Many spectra submitted for analysis include either a calibration spectrum using ^{137}Cs or no calibration spectrum at all. The single line provided by ^{137}Cs is insufficient to adequately calibrate nonlinear spectra. A calibration source that provides several lines that are well-spaced, from the low energy cutoff to the full energy range of the detector, is needed for a satisfactory energy calibration.

This paper defines the requirements of an energy calibration for the purposes of National Reachback, outlines a method to validate whether a given spectrum meets that definition, discusses general source considerations, and provides a specific operating procedure for calibrating the GR-135.

ACKNOWLEDGMENTS

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Nomenclature

DOE	Department of Energy
DOT	Department of Transportation
FWHM	full width at half maximum
NRC	Nuclear Regulatory Commission
SNL	Sandia National Laboratories
SNM	special nuclear material

1. DEFINITION

For the purposes of performing nuclide identification, a well-calibrated spectrum is one in which the energy assigned to the center of each channel differs from the true energy by no more than 25% of the FWHM at the assigned energy.

1.1. Discussion

Gamma-ray spectrometers convert photon interactions in the detector material into electronic pulses whose pulse height (or voltage) is directly related to the total energy deposited in the detector. These pulses are processed by the detector electronics into a histogram of pulses per channel where the channels are contiguous bins[1]. Energy calibration is simply the mapping of those channels to the energy deposited in the detector.

Energy resolution is a number defined as the full width at half maximum (FWHM), measured in energy units, of the full-energy peak formed by the detection of mono-energetic gamma-rays. In general, the energy resolution is not constant across the entire energy range of the detector, but varies as a function of energy.

For a properly calibrated spectrum, the energy assigned to each channel will be within 25% FWHM of the true energy for that channel.

1.2. Summary of discussions with the National Reachback community

In discussing the definition and its possible impact with the National Reachback community, two points were made that are worth noting. First, the calibration step of the analysis is critical to ensure accurate, timely, results. Therefore, having a standardized procedure for taking calibration spectra would be very helpful. Second, having a standard definition of well calibrated spectra might also encourage detector manufacturers to build detectors that use better self-calibration approaches and thus could take spectra that are better calibrated a priori.

[1] For the causes of non-linearity and energy resolution in scintillators, see: R. Whitlock, B. Carnahan, and M. Black *Overview: The Impact of Scintillator Non-Linearity on Energy Calibration*, DNDO Document 600-ECSP-121040v1.00, December 2, 2013.

2. VALIDATING AN ENERGY-CALIBRATED SPECTRUM

2.1. General Considerations

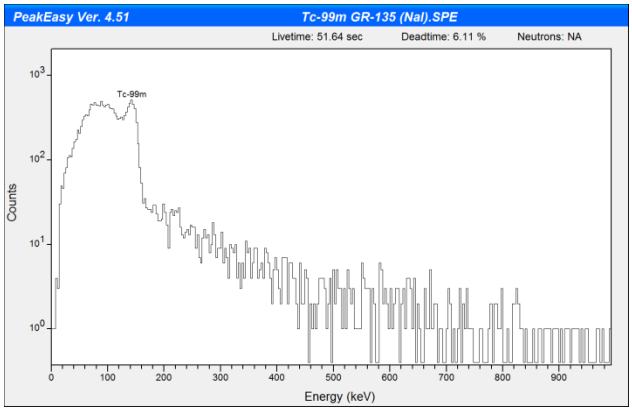

Figure 1. Tc-99m spectrum from PeakEasy Library.

The range over which an energy-calibration is required for a particular spectrum depends on the nuclides or nuclear reactions present. For example, if the spectrum contains only Tc-99m {Figure 1} (emitting a strong line at 141 keV) it might be unnecessary to calibrate the spectrum beyond a few hundred keV. However, analysts rarely know what nuclides to expect in a given unknown spectrum, therefore a calibration over the full energy range of the detector is advisable. The unknown nuclides may not result in any distinct peaks, but may instead simply elevate the continuum over a broad range of energies. Several areas where indications of special nuclear material (SNM) and nuclear interactions can be seen are of particular interest {Table 1}

Table 1. Lines of interest. These lines represent a few of the nuclides and nuclear reactions that may be present in unknown spectra. This table is incomplete and should be considered a small sample of nuclides and nuclear reactions of interest to the analyst.

40 – 100 keV	x-rays and Am-241
140 – 200 keV	^{235}U
350 – 450 keV	^{239}Pu
600 – 700 keV	^{239}Pu & ^{240}Pu
800 – 1000 keV	Fe(n, n'), ^{228}Ac (distinguish ^{232}Th from ^{232}U), & ^{238}U
2223 keV	H(n,g)
2614 keV	^{232}U & ^{232}Th
4440 keV	Be(α,n)
7650 keV	Fe(n,g)
10830 keV	N(n,g)

In practice, it is important to ensure that an unknown spectrum is calibrated over the full-energy range of the acquired spectrum.

This is best accomplished by using a calibration source that has numerous strong peaks that span the full energy range of the detector; a single source or multiple sources measured simultaneously may be used. Unfortunately, there are no long-lived nuclides that produce

gamma rays above 3000 keV. If it is necessary to include peaks above 3000 keV, it is possible to create sources that utilize nuclear reactions to produce higher energy gamma rays. These sources typically require higher activity sources to provide significant yield for the gamma ray of interest. They also frequently produce secondary types of radiation, making the calibration source more difficult to ship and handle. This makes producing and distributing a standardized calibration source that extends above 3000 keV difficult.

The calibration source should be counted for a sufficient time, at a reasonable distance, to allow the development of significant peaks. This distance and time will depend upon the source and detector as well as the background. A significant peak, for the purpose of calibration, should be at least 50σ above background. This value was determined by performing a visual inspection of several spectra from HPGe, NaI, and $LaBr_3$ detectors. A 50σ peak is unlikely to be missed and will allow the spectral peaks to be associated with the expected calibration lines.

2.1. Validation Method

A given spectrum may or may not contain sufficient information to determine if it meets the definition established in the first section. The following procedure can be applied to any spectrum to determine if it is calibrated.

- Define the full energy range of the detector.
    - From the low energy cutoff of the detector to the full energy scale of the detector.
- Determine which source, or sources, were used for the calibration spectrum.
    - This information must be provided with the spectrum.
- Evaluate the spectrum for sufficient peaks.
    - Peaks must at least 50 sigma above background.
    - The peaks should be spaced over the full energy range of the detector (up to ~3000 keV).
    - At least one peak should be between the low energy cutoff and 200 keV.
    - A minimum of two more peaks should be clearly visible in the spectrum, one near the middle of the energy range and a second near the upper energy limit.
- Fit the full energy peaks in the calibration spectrum.
    - Label the centroid of the fit to each peak as the peak energy.
- Compare these peak energies to the expected energies from the declared calibration source.
    - All full energy peaks should be within 25% of the FWHM of the expected energy.

3. SOURCE CONSIDERATIONS

The source should be selected so that there are significant peaks distributed over the full energy range of the detector. A minimum of three peaks with intensity of at least 50σ should be visible in the spectrum. These peaks should be spaced across the full energy range, with at least one peak below 200 keV, one peak near the middle of the energy range, and one near the upper limit. The source activity should be chosen so that a calibration spectrum can be acquired in a reasonable amount of time, with the source located a short distance away from the detector. The time and distance required to acquire the calibration spectrum will vary with application, however a good target should be about five minutes at 2-20 cm. If the source is too weak, it will not result in 50σ peaks when located 2-20 cm from the detector. If the source is positioned very close to the detector, coincidence peaks may complicate the spectrum. A very strong source positioned too close to the detector will also result in random sum peaks and high dead-time.

The source should have a sufficiently long half-life to ensure that the activity of the source does not change significantly during the period of the measurement. It is also highly desirable to use a source with a half-life long enough to not require frequent source replacement. This trade-off between useful source lifetime and high source activity will be an important consideration unique to the application.

While the source peak distribution and half-life are the two most important considerations, other key considerations include:

- Regulatory restrictions (i.e., is a license needed?)
- Cost of procurement / Availability
- Contamination potential
- Form factor (e.g., size, weight, solid, liquid, powder)
- Potential to be useful as a source for determining the detector's detection efficiency
- Single source solutions are preferable over multiple-source solutions

The best candidates for calibration sources typically include naturally occurring nuclides that are in equilibrium with their daughters, e.g. ^{232}Th and ^{226}Ra. Other sources commonly used for calibration include ^{22}Na, ^{60}Co, ^{137}Cs, ^{133}Ba, ^{152}Eu, and ^{241}Am. Depending upon the resolution of the detector being used, the distance between source peaks should be considered. For lower resolution detectors, peak interference can make it much more difficult to accurately determine the centroid of any given peak. The table below lists the lowest and highest energy, and number of significant gamma ray lines for each of these sources.

Table 2. Energy range for significant intensity gamma rays listed for common nuclides used as calibration sources.

	Lowest Energy Gamma	Highest Energy Gamma	# of Significant Gammas
^{232}Th	239 keV	2614 keV	8
^{226}Ra	186 keV	2204 keV	12
^{22}Na	511 keV	1275 keV	2
^{60}Co	1173 keV	1332 keV	2
^{137}Cs	662 keV	-	1
^{133}Ba	81 keV	384 keV	5
^{152}Eu	122 keV	1408 keV	7
^{241}Am	59 keV	-	1

This table should not be considered an exhaustive list of all possible calibration sources. Calibrations may be performed using a wide variety of sources for reasons including: suitability of the source for the measurement and availability of the source.

Depending upon the full energy range of the detector and the requirements of the measurement scenario, it may be necessary to calibrate the spectrum above 2614 keV. This is difficult using standard long-lived nuclides. To provide full energy peaks above this energy a source that includes nuclear reactions [i.e. (n,p), (n,α), (α,n), (p,n), (n,n'), (n,γ), etc…] must be used. For example, alpha particles reacting with beryllium will produce a full energy peak at 4440 keV and neutrons reacting with iron will produce lines at 7631 keV and 7645 keV.

$$^{9}_{4}Be + \alpha \rightarrow ^{12}_{6}C + n + \gamma$$

$$^{56}_{26}Fe + n \rightarrow ^{57}_{26}Fe + \gamma$$

4. SUGGESTED OPERATING PROCEDURE FOR ACQUIRING A CALIBRATION SPECTRUM FOR THE SAIC GR-135

4.1. Source Selection

The full energy range for the GR-135 is approximately 20 keV to 3500 keV. Very few long-lived nuclides emit gamma rays over a range this large. Looking at the table above, we could combine sources to produce a calibration standard that would span the entire energy range. One possible combination, using ^{241}Am and ^{232}Th, would provide significant, well-spaced, peaks from 59 keV to 2614 keV.

The optimum source strengths, distance, and acquisition time should be chosen to ensure a low dead-time and provide significant (>50 sigma) full energy peaks. The source strength chosen will dictate the distance and time used during the calibration. A weak source will need to be counted longer, closer to the detector, while a strong source can be counted for a shorter time and can be moved farther from the detector. For the purposes of this paper, we will choose a source activity for both the ^{241}Am and ^{232}Th of 1 µCi.

The optimum distance can be determined by moving the source closer to the detector and analyzing the spectrum for dead-time, coincidence peaks, and random sum peaks. Figure 2 shows the effect of moving this calibration from 10 cm (blue line) to 0.1 cm (black line). At 2 cm, we can clearly see a coincidence peak near 3200 keV. The dead-time has also increased from 1.5% to 22%. Moving even closer to the detector, we can see that although the total number of counts is significantly higher, the peak to noise ratio is getting worse. This is due to an increase in the dead-time, now 62% at 0.1 cm, degrading the quality of the spectrum.

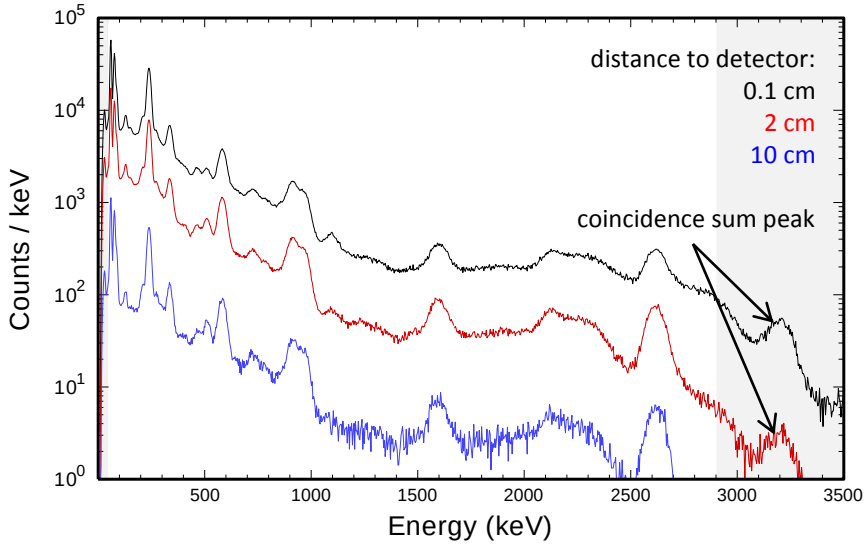

Figure 2. GADRAS simulation showing the effect of moving the ^{241}Am/^{232}Th source closer to the detector. As the source moves closer the dead time increases from 1.5% at 10 cm to 62% at 0.1 cm, and a coincidence peak can be seen near 3200 keV.

If we fix the source distance at 10 cm, we can now determine how long to count the source. Table 3 shows how the sigma value changes for the four major peaks in the ^{241}Am/^{232}Th source. A minimum count time of 240 seconds would be required to ensure all four peaks are at least 50σ above background. However, since the 583 keV line is close to the 50σ minimum, increasing the count time to 300 seconds is advisable.

Table 3. Number of sigma above background for the four major lines in the calibration source. As the count time increases, the peaks become more defined.

Time (s)	Calibration Line			
	59 keV	238 keV	583 keV	2614 keV
60	85σ	60σ	25σ	40σ
120	95σ	84σ	35σ	45σ
180	110σ	90σ	45σ	60σ
240	152σ	115σ	55σ	62σ
300	176σ	145σ	70σ	65σ

For the source under discussion, 1 µCi of ^{241}Am and 1 µCi ^{232}Th, performing a calibration at a distance of 10 cm for 300 seconds results in a spectrum with an acceptably low 1% dead-time and significant peaks across the entire energy range{Figure 3}.

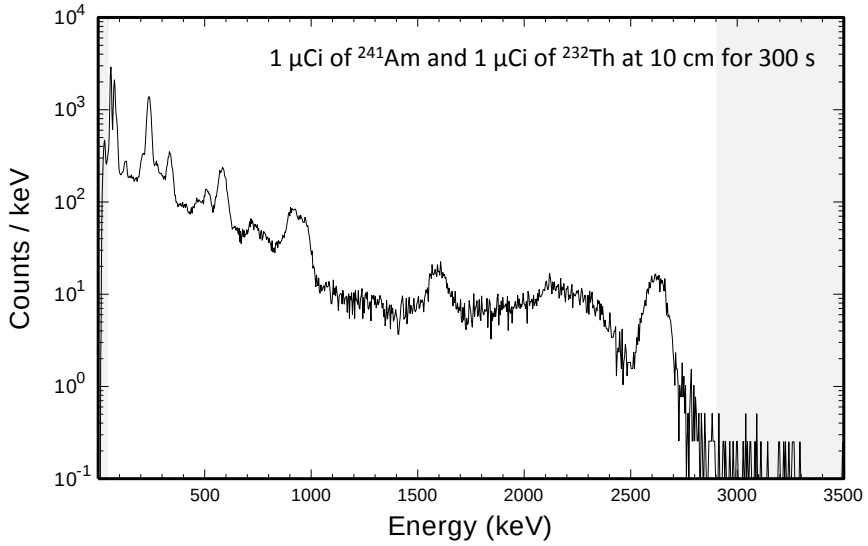

Figure 3. Simulated calibration spectrum containing 1 µCi of ^{241}Am and 1 µCi of ^{232}Th, including estimated background for Livermore, CA, measured for 300 seconds at 10 cm using a GR-135 *(simulated using GADRAS 18.3.1).*

These two nuclides are commonly used and are available at a range of activities. The sources discussed above may require special packaging to comply with shipping regulations and may require the user to have special training to handle these sources. Department of Transportation (DOT) and Nuclear Regulatory Commission (NRC) regulations should be consulted to determine the shipping requirements for radioactive sources. The contamination risk associated with the

source would depend on the configuration. However, both of these sources are typically sold as sealed sources minimizing any risk of contamination.

4.2. SAIC GR-135 Calibration Procedure

4.2.1. Pre-Calibration Considerations

To ensure the best possible calibration spectrum, the following factors should be considered:

- The detector should be free of any radioactive contamination.
- The batteries should have enough reserve to complete all required measurements without needing replacement or recharging; alternatively an external power source may be used if available.
- The instrument should be turned on and allowed to acclimate, in a location at the same temperature as the location in which the item of interest will be measured.

To ensure the highest quality calibration spectrum, the measurement location should be as free as possible from any other radiation source.

4.2.2. Calibration Procedure for the GR-135

Using the source selected above, 1 µCi of ^{241}Am and 1 µCi ^{232}Th:

- If possible, place the detector on a table or other surface raised at least 50 cm above the floor (or ground).
- Place the calibration source 10 cm from the front face of the GR-135, at the same height as the detector.
- Acquire at least a five minute spectrum of the source.
- Record the time and/or spectrum number of the calibration spectrum, as well as the sources used to perform the calibration.

When taking data intended for analysis purposes, a calibration should be performed if any of the following criteria are true:

- The detector has experienced a significant (>15° F) change in temperature between acquiring the unknown spectrum and acquiring the calibration spectrum.
- More than four hours have elapsed between acquiring the unknown spectrum and acquiring the calibration spectrum.
- The detector was dropped or jolted hard in the time interval between acquiring the calibration and the unknown spectrum.

Calibration may be completed before or after measuring an unknown item.

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