

Detector Resolution Required for Accurate Identification in Common Gamma-Ray Masking Situations

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Abstract:

Accurate nuclide identification depends on the ability to determine if specific peaks are present in the spectrum. Several current handheld nuclide identifiers and portal monitors use a variant of a peak quality value for this. The peak quality is usually calculated as the peak area divided by the uncertainty of the peak area and when this quotient is above a threshold value, the peak is said to be present. Other works (1, 2) have developed a formalism to calculate the peak uncertainty for interfering peaks based on the detector resolution, background, individual peak areas, and peak separation. The threshold on peak uncertainty determines the minimum activity that will be identified or detected. Care must be used in the selection of the threshold in order to comply with the false positive and false negative requirements of the detection system regime, or “concept of operations”. The performance standards for the handheld identifiers and portal monitors specify the nuclides required to be identified. From this list and other commonly expected nuclides, the energies of the expected gamma rays can be tallied, yielding a table of the separations of adjacent peaks possible in the collected spectrum. Using the formalism, the peak quality value can be determined as a function of the detector resolution, peak area and background for the energy separations in the table determined above. Results are shown for the cases of HEU and plutonium with the masking nuclides of NORM, ^{133}Ba , or ^{57}Co for both germanium and sodium iodide detectors. Typical resolutions, efficiencies and counting times were used.

I. Introduction

The control of radionuclides at border crossings and the location of sources in search situations are complicated by the natural background, medical isotopes, goods in transit containing high natural emitters, and legitimate radionuclide shipments. To differentiate between legitimate and illicit materials, it is necessary to identify, with confidence, the nuclides involved. This is commonly done by their gamma-ray signatures.

The situation is further complicated by the need to have a low rate of false positives and a low rate of false negatives in a reasonable counting time. For freight crossings at a border, the counting time for the entire container may be less than 60 seconds. Too many false positives cause long delays and often result in the detection device being turned off or ignored. Too many false negatives means there will be the potential for illicit material to be trafficked undetected.

The rate of false positives and false negatives is related to the ability to accurately detect a peak in the spectrum. A peak is said to be present in the spectrum when a parameter based on the peak region of the spectrum exceeds a specified threshold value. Others (1, 2, 3) have used the “precision” of a peak for this parameter. The precision is defined as the uncertainty in the net peak area (σ) divided by the net peak area (A). Both σ and A depend on the background counts per unit energy in the spectrum, the resolution of the detector, and interfering peaks. The inverse of the sensitivity, or peak quality factor, is used below to show the resolution needed to accurately identify peaks from common materials in expected masking situations.

II. Nuclides and Gamma Rays

The nuclides and principle gamma rays for nuclides that could be used in an Improvised Nuclear Device (IND), Improvised Explosive Device (IED), or Radiological Dispersal Device (RDD), as well as those used extensively in commerce are given in several works (4, 5, 6). The principle nuclides from these works are shown in Table I. The table also includes several nuclides commonly used for calibration of instruments. Arlt (7) has described cases where the gamma-ray lines in this table with small separation have caused misidentification due to the spectral resolution of the instrument. Reinhard (8) describes cases where masking of plutonium is possible with low to medium resolution detectors and not possible with high resolution detectors.

Terracol (1) has proposed a general rule that the line separation should be greater than the detector resolution for “reasonable calculations.” Using Table I and assuming that the high resolution (HPGe) detector to have a resolution of 1.0 keV at 122 keV and 2.5 keV at 1.3 MeV, there are 14 cases out of 135 gamma rays where the separation of the lines in the table is less than this resolution. For the low resolution detector (sodium iodide or NaI)

at 9% resolution, there are 123 cases where the separation is less than the resolution.

| Table I Principle Nuclides of Interest | | | | | | | | | |
|--|---------------------|--------------|------------------------|--------------|------------------------|--------------|------------------------------------|--------------|------------------------|
| Energy (keV) | Nuclide | Energy (keV) | Nuclide | Energy (keV) | Nuclide | Energy (keV) | Nuclide | Energy (keV) | Nuclide |
| 35.46 | ¹²⁵ I | 193.59 | ²³³ U | 356.00 | ¹³³ Ba | 646.02 | ²³³ Pa | 1063.60 | ²⁰⁷ Pb |
| 59.54 | ²⁴¹ Am | 208.36 | ¹⁷⁷ Lu | 357.45 | ¹⁰³ Pd | 657.06 | ⁷⁶ Br | 1093.67 | ¹⁷² Lu |
| 60.01 | ¹⁵⁵ Eu | 210.94 | ²³³ U | 363.50 | ¹⁵⁹ Gd | 661.60 | ¹³⁷ Cs | 1099.25 | ⁵⁹ Fe |
| 69.67 | ¹⁵³ Sm | 227.08 | ¹⁸⁸ W | 364.50 | ¹³¹ I | 662.45 | ²⁴¹ Am | 1112.07 | ¹⁵² Eu |
| 70.82 | ²⁰⁴ Tl | 238.63 | ^{228, 232} Th | 375.00 | ²³⁹ Pu | 669.60 | ²¹¹ At | 1173.20 | ⁶⁰ Co |
| 80.20 | ²⁰⁴ Tl | 245.35 | ¹¹¹ In | 383.00 | ¹³³ Ba | 687.00 | ²¹¹ At | 1189.05 | ¹⁸² Ta |
| 81.00 | ¹³³ Xe | 249.79 | ¹³⁵ Xe | 388.32 | ^{252, 249} Cf | 687.59 | ²⁴⁰ Pu | 1205.70 | ²⁰⁰ Tl |
| 86.54 | ¹⁵⁵ Eu | 255.06 | ¹¹³ Sn | 391.68 | ¹¹³ Sn | 711.68 | ^{166m} Ho | 1221.41 | ¹⁸² Ta |
| 88.00 | ¹⁰⁹ Cd | 264.70 | ⁷⁵ Se | 393.53 | ⁶⁷ Ga | 722.00 | ²⁴¹ Am | 1274.50 | ²² Na |
| 97.43 | ¹⁵³ Gd | 279.50 | ⁷⁵ Se | 410.90 | ¹⁶⁶ Ho | 739.50 | ^{99m} Mo-Tc | 1291.60 | ⁵⁹ Fe |
| 103.18 | ¹⁵³ Sm | 290.67 | ¹⁸⁸ W | 411.80 | ¹⁹⁸ Au | 742.64 | ²¹¹ At | 1332.50 | ⁶⁰ Co |
| 105.31 | ¹⁵⁵ Eu | 293.55 | ¹⁹⁴ Ir | 413.71 | ²³⁹ Pu | 776.52 | ⁸² Sr- ⁸² Rb | 1345.78 | ⁶⁴ Cu |
| 112.95 | ¹⁷⁷ Lu | 300.22 | ⁶⁷ Ga | 415.93 | ²³³ Pa | 777.90 | ⁹⁹ Mo | 1408.00 | ¹⁵² Eu |
| 122.06 | ⁵⁷ Co | 300.34 | ²⁴¹ Pu | 439.56 | ²⁰² Tl | 803.13 | ²¹⁰ Po | 1460.75 | ⁴⁰ K |
| 129.28 | ²³⁹ Pu | 302.00 | ¹³³ Ba | 477.99 | ¹⁸⁸ Re | 807.79 | ⁶⁴ Ga | 1596.50 | ¹⁴⁰ La |
| 135.28 | ²⁰¹ Tl | 308.46 | ¹⁹² Ir | 487.00 | ¹⁴⁰ La | 809.88 | ²³⁸ U | 1761.00 | ⁹⁰ Sr-Y |
| 136.47 | ⁵⁷ Co | 311.90 | ²³⁹ Pu | 497.08 | ¹⁰³ Ru | 810.28 | ^{166m} Ho | 1771.50 | ⁵⁶ Co |
| 140.50 | ^{99m} MoTc | 312.17 | ²⁴¹ Pu | 511.00 | Annil | 828.00 | ²⁰⁰ Tl | 1836.00 | ⁸⁸ Y |
| 143.79 | HEU | 316.51 | ¹⁹² Ir | 528.96 | ¹²³ I | 834.80 | ⁵⁴ Mn | 1853.67 | ⁷⁶ Br |
| 158.97 | ¹²³ I | 320.08 | ⁵¹ Cr | 529.80 | ¹⁶⁶ Ho | 846.80 | n on Fe | 2032.30 | n on Li |
| 160.31 | ²⁴⁰ Pu | 328.46 | ¹⁹⁴ Ir | 559.08 | ⁷⁶ Br | 898.00 | ⁸⁸ Y | 2097.80 | ¹⁵⁶ Eu |
| 163.93 | ^{131m} Xe | 333.51 | ^{252, 249} Cf | 569.70 | ²⁰⁷ Pb | 900.73 | ¹⁷² Lu | 2184.00 | ¹⁵⁶ Eu |
| 167.40 | ²⁰¹ Tl | 340.60 | ²³⁷ Np | 583.20 | ^{228, 232} Th | 911.00 | ²³² Th | 2186.00 | ⁹⁰ Sr-Y |
| 171.28 | ¹¹¹ In | 340.81 | ²⁴¹ Pu | 609.00 | ²²⁶ Ra | 959.70 | ²⁰² Tl | 2223.25 | n on H |
| 181.56 | ¹⁷² Lu | 344.94 | ²³⁹ Pu | 632.99 | ¹⁸⁸ Re | 980.53 | n on Li | 2598.00 | ⁵⁶ Co |
| 184.58 | ⁶⁷ Ga | 348.13 | ¹⁵⁹ Gd | 637.00 | ¹³¹ I | 991.56 | ⁶⁴ Ga | 2614.53 | ^{228, 232} Th |
| 185.70 | HEU | 351.87 | ²¹⁴ Pb | 642.33 | ²⁴⁰ Pu | 1001.00 | ²³⁸ U | | |

III. Methods

To determine the resolution needed to obtain a given precision, the work of Ryder (2) can be used. Based on this work, Terracol gives the formulas for peak variance as:

$$\sigma_1^2 = aB + bN_1 + cN_2$$

$$\sigma_2^2 = aB + bN_2 + cN_1$$

Where: B is the background under the peaks
 N_i is the net peak area at energy E_i
 a, b, c are functions of resolution and peak separation

Both Ryder and Terracol express precision or sensitivity as: $\frac{\sigma}{N}$. A representative plot, based on Ref 1 (see Fig. 1), shows the $\frac{\sigma}{N}$ vs peak separation for several resolutions, all other parameters, such as background per unit energy and number of counts in the net peak area are constant. For a given

resolution, the precision improves with increasing separation to a limit and then remains constant. Note also in this figure that the limit increases with resolution because of the inclusion of more background counts under the peak region.

Previous works (9, 10) on MDA in relation to false positives and false negatives used the inverse of the precision defined above for the peak quality parameter (Q). A plot of Q for the conditions of Fig. 1 is shown in Fig. 2. Again, Q reaches a limit where increases in peak separation do not make improvements and the actual value of the limit depends only on the

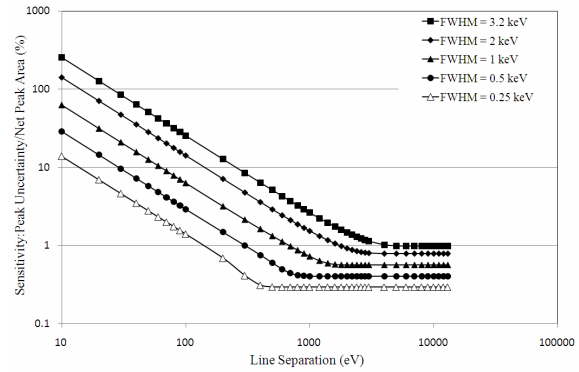


Figure 1 Spectrometer Sensitivity as a Function of Resolution for Two Peaks of Equal Amplitude with Background

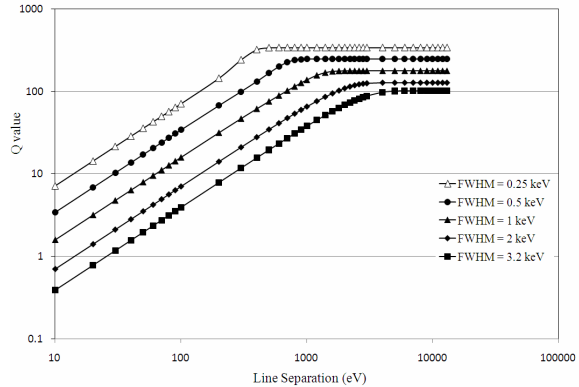


Figure 2 Detector Q value as a Function of Resolution for Conditions of Fig. 1.

resolution. That is, for line separations on the sloping part of the curve, higher efficiency (equivalent to longer counting times), to improve the counting statistics, will not overcome the resolution deficiency.

IV. Examples

In the above example, only the resolution and line separation are varied, that is, the net peak area and background are the same for both detectors. This is not always the situation especially when evaluating different types of detectors, such as HPGe and NaI. Two cases are discussed here: Handheld Radioisotope Identifiers (RIIDs) and Spectroscopy-based Portal Monitors (ASP).

Handheld RIIDs

A situation close to the assumptions in Ref 2 is the Handheld RIID. The ORTEC Detective was designed to have an HPGe detector with approximately the same low-energy absolute “geometric” efficiency as the commonly used NaI-based RIIDs at that time. It has a HPGe crystal of 50mm diameter and 30 mm length.

Of high interest for RIIDs is the identification of ^{235}U (HEU) in the presence of masking radiation. HEU could be masked with ^{67}Ga (a nuclear medicine isotope) or ^{172}Lu (radiotracer uses). Both of which have gamma rays close to the principle gamma ray, 185 keV, of ^{235}U . Figure 3 shows the simulated spectra for ^{172}Lu and ^{235}U for the case of two peaks of equal net area on similar backgrounds. The two detector types are assumed to have the same absolute efficiency and backgrounds. Figure 4 shows the Q value of both detectors for this case.

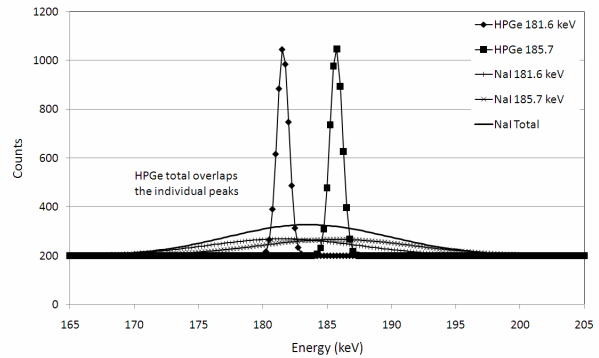


Figure 3 HPGe and NaI Spectra for ^{172}Lu and ^{235}U Peaks

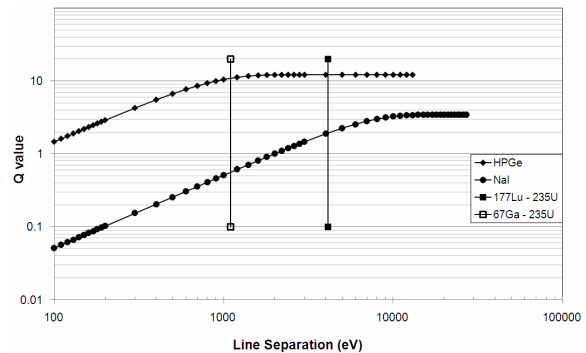


Figure 4 Q value for 185 keV Peak with an Interfering Peak of the Same Amplitude in HPGe and NaI Detectors of Similar Efficiencies and Backgrounds

Note that the NaI detector Q value reaches its limit at a much lower Q value than the HPGe detector. The Q value has reached its limit at the $^{172}\text{Lu} - ^{235}\text{U}$ separation for the HPGe detector, whereas it has not for the NaI detector.

Portal Monitor

The ORTEC IDM (9) was designed for portal monitor applications and has a detector of 85 mm diameter by 30 mm length. Many NaI-based portal monitors use 4 x 2 x 16 inch detectors oriented with the 4 x 16 inch side as the active area. Obviously, in this situation with the possibility of multiple detectors of any type, the assumptions of very similar efficiencies and backgrounds are not valid.

To compare these two detectors in a real world situation, the background count rate and efficiency at an energy, where there are close gamma rays, is needed. For this the background inside a building was collected with an IDM and a 4 x 4 x 16 NaI detector for 24 hours to get a long-time average background. At low energies, the gamma rays are totally absorbed in both detectors, so the efficiency for a far-field source is the surface area of the detector. In addition, portal monitors typically have a short data collection time, possibly only seconds, corresponding to the transit time of the truck or cargo container through the portal.

| Detector | HPGe | NaI |
|----------------------|-------|------|
| Energy (keV) | 375 | 375 |
| Resolution (keV) | 1.36 | 33.7 |
| Relative Efficiency | 1 | 7.27 |
| Background (c/s/keV) | 0.15 | 0.90 |
| Channel width (keV) | 0.185 | 2.99 |

Using these measured values, the Q values for both detectors can be used to compare the detectors in a real situation. For this example, the case of ^{239}Pu masked by ^{133}Ba was selected. The gamma-ray energies are 375 and 383 keV. The resolution of the HPGe detector was taken as the average of 10 IDMs or 1.36 keV. The resolution of the NaI was taken as 9% or 33.7 keV. The ratio of the two efficiencies is 7.27:1 (NaI:HPGe). The HPGe data were calculated for 16000 channels and the NaI was 1024 channels. The background in the 350 to 450 keV range is 0.15 counts/sec/keV for HPGe and 0.90 counts/sec/keV for NaI. These values are summarized in Table II. Using these assumptions, as well as the assumption of a pure Gaussian peak shapes, spectra can be generated to represent the situation. Such spectra for the case of the peak area from the masking nuclide at ten times the peak area of the target peak are shown in Fig. 5. The statistical variation in the channel data and the background are not shown. Figure 5 shows the complete separation of the two peaks for HPGe and the nearly complete overlap of the peaks for NaI. Note that the NaI total spectrum is dominated by the contribution of the mask even in the spectrum channels where the target gamma ray is most intense.

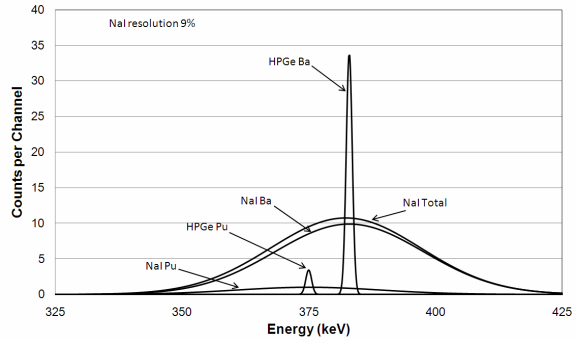


Figure 5 HPGe and NaI Spectra for 375 and 383 keV Peaks with 1:10 Relative Amplitude

The Q values (or peak sensitivity) calculated in the Ryder method assumes that the peak areas of the component peaks are well known. With this assumption, the Q values were calculated for this case in both HPGe and NaI for different peak area ratios as shown in Fig. 6. The peak areas and backgrounds are based on the values in Table II with a collection time of 20 seconds. Note that for HPGe the Q value doesn't change. This is expected since the HPGe peaks shown in Fig. 5 are completely resolved. For the NaI, the Q value is above the HPGe value for no mask due to the much higher absolute efficiency, but decreases rapidly with increasing mask. In actual spectra, the contributions of the

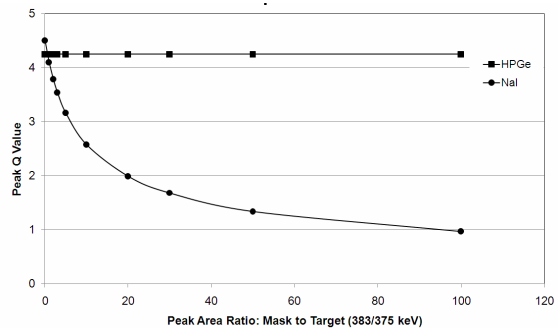


Figure 6 Q Value for Peak with Mask from 0 to 100 times Target Amplitude

individual components are not known and must be derived from the total spectrum. This will have a tendency to increase the net peak uncertainty, especially for low count/channel spectra due to the added uncertainty in the peak extraction process.

Choice of Q Threshold Values in an Instrument

The need to have Q threshold values as high as possible is based on the variation of Q for the null or background situation. Previous work with HPGe data (10) showed that for spectra with low counts per energy window, either from short data collection time or low count rates, Q has to be greater than 3.5 to meet the 1:10000 false-positive requirement. This value of Q would have a false negative rate better than 1:1000 for nuclide activities in the background such as ^{40}K . Figure 6 illustrates that the better resolution HPGe detector is able to detect and identify the target in the presence of the mask for all cases while the low resolution NaI detector would only operate adequately in the simplest masking cases.

It should be noted that the Ryder method predicts better peak quality values than realizable in practice because it does not include the uncertainty of the “extraction” of the peak area from the total spectrum. In addition, it does not account for the statistical variation in the data for low-count spectra.

Including both of these uncertainties will decrease the Q values further for the NaI detector faster than for the HPGe detector. The variation in the Q value for the null sample requires that Q be relatively large to have the false positive rate below 1:10000.

The Q values shown in Fig. 5 illustrate that gross count efficiency is not a good indicator of performance in trafficking situations. The presence of even small quantities of a masking nuclide rapidly eliminates any efficiency advantage of the NaI detector because the NaI detector Q value for the peak of interest is significantly reduce by the mask.

V. Conclusion

Using the formalism developed by Ryder for calculating the net peak area uncertainty for overlapping peaks, it has been shown that the resolution of HPGe detectors in RIIDS and portal monitoring systems is highly advantageous to reliable detection and identification of nuclides of interest

when masking nuclides are present. It has also been shown that the HPGe detector exhibits a large advantage in resistance to false positives by virtue of its intrinsically higher Q-values, which derive directly from the huge advantage in energy resolution. The use of high resolution detectors will help in solving the problem of false positives in the currently operating low-resolution portal monitors.

VI. References

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