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LOW-LEVEL GAMMA-RAY SPECTROMETRY

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Abstract

Low-level gamma-ray spectrometry generally equates to high-sensitivity gamma-ray spectrometry that can be attained by background reduction, selective signal identification, or some combination of both. Various methods for selectively identifying gamma-ray events and for reducing the background in gamma-ray spectrometers are given. The relative magnitude of each effect on overall sensitivity and the relative "cost" for implementing them are given so that a cost/benefit comparison can be made and a sufficiently sensitive spectrometer system can be designed for any application without going to excessive or unnecessary expense.

Introduction

Low-level gamma-ray spectrometry is, by definition, the ability to measure low levels of gamma radiation. This does not necessarily imply that a sample to be measured has only small quantities of radiation present in it; it simply means that it is necessary to measure the gamma-rays emitted by some particular radionuclide which is only present in small quantities. There may be other gamma-emitting radionuclides present in the sample in substantial quantities.

This discussion will describe the various mechanisms for selectively and nondestructively measuring small quantities of gamma radiation. Everything discussed is equally applicable to the measurement of x-rays and generally

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applicable to the measurement of any other type of radiation. Also, the discussion will use germanium spectrometers as the primary example, but the technology will in most cases be applicable to the use of NaI(Tl), BGO, plastic, or any other scintillation spectrometer. In addition, the discussion will assume a sample is being measured for some inherent radioactive component, although this interpretation may be very broad, such as the measurement of double-beta decay of ^{76}Ge in a sample that is the spectrometer itself.

In general, steps taken to improve the sensitivity of a gamma-ray spectrometry system for making any particular measurement fall into the category of either 1) selective signal identification or 2) background reduction. Selective signal identification involves mechanisms of observing only the gamma rays of interest, while background reduction removes potential interferences either generally or selectively in the region of interest. The balance of this paper will discuss various mechanisms for each, the relative benefit of each mechanism, and the relative "cost" of each mechanism.

Signal Identification

Selectively identifying gamma rays as emanating from a specific isotope is the simplest method of quantifying that isotope. The most fundamental method of identifying gamma rays emitted by a specific radioisotope is based on measuring the unique energy of those gamma rays. The more precisely and selectively that the gamma-ray energy can be determined, the more unique will be the fingerprint. Consequently, the better the energy resolution of the spectrometer, the more capable it is of selective signal identification. Therefore, given that all other factors are equal (efficiency, background, etc.), because of its superior energy resolution, a germanium diode spectrometer is superior to a NaI(Tl) spectrometer, which is in turn superior to a plastic scintillator.

The next logical step in selective signal identification concerns the use of coincidence/noncoincidence/anticoincidence techniques. Some radionuclides decay by gamma-ray cascades with time constants short relative to the electronic resolving time of typical instrumentation. The simplest way to

selectively measure this type of cascade is to measure the gamma rays incident on a spectrometer as a function of energy and in time coincidence with another gamma-ray event in another detector. The common example of this system is a germanium spectrometer surrounded by a NaI(Tl) or plastic scintillator coupled through an electronic gate such that energy spectral data from the germanium spectrometer is only accepted when it is in coincidence with an event gated by the NaI(Tl) or plastic. A beta detector can be added to the system as yet another gate or selection criteria.^(1,2) Bulk shielding between the detectors may also be used to prevent scattered single gamma rays from appearing to be coincidence events.⁽³⁾

The next level of sophistication involves energy gating the NaI(Tl) or plastic such that the energy spectrum in the germanium spectrometer is only stored when gamma rays of specific energy are observed in time coincidence. In this manifestation, a NaI(Tl) spectrometer is superior to a plastic scintillator because of its superior energy resolution. Such a system, for example, could be used to measure small amounts of ^{60}Co in a sample containing large concentrations of ^{40}K . By setting an energy gate from 1100 to 1400 keV in the NaI(Tl) spectrometer and recording the time coincidence energy spectrum incident on the germanium spectrometer, only the ^{60}Co will be observed. Of course, the maximum amount of information is obtained by utilizing the entire energy spectrum in the NaI(Tl) gating spectrometer, and splitting it electronically into many different energy intervals, corresponding to the isotopes of interest. Each of these energy interval gates would route the coincident germanium spectrum to a separate storage device. Only the electronic capacity of the system limits the number of energy selected spectra which can be simultaneously obtained and stored. Theoretically, the gamma rays from the decay of each isotope in a sample can be stored in a separate spectrum.

A corollary to the coincidence system is the noncoincidence system. The physical arrangement of the spectrometers can be the same and, in fact, both systems can be operated simultaneously. In the noncoincidence mode, only those gamma rays incident on the germanium spectrometer are stored which are not in coincidence with a gamma-ray event in the NaI(Tl) or plastic scintillator. In this manner, in principle only those radioisotopes that do not decay through multiple time coincident gamma-ray cascades are stored in this spectrum.

A system somewhat similar to the noncoincidence system is the anticoincidence system. This system is generally somewhat less sophisticated than the noncoincidence system but operates in exactly the same manner. The major purpose of this system is to cancel the storage of any event in the primary spectrometer which is in coincidence with an event in the secondary or guard counter. This is primarily a background reduction technique, rather than a signal selection technique. A major application of this technique is to provide Compton reduction by canceling events that scatter in the primary detector and deposit additional energy in the guard counter.

Another method of selective signal identification is based on geometry or directionality. The simplest example of this technique is the opposed coincidence measurement of the two 511-keV photons following positron annihilation. Another manifestation of this technique is the summing of the energy from coincident events in two or more detectors. This provides a mechanism for adding back together the total energy of a gamma ray that has undergone scattering in one detector and deposition of the balance of the energy in one or more additional detectors. In the most sophisticated version of this technique, multiple detectors are used to simultaneously record the energy of all coincident events, and the spatial arrangement of the detectors is folded into the analysis, à la computerized tomography, to determine if the coincident event is the sum of scattered depositions from a single gamma ray or if the event is a true coincidence of separate gamma rays.⁽⁴⁾

Nearly all of these highly sophisticated techniques can be combined with other techniques to maximize the selectivity/sensitivity of the identification. Another method for increasing the selectivity of gamma-ray measurements that can be used in concert or as a stand-alone method with either single or multiple detectors is time-tagged data. Each energy deposition in each detector is recorded along with its time of arrival. This type of data is most conveniently acquired and analyzed with a computer. The time tag itself can be adjusted from nanoseconds to minutes, depending on the type of information desired. Time tags can be used to selectively identify spurious events such as multiple pulsing, ringing, microphonics, etc. or to search for coincident events cascading through measurably long half-life excited states.

One extremely sophisticated technique that can be used to identify the origin of energy deposition events in a spectrometer is pulse shape

determination. This technique also requires a computer to achieve maximum effectiveness and usually involves digitizing the shape of the electronic output pulse from the spectrometer. Pulses generated by beta particles, for example, have a different characteristic shape than those generated by gamma rays, and this information can be used to selectively identify the event.

Almost all of these sophisticated event sorting mechanisms are predicated on fairly low event rates, since a great deal of electronic information must be accumulated and reduced. Hence, event rates should be limited to the order of 100 Hz.

Background Reduction

Whereas selective signal identification is very useful in identifying low levels of specific gamma rays from mixtures of radionuclides, background reduction is more important when the total activity of the sample is very low or when only a single isotope is present. Simply stated, the signal-to-noise ratio must exceed some threshold value for a positive determination to be made. The following discussion treats the various sources of background in descending order of importance and describes methods for eliminating or mitigating them. For the purpose of this example, a simple germanium spectrometer in a bulk shield will be used. Without a bulk shield, the background from the primordial radionuclides in the laboratory environment would dominate the spectrum and preclude any discussion of low-level measurements. Any of the signal selection techniques discussed in the previous section can be combined with background reduction to obtain the maximum effect, but all the background reduction efforts will necessarily apply to any hardware used. For example, a NaI(Tl) anticoincidence shield can be added to the germanium spectrometer, but the low-background effort must also then include the NaI(Tl) system as well, or else the background added by the anticoincidence shield may more than negate the positive effect of the shield itself.

The major source of background in most shielded laboratory-based spectrometer systems is cosmic rays.^(5,6) On the order of 90% of all background stems from the interaction of cosmic rays, primarily muons, in the detector, component parts, and bulk shield, which is typically lead. Not only

do the muon interactions themselves contribute to the background, but the secondary gamma rays also are a major contributor. Neutrons produced by spallation of the high-Z nuclei in the bulk shield contribute by both activation and excitation of the detector and component parts. A low-Z shield would be beneficial, but its size may be prohibitively large. High-density shields are used because a reasonable amount of shielding can be obtained in a compact space. Hence, the most practical method of minimizing the secondary neutron effects is to surround the detector with a high neutron capture cross-section material such as cadmium, boron, or lithium. Of these, cadmium and boron have prompt capture gamma rays themselves that will contribute to the background. Most of the neutrons are born fast and must be thermalized before they can be effectively captured by some agent. Hydrogenous materials work best for thermalizing, but they then produce 2225-keV gamma rays from neutron capture by hydrogen. Hence, some layered arrangement of bulk shield, moderator, and capture material will provide the best protection against neutron effects.

The primary effects of the muons themselves and the secondary gamma rays can best be diminished by use of an antic cosmic shield. This is similar to an anticoincidence shield but does not require the use of an expensive scintillator. In fact, relatively inexpensive plastic or liquid scintillator is preferred since it will also serve as the neutron moderating agent. By applying a blocking pulse of a few milliseconds following a cosmic muon interaction, the background of a typical laboratory spectrometer can be reduced by about an order of magnitude.

The next major source of background in a germanium spectrometer is the presence of primordial and manmade radionuclides in the component parts of the system.^(7,8) Included in this category are not only thorium, uranium, and potassium present in the various components but also manmade ^{60}Co in steel components, manmade ^{137}Cs from weapons and Chernobyl fallout in all components, ^{210}Pb in the bulk lead shield, and radon in the air. Recently, detector manufacturers have recognized the importance of careful selection of the raw materials from which the detector components are produced, and for only modest increases in cost, detectors can now be procured that have only very low levels of intrinsic radioactivity. It is also relatively inexpensive to prevent the natural radon and its progeny from contributing to the background of the system by simply piping the nitrogen gas boiling off the

dewar into the detector cavity and sealing the system adequately to maintain a positive pressure.

Most of the background reduction techniques discussed so far have been relatively inexpensive and effective. Backgrounds can be reduced more than two orders of magnitude using these simple steps. A system utilizing these background reduction efforts is shown in Figure 1, and a background spectrum from this system is shown in Figure 2. Additional background reductions of another four orders of magnitude are possible, but the cost/benefit ratio increases astronomically.

Another order of magnitude in background reduction over that shown in Figure 2 for the lower energy region can be obtained by shielding the detector from the contemporary bulk lead shield. Ordinary commercial lead bricks have significant concentrations of ^{210}Pb in them from the presence of uranium in the lead ore. Although the uranium is removed in the smelting and refining process, the equilibrium ^{210}Pb daughter is chemically inseparable and remains in the bricks. The 22.3-year half-life ^{210}Pb decays through an equilibrium daughter, ^{210}Bi , which decays with a high energy beta emission. This beta particle interacts with the high-Z lead and produces copious quantities of bremsstrahlung radiation and lead x-rays.⁽⁹⁾ A spectrum where this bremsstrahlung constituted the majority of the background is shown in Figure 3. Several potential inner shielding materials are available to separate the detector from the contemporary lead, but care must be taken that the chosen material does not introduce significant levels of its own background. The most obvious inner shield is lead that does not contain ^{210}Pb . This lead can be in the form of ancient lead where the ^{210}Pb has undergone sufficient radioactive decay to be insignificant, or in the form of currently available commercial lead bricks obtained from selected lead ores having extremely low concentrations of uranium. These commercial lead bricks are expensive. Ancient lead, from any source, will likely have to be cleaned and perhaps cast into suitable forms. Care must also be exercised so that these cleaning and casting operations do not introduce radioactive contaminants. Also, ancient lead may not be as free of uranium (radium) as currently smelted lead, and although the original equilibrium ^{210}Pb from the ore will have decayed, there may still be equilibrium ^{210}Bi from the uranium (radium) in the lead.

Although antic cosmic shielding in the laboratory is very effective in significantly reducing the background, it cannot be 100% effective in eliminating the effects of cosmic rays as long as cosmic muons are present. Cosmic muons cannot be totally eliminated, but substantial overburdens can reduce their effects to negligible levels. Installing the counting equipment in a mine, or tunnel, or even a hydroelectric dam can have profound effects on background reduction. Generally, this step is not financially expensive, but if an underground facility is not located proximate to the laboratory, the time burden for changing samples and tending the equipment may be prohibitive. A spectrum taken with a system similar to that shown in Figure 1 but located 37 m deep in a hydroelectric dam is shown in Figure 4. Another spectrum from a similar system located 670 m deep in an old iron ore mine is shown in Figure 5.

Since cosmic rays produce most of the background in surface spectrometers, and since a significant part of this background is due to the radioactivation of component parts of the spectrometer system, unless the system can be located underground, an equilibrium background of cosmogenic radioactivities will be persistent.^(8,10) Some material selection may be helpful in surface-based systems, but activation of the detector crystal itself cannot be avoided.

Even in cases where extreme care has been taken in all steps and the system has been shielded from cosmic rays nearly constantly from the time it was manufactured, some sources of background remain. Figure 6 is a spectrum taken with a system incorporating all of the above precautions and at a depth of 1480 m in an operating gold mine. The inner shield of this system was fabricated from 450-year-old lead recovered from a sunken Spanish galleon. Note that nearly all of the background up to 850 keV is from the ^{210}Bi bremsstrahlung in this old lead, presumably from uranium or radium impurities in the original cast ballast.

To what ultimate level can background be reduced? Let us assume that an inner-inner shield of copper can be electroformed in such a manner that all the cosmogenic radionuclides are removed and any additional formation of them is precluded. Then, by shielding the detector from the Spanish lead, the background up to 850 keV can be virtually eliminated. What about the rest of the spectrum? The major portion of the remaining spectrum, shown in Figure 7, will be due to cosmogenic ^{68}Ge in the germanium crystal itself.⁽¹¹⁾ The only

reasonable way to eliminate this source of background is to isotopically enrich the germanium in the heavy isotopes, depleting it in the light isotopes and eliminating the ^{68}Ge . This is, of course, enormously expensive, but it can be done. Even so, not all background will be eliminated! Figure 8 shows the remaining spectrum created by the double-beta decay of ^{76}Ge .⁽¹²⁾

Although it is possible to further isotopically separate the germanium which had been enriched in the heavy isotopes to eliminate the ^{76}Ge , leaving a detector composed of only $^{72,73,74}\text{Ge}$, there appears to be no practical reason for doing so. However, one must wonder what the remaining background would be.

Conclusion

Although virtually any level of background reduction is attainable, a practical choice must be made on a cost/benefit basis. If the primary use of the system will be to count environmental samples, it will do no good to reduce the system background below that which will be produced by the radioactivities in the sample itself.

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Figure Captions

- 1) A relatively inexpensive very-low-background germanium gamma-ray spectrometer for sample counting in a laboratory.
- 2) A background spectrum from the spectrometer shown in Figure 1.
- 3) A background spectrum from a germanium spectrometer enclosed in a contemporary lead shield. The dominant components of this spectrum are the bremsstrahlung and lead x-rays from the beta decay of ^{210}Bi in the lead shield.
- 4) A background spectrum from a system similar to that which produced the spectrum shown in Figure 2, except this system has 37 m of overburden.
- 5) A background spectrum similar to Figure 2 and Figure 4 but with 670 m of overburden.
- 6) A background spectrum from a system incorporating nearly all of the background reduction techniques described in the text and having 1480 m of overburden. The majority of the background up to 850 keV is from ^{210}Bi bremsstrahlung in the 450-year-old lead shield. The majority of the background above 850 keV is due to cosmogenic ^{68}Ge in the germanium crystal.
- 7) The ^{68}Ge component of the spectrum shown in Figure 6.
- 8) The "background" spectrum from the double-beta decay of ^{76}Ge in a germanium spectrometer.

Detector and Shield

Figure 1

Figure 2

Figure 3

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

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