Coincidence Summing Considerations When Using Marinelli-Beaker Geometries in Germanium Gamma-Ray Spectroscopy

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This article discusses some of the coincidence summing problems encountered when measuring aqueous samples in Marinelli beakers using germanium gamma-ray spectroscopy. Determination of the coincident gamma-ray radionuclides ^{134}Cs and ^{60}Co is discussed. Calibrations with the coincident gamma-ray radionuclides ^{88}Y, ^{60}Co, ^{133}Ba, ^{152}Eu, and ^{154}Eu are compared to essentially coincidence-free calibrations.

For these measurements, standards were prepared by adding 500 ± 2 mL of a liquid of known radionuclide concentration to 500-mL, plastic Marinelli beakers of the type described in ANSI/IEEE Std 680-1978. Gamma-ray measurements were performed using a 19.4%, 84-cm^3, p-type, closed coaxial, intrinsic germanium detector. Essentially coincidence-free standards for this particular detector/geometry combination were prepared using ^{241}Am (59.5 keV), ^{109}Cd (88 keV), ^{57}Co (122 keV), ^{139}Ce (165.9 keV), ^{203}Hg (279.2 keV), ^{113}Sn (391.7 keV), ^{137}Cs (661.6 keV), ^{58}Co (810 keV), ^{54}Mn (834.8 keV), and ^{65}Zn (1115.5 keV). Decay data used in this work was obtained from NIST or NCRP Report 58. The counting efficiency vs gamma-ray-energy data was fit to a 5th order polynomial with a maximum deviation of 3%. This curve fit was considered adequate for demonstrating the magnitude of the coincidence summing effects in measurements with Marinelli beakers.

Two coincident gamma-ray radionuclides, ^{134}Cs and ^{60}Co, are environmentally important as well as convenient for use in interlaboratory, cross-check programs.[1] General coincidence summing problems and specific coincidence summing problems encountered when measuring these two radionuclides on filter papers were discussed in the last issue.[2] In order to perform accurate measurements of ^{134}Cs and ^{60}Co in liquid samples using Marinelli beakers, coincidence summing effects should be investigated.

Liquid samples of ^{134}Cs and ^{60}Co were prepared in Marinelli beakers and measured by germanium gamma-ray spectroscopy using the efficiency curve from the coincidence-free standards. The measured activity of ^{134}Cs using the 605-keV gamma ray was only 85% of the known value. Obviously, coincidence summing correction factors must be known in order to accurately determine the activity of ^{134}Cs in unknown liquid samples in Marinelli beakers. As with the filter-paper geometry discussed in the last issue, coincidence summing correction factors will probably be different for each detector.

The measured activity of ^{60}Co in the Marinelli-beaker sample was 96% of the known value when using the 1173-keV gamma ray. The 1332-keV gamma ray was not used because it would have required an extrapolation of 220 keV past the highest energy point on the coincidence-free efficiency curve.

Turning from the measurement process to the calibration process, the coincidence-free standards described above suffer from two major deficiencies. First, the energy range is restricted by the highest energy gamma ray (1115.5 keV). Second, some of the components have relatively short half-lives. Several remedies to these deficiencies have been used but they involve radionuclides with potential coincidence summing problems.

The coincident gamma-ray radionuclides ^{60}Co, ^{88}Y, ^{133}Ba, ^{152}Eu, and ^{154}Eu have been used in various combinations to extend the energy range or the useful life of gamma-ray calibration standards. When using calibration standards containing any of these radionuclides in high-efficiency counting situations, coincidence summing problems should be assessed.

Marinelli-beaker standards, with known activities of the five, coincident gamma-ray radionuclides, were measured by germanium gamma-ray spectroscopy using the coincidence-free efficiency curve. As discussed above, the measured value for ^{60}Co (1173 keV) was 96% of the known value. Examination of the decay scheme for ^{60}Co indicates that the expected coincidence summing correction for the 1332-keV gamma ray is approximately equal to the coincidence summing correction for the 1173-keV gamma ray. For this detector both the 1173-keV and 1332-keV gamma rays from ^{60}Co require a 4-5% correction for coincidence summing.

The measured value for ^{88}Y using the 898-keV gamma ray was 98% of the known value. The 1836-keV gamma ray was not used due, again, to the large extrapolation. Examination of the decay scheme for ^{88}Y indicates that the coincidence summing correction for the 1836-keV gamma ray should be of the same order of magnitude as for the 898-keV gamma ray. The coincidence summing corrections for both the 898-keV and 1836-keV gamma rays for this detector/geometry combination are on the order of a few percent.

Figure 1 shows the ratio of measured-to-known values determined from the ^{154}Eu Marinelli-beaker standard. The following gamma rays were used: 123.1 keV, 247.7 keV, 591.8 keV, 723.3 keV, 873.2 keV, 996.3 keV, 1004.7 keV, and 1274.5 keV. The results in Figure 1 emphasize the fact that the coincidence summing factor varies for each gamma-ray transition. The results also show that in order to use ^{154}Eu for calibration in the Marinelli-beaker geometry, coincidence summing corrections—some as large as 17%—must be applied.

The ratios of measured-to-known values, determined from eight gamma rays of ^{152}Eu, are shown in Figure 2. Gamma rays at 121.8 keV; 244.7 keV; 344.3 keV; 444 keV; 778.9 keV; 964.1 keV; 1085.9 keV, and 1112.1 keV were used. These results show that coincidence summing corrections must be applied in order to use ^{152}Eu for accurate calibrations in the Marinelli-beaker geometry.

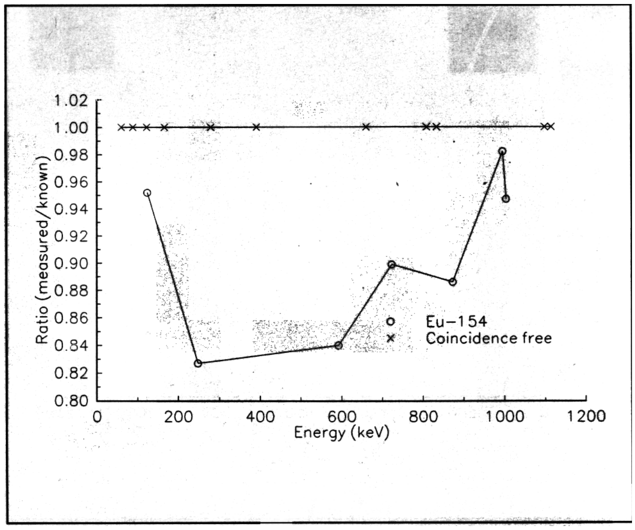

Figure 1 Effects of coincidence summing in the ^{154}Eu Marinelli-beaker standard.

Five gamma rays from ^{133}Ba (81 keV, 276.3 keV, 302.7 keV, 355.9 keV, and 383.7 keV) were measured to obtain the results in Figure 3. These results show that ^{133}Ba also requires coincidence summing corrections in the Marinelli-beaker geometry. Even though the 383.7-keV efficiency is very close to the coincidence-free curve in this case, the coincidence summing correction for that gamma-ray line should always be investigated carefully as there is a "summing in" contribution.[3]

The measured values presented here apply only to this particular detector and beaker and are presented only to indicate the order of magnitude of coincidence summing problems. More efficient detectors might be expected to have larger coincidence summing problems. Also, extended-range detectors with higher efficiency in the low-energy (less than 60 keV) range would be expected to have different coincidence summing characteristics due to the increased probability of x-ray/gamma-ray coincidence summing.

What can be done about coincidence summing problems in the Marinelli-beaker geometry? The

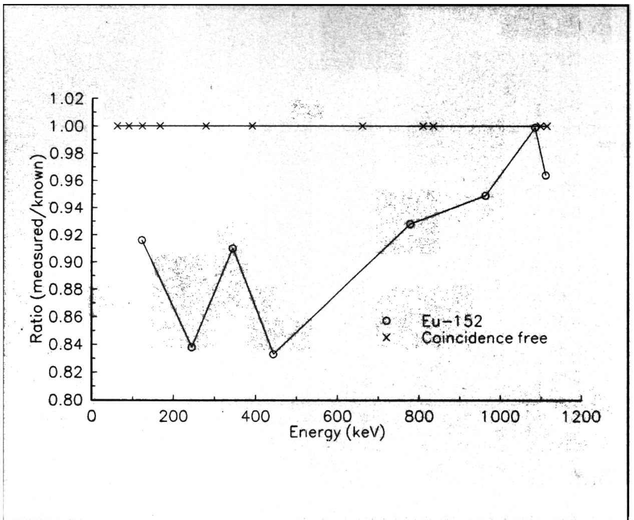

Figure 2 Effects of coincidence summing in the ^{152}Eu Marinelli-beaker standard.

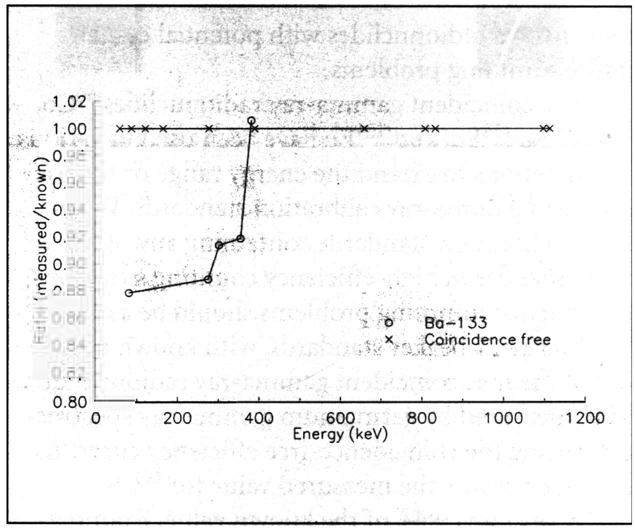

Figure 3 Effects of coincidence summing in the ^{133}Ba Marinelli-beaker standard.

ideal but generally impractical scheme would be to calibrate with certified standards of each radionuclide to be measured and use radionuclide-specific efficiencies, not efficiency curves. Another approach would be to calibrate with coincidence-free radionuclides and measure coincidence summing corrections with known (not necessarily certified) solutions of the coincident gamma-ray radionuclides to be measured. If these two schemes are not practical, efficiency calibrations can be performed with coincident gamma-ray radionuclides. Obviously, if the coincidence summing correction is known to be small compared to the acceptable uncertainty in a measurement, it can be ignored. Relatively small coincidence summing correction factors can be roughly estimated without adding significantly to the uncertainty in measurements. If calibrations are performed with coincident gamma-ray radionuclides known to have large (10-20%) coincidence summing effects, accurate corrections must be performed to obtain accurate measurements.

The easiest approach appears to be to calibrate with standards having little or no coincidence summing and either estimate the correction factors or neglect them and increase the uncertainty in the final measurements. Correction factors for radionuclides being measured (e.g., ^{134}Cs) can be directly determined. For this procedure solutions can be measured in-house using counting geometries which minimize coincidence summing. Aliquots can be transferred gravimetrically to Marinelli beakers and the coincidence summing correction factors derived. The alternative, accurate determination of the coincidence summing correction factors in the primary calibration standards, is a more complicated process. In the future, calculational procedures may prove valuable if the characteristics of each germanium detector can be accurately determined.[4]

References

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