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A METHOD FOR ASSESSING AND CORRECTING COINCIDENCE SUMMING EFFECTS FOR GERMANIUM DETECTOR EFFICIENCY CALIBRATIONS

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The addition of ^{54}Mn and ^{65}Zn to a nine-radionuclide standard (containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y, and ^{60}Co) provides the capability to determine the extent of coincidence summing for gamma rays from ^{88}Y and ^{60}Co. A method for correcting the efficiency points at 1332 keV (^{60}Co) and 1836 keV (^{88}Y) for coincidence summing is presented.

Efficiency calibrations of intrinsic germanium (Ge) detectors for analysis of gamma-ray emitters by spectrometric methods are generally performed with multi-line gamma-ray standards that cover the energy range of interest. Efficiency-energy pairs are generally processed with a fitting function that predicts the full-energy-peak detector efficiency (hereafter referred to as gamma-ray efficiency) as a function of gamma-ray energy. The most widely used multi-line gamma-ray standards are a nine-radionuclide mixture of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y, and ^{60}Co; and a three-radionuclide mixture of ^{125}Sb, ^{154}Eu, and ^{155}Eu, that were developed by the National Institute of Standards and Technology (NIST). The nine-radionuclide mixture is sometimes modified by the addition ^{85}Sr (514 keV) to provide an additional calibration point at 514 keV. The addition of ^{85}Sr is generally not recommended since the annihilation radiation from positrons produce 511-keV gamma rays that may interfere with the measurement of the 514-keV gamma rays as the ^{85}Sr decays to lower levels. Although both mixtures contain radionuclides that exhibit coincidence or cascade summing when counting in close proximity to the detector, the effects are generally much worse with the three-radionuclide mixture. For this reason the nine-radionuclide mixture is the preferred standard when counting samples in close proximity to the detector.

Coincidence or cascade summing (hereafter referred to as summing) occurs when two or more gamma rays originating from a single disintegration results in only one summed pulse.[1] Summing occurs for all coincident gamma rays that are detected including events which do not result in full-energy deposition such as scattered or Compton radiation. When two gamma rays sum, the apparent efficiency for each gamma ray is less than a gamma ray of the same energy with no coincidence summing. This is commonly referred to as "summing out". More complex cases of coincidence summing can occur with multiple gamma rays including "summing in" where the apparent efficiency for a gamma ray is greater. This occurs when two gamma rays in coincidence sum to give a photo peak with the same energy as one that occurs from a single transition. Only the simple case of two gamma rays in coincidence will be treated in this paper.

The effect of cascade summing with both multi-line mixtures is illustrated in Figure 1. Gamma-ray efficiencies are plotted versus energy for 47-mm diameter filter paper geometries

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prepared with both mixtures and counted at 0 and 10 cm from a 20% Ge detector. At 10 cm there are no apparent coincident summing effects and all points above 150 keV fall on a straight line for both mixtures. When the nine-radionuclide mixture is counted at 0 cm from the detector, the efficiencies show cascade summing effects for the gamma rays from ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1333 keV). These efficiency points fall below the line generated from the gamma-ray efficiencies with no cascade summing effects. When counted at 0 cm, the efficiencies determined with the gamma rays from the ^{125}Sb, ^{154}Eu, and ^{155}Eu standard exhibited serious summing effects and differed significantly from the expected efficiencies for noncoincident gamma rays. Attempts to fit the efficiency data points from the ^{125}Sb, ^{154}Eu, and ^{155}Eu source with a function commonly used for efficiency curves were unsuccessful (the least-squares fit diverged).

Coincidence summing is independent of the gamma-ray activity of the sample measured and is proportional to the detection efficiency of each gamma ray in coincidence. Corrections for summing are more important for high-efficiency detectors and geometries such as those commonly used for counting low-level environmental samples. Random summing which can

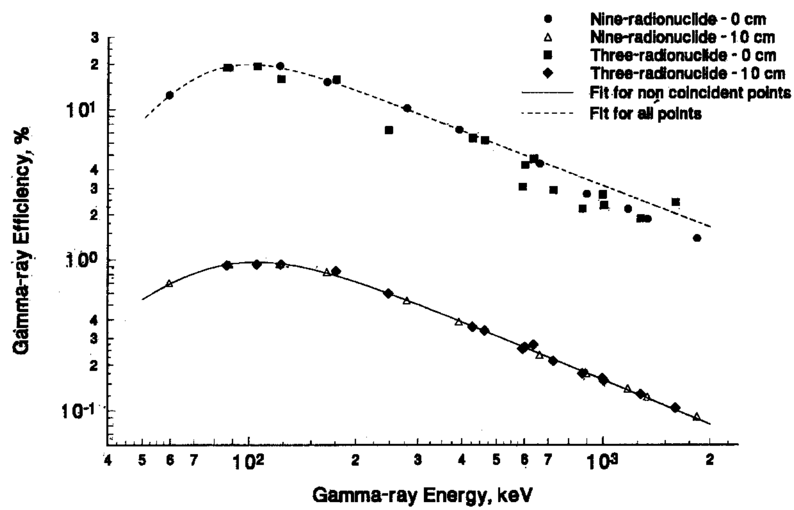

Fig. 1 Efficiency plot of three- and nine-radionuclide mixtures.

result in count rate losses is related to the count rate, but will not be treated in this paper. The coincidence summing correction factor, C_c is the multiplicative factor to convert a measured gamma-ray efficiency with summing effects to an efficiency with no summing effects. The coincidence summing correction factor can be calculated from the following equation [1]:

$$C_c = \frac{1}{1 - \sum_{i=1}^{i=j} f_i \cdot \varepsilon_t(i)} \qquad [1]$$

where f_i = the fraction of coincidence photons of energy i in coincidence with the gamma ray of interest, and $\varepsilon_t(i)$ = the total efficiency of the coincidence photon of energy i. For a simple two gamma-ray cascade like ^{60}Co the equation is:

$$C_{1173} = \frac{1}{1 - 1 \cdot \varepsilon_t\,(1333)} \qquad [2]$$

where C_{1173} is the coincidence summing correction factor for the 1173-keV gamma ray from cascade summing with the 1333-keV gamma ray. In this case, $f_i = 1.0$ since every ^{60}Co decay results in a 1173- and 1333-keV gamma ray in coincidence.

In principle, the above equation can be used to calculate correction factors for summing effects; however, this approach requires a determination of the total efficiency for each coincident gamma ray. Since the total efficiency must be measured for each geometry with single radionuclide gamma-ray sources, this approach would be very time consuming and expensive. In this paper an empirical approach for correcting the measured efficiencies of gamma rays from ^{88}Y and ^{60}Co for cascade summing effects is presented. This method can be used in conjunction with routine geometry calibrations and does not require separate determination of total gamma-ray-detection efficiencies. The resulting efficiency curves represent photon efficiencies with no summing effects and would generate accurate data for gamma-ray-emitting radionuclides that do not have coincident gamma rays. The measurement of gamma-ray-emitting radionuclides with coincident gamma rays may require the measurement of specific radionuclide gamma-ray efficiencies if summing effects are significant.

Materials and Equipment

The gamma-ray spectroscopy system used in this work consisted of a 89.5 cm^3 p-type, intrinsic Ge detector with a relative efficiency of 20% interfaced to a 4096 channel Canberra Accu-Spec MCA card in an IBM-compatible computer. Data analysis including peak searches, efficiency calculations, and efficiency curve fitting was performed with Canberra's Accu-Spec Radionuclide Analysis Software.

The radionuclide sources used in this work were NIST-traceable standards from Analytics, Inc. with the exception of the ^{124}Sb, ^{154}Eu, and ^{155}Eu source which was prepared from a NIST solution. Filter paper sources with active areas of 47 mm in diameter were prepared from the nine-radionuclide mixture and the three-radionuclide mixture by the method described elsewhere.[2] Point sources for the determination of total gamma-ray efficiencies were prepared for each of the following radionuclides: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y, and ^{60}Co. A point source of the nine-radionuclide mixture plus ^{54}Mn and ^{65}Zn was used for determining cascade summing corrections.

Method for Determining Cascade Summing Corrections

This method employs a radionuclide standard which contains ^{54}Mn and ^{65}Zn added to the nine-radionuclide mixture to provide two additional gamma rays at 834.8 and 1115 keV. The ^{54}Mn and ^{65}Zn gamma rays have no coincidence summing and are close in energy to the 898- and 1173-keV gamma rays from ^{88}Y and ^{60}Co. The 834.8- and 1115-keV gamma rays are used with the gamma rays in the mixture that have no summing effects to generate an efficiency equation free of summing effects. The efficiency for a 1173-keV gamma ray free of summing effects is calculated from the equation and used with the measured efficiency to generate a correction factor for summing.

A source is prepared with the nine-radionuclide mixture plus ^{54}Mn and ^{65}Zn in the desired geometry, and the gamma-ray efficiencies are determined for all the gamma rays in the mixture. These data and a plot of the efficiencies are presented in Figure 2 and show the effect of summing. Above 150 keV the gamma-ray efficiencies of gamma rays with no coincidence summing fall on a straight line. The gamma rays from ^{88}Y and ^{60}Co (898, 1173, 1333, and 1836 keV) have coincident gamma rays and also fall on a straight line, but this line is about 18% lower due to losses in the full-energy photo peak from summing. For this particular geometry the 165.9-keV gamma ray appears to be about 6% below the expected efficiency. This can be attributed to ^{139}Ce x rays summing with the 165.9-keV gamma rays from the decay of ^{139}Ce. Since this a point source with a thickness of only a few mg/cm^2, the effect is maximized because the ^{139}Ce x rays are not significantly attenuated within the source. Coincidence summing effects with x-rays is generally not a problem for "p-type" Ge detectors since the 33- to 38-keV x rays from ^{139}Ce are greatly attenuated by the end cap and the sample. This effect can be eliminated by the addition of an appropriate thickness of metal absorber (i.e., Cu).[3]

The next step is to generate an efficiency curve (equation) with no summing effects. This is accomplished by deleting the ^{88}Y and ^{60}Co efficiencies and using only the gamma-ray efficiencies with no summing effects. An equation relating efficiency to gamma-ray energy is obtained using a fitting function and the measured gamma-ray efficiencies. The resulting efficiency equation and plot are shown in Figure 3. The efficiency for a 1173-keV gamma ray is determined from the equation and represents the efficiency for a 1173-keV gamma ray with no summing effects. The calculated value for a 1173-keV gamma ray was 3.08% compared to the measured value of 2.61%.

The ratio of the calculated efficiency to the measured efficiency, C_{1173} is the correction factor for cascade summing and given by

$$C_{1173} = \frac{\varepsilon_{1173}\,(\mathit{fitted})}{\varepsilon_{1173}\,(\mathit{measured})} = \frac{3.08\%}{2.61\%} = 1.18$$

Energy	Observed Efficiency, %	Calculated Efficiency, %	% Difference
59.5	14.50	14.73	1.59
88	22.48	22.56	0.36
122	23.84	23.24	-2.52
165.9	18.44	19.59	6.24
279.2	12.65	12.22	-3.40
391.69	8.84	8.64	-2.26
661.6	5.26	4.99	-5.13
834.8	4.14	3.91	-5.56
898.03	3.28	3.62	10.37
1115	3.15	2.88	-8.57
1173.2	2.61	2.73	4.60
1332.5	2.23	2.39	7.17
1836	1.64	1.70	3.66

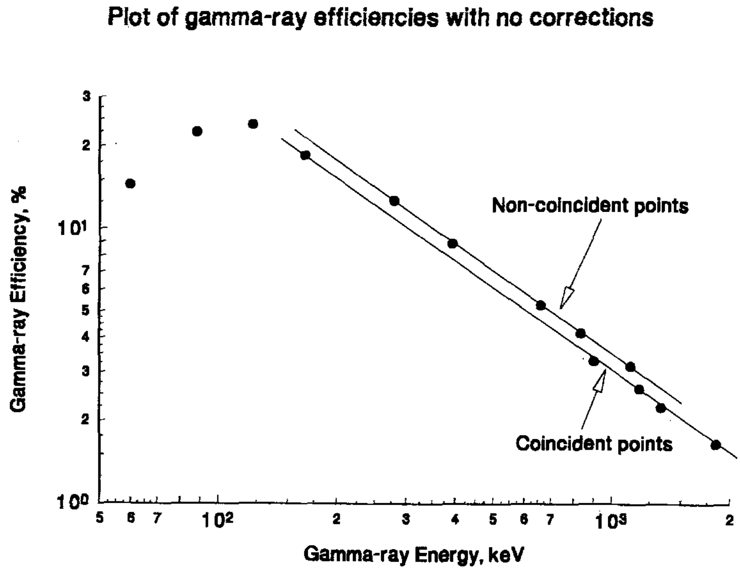

Fig. 2 Point source efficiencies for mixed gamma-ray source plus ^{54}Mn and ^{65}Zn in contact with detector.

Energy	Observed Efficiency, %	Calculated Efficiency, %	% Difference
59.5	14.50	14.28	-1.52
88	22.48	23.12	2.85
122	23.84	23.09	-3.15
165.9	18.44	19.07	3.42
279.2	12.65	12.08	-4.51
391.7	8.84	8.78	-0.68
661.6	5.26	5.32	1.14
834.8	4.14	4.26	2.90
1115	3.15	3.23	2.54

$$Efficiency\ (\%) = \frac{1}{0.37662 \cdot En^{.95645} + 99111 \cdot En^{-2.97897}} \cdot 100,$$ where En is given in keV.

where ε_{1173} *(fitted)* = 3.08% and ε_{1173} *(measured)* = 2.61% as described above. The experimental efficiencies for the 1333- and 1836-keV gamma rays are multiplied by the correction factor, 1.18, to obtain efficiencies corrected for summing effects. A new efficiency function or curve is generated with the corrected efficiencies for the 1333- and 1836-keV gamma rays and the efficiencies of the gamma rays with no summing. The result is an efficiency curve or equation over the energy range from 59.5 to 1836 keV that is valid for gamma rays with no summing effects. The data and efficiency plot utilizing the corrected efficiencies are shown in Figure 4.

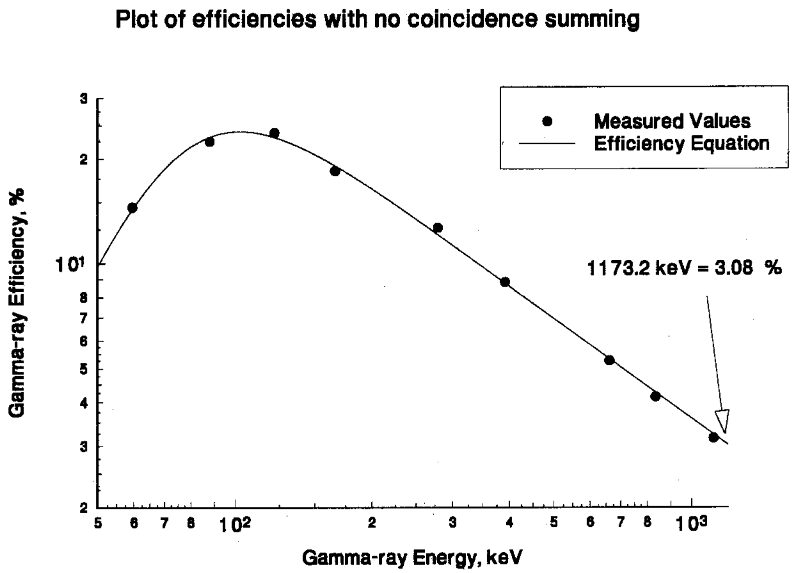

Fig. 3 Point source efficiencies for mixed gamma-ray source with no coincident gamma rays.

Discussion of the Method

The methodology described above for generating an efficiency curve that is free of coincident summing effects is based on two assumptions:

Assumption 1: The fitted efficiency for the 1173 keV accurately reflects the efficiency expected for a gamma ray of that energy with no summing effects.

Assumption 2: The correction factor for the 1173-keV gamma ray from ^{60}Co applies to the 1333- and 1836-keV gamma rays from ^{60}Co and ^{88}Y, respectively.

The validity of Assumption 1 is dependent on the uncertainty associated with the eight measured efficiency points and the representativeness of the resulting efficiency equation to the actual efficiency/energy relationship over the energy range of interest. The accuracy of the eight efficiency calibration points is dependent on the accuracy of the calibration source and the spectrum data reduction routines. In general the uncertainty associated with the efficiency measurements are less than about 5% at the three sigma level. The efficiency equation used in this work was determined by a nonlinear least-squares fit that determines four coefficients A, B, C, and D for the equation:

$$efficiency = \frac{1}{A \cdot M^B + C \cdot M^D} \qquad [3]$$

where M is the gamma-ray energy in MeV. In general the deviation of the fitted efficiency points from the experimental efficiency points generally ranged from 0-4%. Operational experience with this particular fit has shown that it accurately fits the efficiency equation for Ge detectors in this energy range from 50-2000 keV.

Assumption 2 can be stated mathematically as follows: $C_{1333} = C_{1173}$ and $C_{1836} = C_{1173}$. The validity of Assumption 2 was tested using equation [1] for the correction factors associated with ^{88}Y and ^{60}Co gamma rays. Since equation [1] requires the total efficiencies for the ^{60}Co and ^{88}Y gamma rays, the total detector efficiency for gamma rays as a function of energy was determined for the detector used in this work by counting a series of single radionuclide point sources. A plot of these efficiencies is given in Figure 5. An equation representing the total efficiency as a function of gamma-ray energy was determined using TableCurve[4] and is represented by the solid line in Figure 5. The total efficiencies for the 898-, 1173-, 1333-, and 1836-keV gamma rays were 0.168, 0.155, 0.150, and 0.142, respectively. Substituting these values in equation [1], the following correction factors were obtained:

Energy	Observed Efficiency, %	Calculated Efficiency, %	% Difference
59.5	14.50	14.27	-1.59
88	22.48	23.15	2.98
122	23.84	23.09	-3.15
165.9	18.44	19.04	3.25
279.2	12.65	12.03	-4.90
391.7	8.84	8.73	-1.24
661.6	5.26	5.28	0.38
834.8	4.14	4.22	2.03
1115	3.15	3.20	1.59
1332.5	2.60	2.63	0.58
1836	1.91	1.93	-0.38

$$\mathit{Efficiency}\ (\%) = \frac{1}{0.028573 \cdot En^{1.00329} + 23355 \cdot En^{-2.6229}} \cdot 100, \text{ where } En \text{ is given in keV.}$$

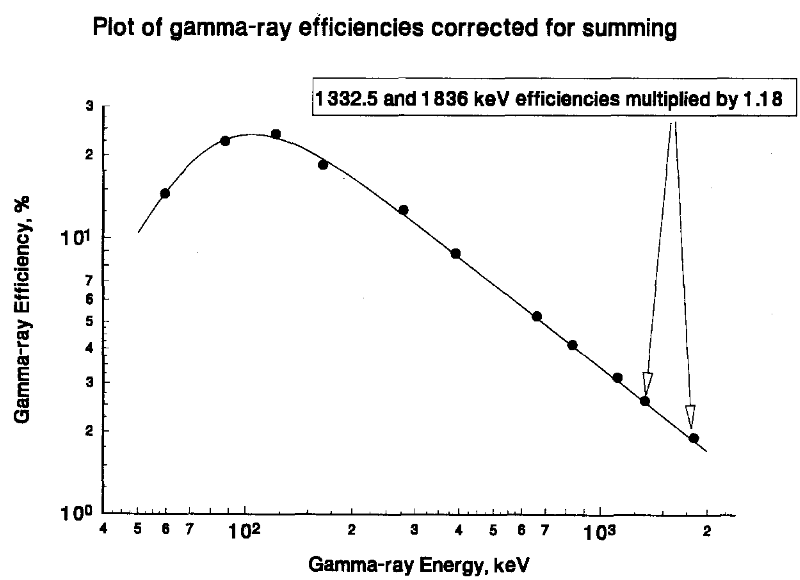

Fig. 4 Point source efficiencies for mixed gamma-ray source after correcting for coincidence summing.

$$C_{898} = \frac{1}{[1 - \varepsilon_t(1836) \cdot 1.0]} = \frac{1}{[1 - .142 \cdot 1.0)]} = 1.166 \qquad [4]$$

$$C_{1173} = \frac{1}{[1 - \varepsilon_t(1333) \cdot 1.0]} = \frac{1}{[1 - .150 \cdot 1.0]} = 1.178 \qquad [5]$$

$$C_{1333} = \frac{1}{[1 - \varepsilon_t(1173) \cdot 1.0]} = \frac{1}{[1 - .155 \cdot 1.0]} = 1.183 \qquad [6]$$

$$C_{1836} = \frac{1}{[1 - \varepsilon_t(898) \cdot 0.937]} = \frac{1}{[1 - .168 \cdot 0.937]} = 1.186 \qquad [7]$$

The correction factor obtained for the 1173 was within 0.5% of the factors for the 1333- and 1836-keV gamma rays. The correction factor for the 898-keV gamma ray was within 1.4% of the factors for the 1333- and 1836-keV gamma rays. This shows that the experimentally determined correction factor for the 1173-keV gamma ray will accurately predict the correction factors for the 1333- and 1836-keV gamma rays.

$$Efficiency\ (\%) = 24.9556 - .017428 \cdot En + .000269215 \cdot En^{1.5} + 80389.3 \cdot \ln(En)/En^2 - 309370.6/En^2$$

where *En* is given in keV.

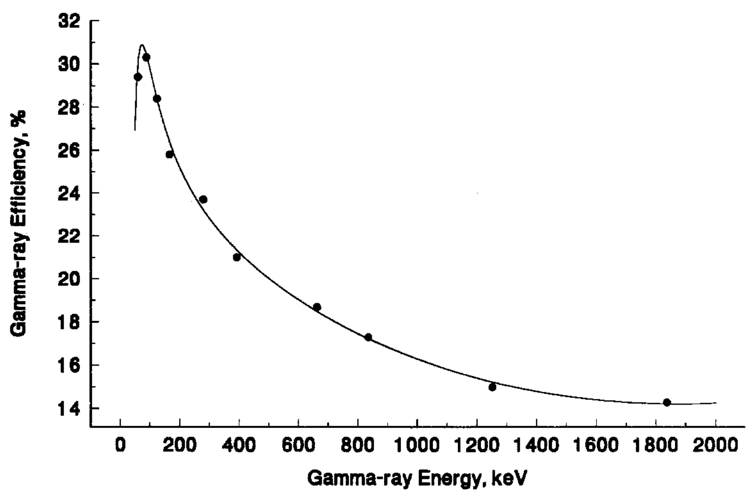

Fig. 5 Total gamma-ray efficiencies for a point source at 0 cm from a 20% Ge detector.

A correction factor can also be obtained for the 898-keV gamma ray using the measured and calculated values and is given by:

$$C_{898} = \frac{\varepsilon_{898}(fitted)}{\varepsilon_{898}(measured)} = \frac{3.98\%}{3.28\%} = 1.21$$

where $\varepsilon_{898}(fitted) = 3.98\%$ and $\varepsilon_{898}(measured) = 3.28\%$.

This value differed by 3.8% from the calculated value of 1.166 given in equation [4] for the 898-keV gamma ray and by 1.5% from the calculated values for the 1333- and 1836-keV gamma rays given in equations [6-7]. These data illustrate that the empirically determined correction factor for the 898-keV gamma ray could also be used for correcting the 1333- and 1836-keV gamma-ray efficiencies. The validity of using the correction factors for the 898- or 1173-keV gamma rays for the 1333- and 1836-keV gamma rays is related to the fact that the total gamma-ray efficiency does not vary greatly above about 1000 keV. Although the total efficiency for the 898-keV gamma ray is somewhat higher than the total efficiency for the 1836-keV gamma ray, the lower fraction of coincident gamma rays associated with the 898-keV gamma ray, .922, compensates for the higher efficiency (See equation [7]).

Conclusion

This method provides a means of assessing and correcting for coincidence summing effects as part of the normal calibration process with little additional effort. The addition of ^{54}Mn and ^{65}Zn to the nine-radionuclide mixture provides additional data points which should improve the overall fit of the efficiency function. This method was only tested on one detector; however, the technique should apply to other detectors, especially larger volume detectors. The total gamma-ray efficiency varies less with larger volume detectors since a larger fraction of the gamma-ray interactions with the detector will result in total energy deposition. A variation of this technique has been used for the calibration of a Ge well detector.[5]

The corrected efficiency curve and the data from the efficiency fit shown in Figure 4 demonstrate that this method for determining coincidence summing correction factors provides a better fit of the efficiency data than the uncorrected efficiencies and yields an efficiency curve with the expected shape (linear on a log-log plot above 200 keV).

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