



Correlation effects in gamma spectroscopy efficiency calibrations and their impact on activity and uncertainty quantification

John M. Kirkpatrick¹ · Troy Anderson¹ · Babatunde Oginni¹ · Henrik Persson¹ · Kara Phillips¹

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Abstract

The requirements of high-precision gamma ray spectroscopy applications increasingly demand greater accuracy from analysis software, in terms of both the determination of nuclide activities and the quantification of measurement uncertainty. To this end, we report on recent work to implement enhanced analysis algorithms for the Genie 2000 software suite that account for the most important effects of correlations between analysis input data in a statistically rigorous way. These effects primarily arise through the calibration procedure, wherein a mathematical function is fit to a set of measured efficiency data points, inevitably resulting in correlations between calculated efficiency values at different energies. While these effects are often relatively small, and thus have historically been considered of minimal significance, they can have important effects on activity results in applications where high precision is called for, especially for multi-line nuclides. The impact of correlations on uncertainty quantification is often more apparent, and may be significant even when precision requirements are not as stringent. Correlation effects are often particularly noticeable in cases where the measured efficiency data are themselves correlated, as when calibration measurements are performed using sources containing multi-line nuclides. In this paper, we discuss the physical mechanisms by which correlations are introduced and describe the mathematical methods by which they are evaluated and propagated by the new algorithms. Quantitative examples are presented to demonstrate the improvement on analysis output.

Keywords Data correlations · Gamma spectroscopy · Efficiency calibration · Uncertainty quantification

Introduction

As part of ongoing efforts for continuous software improvements, work is currently under way to upgrade the internal uncertainty propagation of Mirion Technologies (Canberra)'s Genie 2000 gamma analysis software [1] to include statistically rigorous treatment of the most significant correlation effects. Correlations arise in gamma ray spectroscopic analysis whenever fitting methods are employed, introducing covariant parameter sets, or when input data are correlated. These effects are often small, but their impact on uncertainty estimates and activity quantification can be important for high precision applications where other sources of uncertainty are well controlled.

The primary source of correlation effects is the measured efficiency calibration, in which a parameterized function is fit to a convenient set of measured efficiency data points. This function can then be used to calculate efficiencies at energies of interest differing from the calibration measurements. Correlations between the regression parameters must be taken into account in evaluating the uncertainty of calculated efficiency values. These also introduce correlations between the efficiencies calculated at different energies, with consequences for downstream analysis steps. Activities for multi-line nuclides, calculated as the weighted mean of measured individual line activities, are also affected by correlations between efficiency values. A more complex instance of this effect applies to the unfolding of interferences between nuclides which share gamma emissions at common energies.

Further complexity arises if the measured efficiency data points with which the calibration is performed are themselves correlated, altering the covariance structure of the fit

✉ John M. Kirkpatrick
jkirkpatrick@mirion.com

¹ Mirion Technologies (Canberra), Inc., 800 Research Parkway, Meriden, CT 04650, USA

parameters. This occurs when multi-line nuclides are used in the calibration reference source and when multiple data points share a dependence on the activity of a common nuclide.

To properly account for these effects, new Genie 2000 Efficiency Calibration and Nuclide Identification (NID) algorithms were prototyped using the Python programming language [2]. The primary modifications to both algorithms consisted of (a) generalizing the least squares fitting methods to correctly apply the full covariance matrix between the input data points and (b) saving the full output covariance matrix between the fit parameters to the spectral data file for later use. The effects of these changes on analysis results were demonstrated on simulated spectra created using MCNP-CP [3]. Spectra for several multi-line nuclides were generated singly and in combination, for a point source positioned 30 cm from an n-type HPGe detector with 45% relative efficiency. The spectra were analyzed using standard Genie 2000 algorithms except for the efficiency calibration and NID where the prototype algorithms were used.

Correlations in efficiency calibrations

In a measured efficiency calibration, efficiency data triplets $\{\varepsilon_i, E_i, \sigma(\varepsilon_i)\}$ are fit with a parameterized regression function of linear form

$$y(x) = \sum_{\alpha=0}^n a_{\alpha} f_{\alpha}(x), \quad (1)$$

with basis functions $f_{\alpha}(x)$ and parameters a_{α} . Here x and y may be the efficiency and energy, or their logarithms, depending on the choice of regression function.

The fit is performed by minimizing with respect to the parameters α the Chi squared function, given in vector notation by

$$\chi^2 = (\vec{y}(\vec{x}) - \mathbf{F} \cdot \vec{a})^T \cdot \mathbf{V}^{-1} \cdot (\vec{y}(\vec{x}) - \mathbf{F} \cdot \vec{a}) \quad (2)$$

where \vec{y} is the vector of measured data, \vec{a} is the parameter vector, and \mathbf{F} is a matrix of basis functions with $F_{\alpha i} = f_{\alpha}(x_i)$. The weight function \mathbf{V}^{-1} is the inverse of the covariance matrix of the data; for independent data it is diagonal and (2) becomes the usual Chi squared for uncorrelated fitting.

The outputs of the fit are the parameter values α as well as the parameter covariances, $\text{cov}(a_{\alpha}, a_{\beta})$, given by the inverse of the Hessian matrix of second derivatives of the Chi squared function. Parameter covariances appear even when the measured data are uncorrelated. They are necessary to correctly calculate both the uncertainties of values calculated from the calibration function and also the

correlations between values at different energy values x_i and x_j . In general,

$$\begin{aligned} & \text{cov}(y(x_1), y(x_2)) \\ &= \sum_{\alpha} \left\{ f_{\alpha}(x_1) f_{\alpha}(x_2) \sigma^2(a_{\alpha}) + \sum_{\beta \neq \alpha} f_{\alpha}(x_1) f_{\beta}(x_2) \text{cov}(a_{\alpha} a_{\beta}) \right\}. \end{aligned} \quad (3)$$

When multi-line nuclides are used in the calibration measurement, additional covariance effects occur. The efficiency ε_i measured at energy E_i from nuclide A is normally calculated as

$$\varepsilon_i = \frac{N_i}{I_{iA} A_A T} \quad (4)$$

where N_i is the peak area at energy E_i , I_{iA} is the gamma intensity for nuclide A at energy E_i , A_A is the activity of nuclide A, and T is the live time of the measurement. The variance of the measured efficiency is

$$\sigma^2(\varepsilon_i) = \varepsilon_i^2 \left\{ \frac{\sigma^2(N_i)}{N_i^2} + \frac{\sigma^2(I_{iA})}{I_{iA}^2} + \frac{\sigma^2(A_A)}{A_A^2} + \frac{\sigma^2(T)}{T^2} \right\}. \quad (5)$$

Neglecting the usually trivial uncertainty in the count time T , the covariance between efficiencies measured from two lines of the same nuclide A depends on the variance of the common activity:

$$\text{cov}(\varepsilon_i, \varepsilon_j) = \frac{\sigma^2(A_A)}{A_A^2} \varepsilon_i \varepsilon_j. \quad (6)$$

The data covariance matrix \mathbf{V} is constructed with diagonal elements (5), and with off-diagonal elements given by (6) for lines of a common nuclide or zero for lines of different nuclides. This methodology for handling correlated input data in the efficiency calibrations was described in greater detail in [4].

To demonstrate the effects of correlations in the measured data, simulated spectra were created to perform efficiency calibrations for three different calibration sources, with varying degrees of correlations, using standard Genie 2000 efficiency functions:

- A “Mixed Gamma” source containing seven single-line nuclides and two two-line nuclides shows the effect of a low degree of correlation in the fit;
- An “AmBaCsCo” source (^{241}Am , ^{133}Ba , ^{137}Cs , ^{60}Co) shows the fit with stronger correlations, as ^{133}Ba has six lines and ^{60}Co has two lines, while the other two nuclides have one line each;
- A ^{241}Am – ^{152}Eu source was simulated to show a high degree of correlation, as ^{152}Eu contributes 9 correlated lines, while ^{241}Am adds one uncorrelated point at low energy.

Table 1 Nuclide content and gamma energies of example calibration sources

Mixed Gamma		AmBaCsCo		AmEu	
Nuclide	Energy	Nuclide	Energy	Nuclide	Energy
²⁴¹ Am	59.5	²⁴¹ Am	59.6	²⁴¹ Am	59.6
¹⁰⁹ Cd	88.0	¹³³ Ba	79.6	¹⁵² Eu	121.8
⁵⁷ Co	122.1		81.0		244.7
¹³⁹ Ce	165.9		276.4		344.3
¹¹³ Sn	391.7		302.8		778.9
¹³⁷ Cs	661.7		356.0		867.3
⁵⁴ Mn	834.8		383.9		964.0
⁸⁸ Y	898.0	¹³⁷ Cs	661.7		1085.8
	1836.0	⁶⁰ Co	1173.2		1112.0
⁶⁰ Co	1173.2		1332.5		1408.0
	1332.5				

The nuclides in each calibration standard and the emission energies used for the calibrations are listed in Table 1. In all simulations, the uncertainty on each nuclide activity was taken to be $\pm 3\%$.

Figure 1 shows the effect of correlations on the fit for the three cases. On the top row, the ratio of the efficiency values with correlated data to the values without correlated data is shown. This variation is very small; on the order of 1% over most of the range of the calibrations, although larger values are observed at the extrema of the functions.

The uncertainties, calculated with and without correlated data, are plotted in the bottom row of Fig. 1. The energies and uncertainties of the efficiency data points used in the fit, as well as their nuclides of origin, are indicated by the points. When data correlation are taken into account the uncertainties are higher in regions where nearby data points are correlated, but tend to be slightly lower in regions between clusters of correlated data. The overall effect is greatest of ¹⁵²Eu, as expected. Neglecting correlations in this case would underestimate the uncertainty at most energies, by a maximum of around 1% (absolute) out of 3% near 1100 keV.

Correlation coefficients between different points on the curve are shown in Fig. 2 for the “mixed gamma” simulation, with correlations (right) and without (left). Strong correlation is seen in the region along the diagonal for both cases, as nearby energies are correlated even when the input data are not. Adding the effects of correlated data increases the region of positive correlation between the outputs, in this case primarily at higher energies, as seen in the right hand plot.

NID and interference correction

Weighted mean activities

Covariances in the efficiency calibration have consequences in the Nuclide Identification (NID) analysis step.

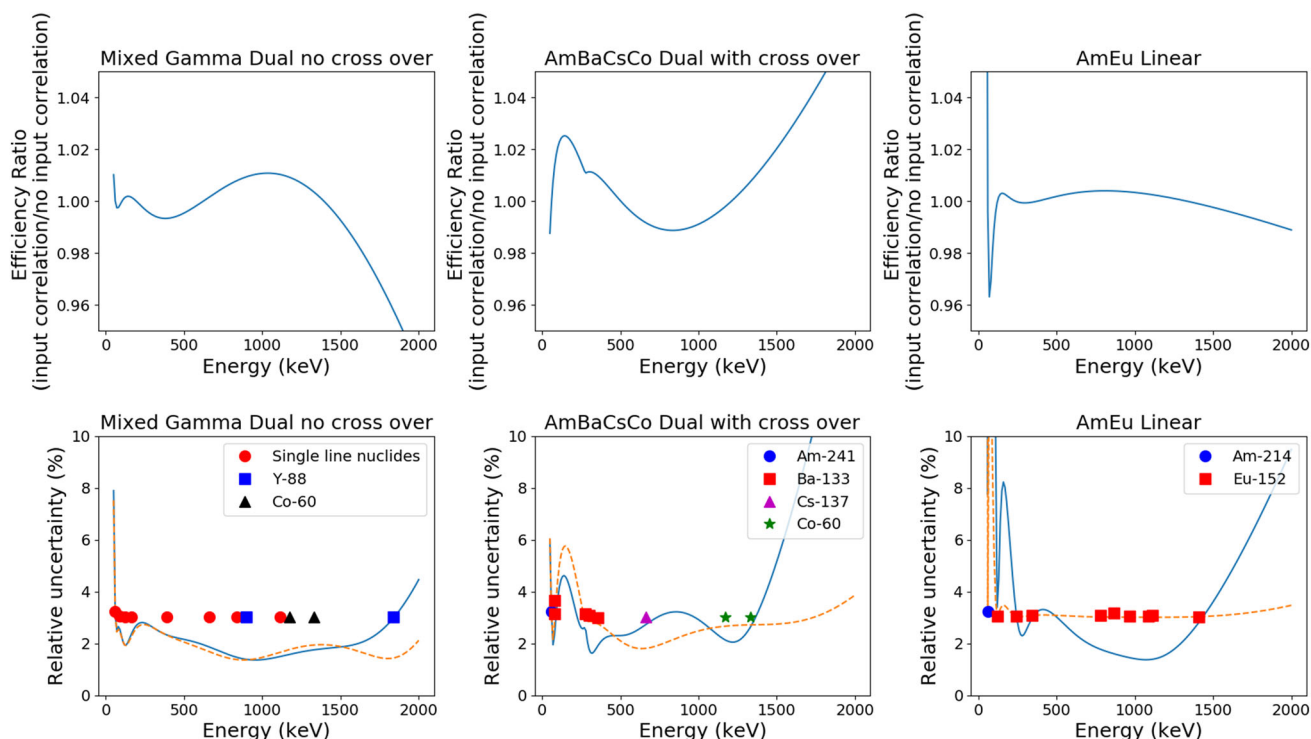


Fig. 1 Top: ratios of efficiency functions with and without data correlations. Bottom: calculated efficiency uncertainties with (solid line) and without (dashed line) data correlations. Energies and uncertainties of measured efficiencies and nuclides of origin indicated by points

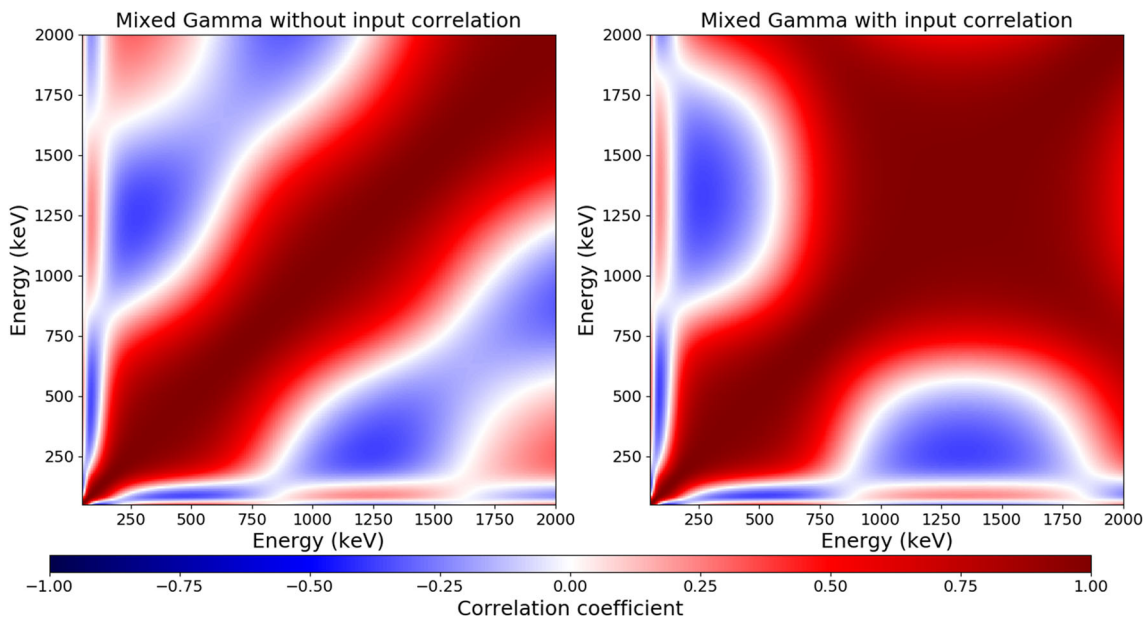


Fig. 2 Correlation coefficients between calculated efficiency values with data correlation effects (right) and without (left)

The weighted mean activities for multi-line nuclides are calculated from individual line activities, evaluated at different energies, which are correlated with one another through their common dependence on the efficiency calibration parameters.

The formula for calculating the weighted mean activity A with covariances is a special case of the least squares fitting problem:

$$A = \sum_{i,j=1}^n V_{ij}^{-1} A_j / \sum_{i,j=1}^n V_{ij}^{-1}. \tag{7}$$

If the individual line activities for nuclide A at energies E_i are calculated as

$$A_{iA} = \frac{N_i}{I_{iA} \epsilon_i T} \tag{8}$$

then the covariances between different lines are

$$\text{cov}(A_{iA}, A_{jA}) = A_i A_j \frac{\text{cov}(\epsilon_i, \epsilon_j)}{\epsilon_i \epsilon_j}, \quad i \neq j. \tag{9}$$

The matrix \mathbf{V} is constructed with off-diagonal elements given by (9) and diagonal elements given by the line activity variances. For uncorrelated data, the inverse covariance matrix \mathbf{V}^{-1} is diagonal.

To demonstrate these effects, spectra were simulated for several multi-line nuclides: ^{57}Co , ^{60}Co , ^{133}Ba , ^{134}Cs , and ^{152}Eu . The statistical (counting) uncertainties on the most significant lines were $< 1\%$. The spectra were analyzed in three different ways for each of the three calibrations created previously: with no correlation effects, with correlations considered between the sample nuclide line activities

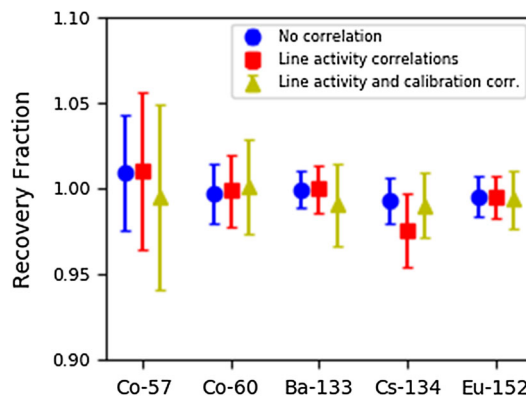


Fig. 3 Weighted Mean Activity recovery fractions for multi-line nuclides, using the Mixed Gamma calibration, for analyses including increasing levels of correlation

but treating the efficiency calibration data as uncorrelated, and including correlation effects between the sample line activities and between the efficiency data points from like nuclides. Weighted mean activities were calculated for each case.

The recovery fractions for each nuclide are plotted in Fig. 3 for all three analyses using the Mixed Gamma calibration—the results are similar for the other calibrations except as noted below. The effect on the activity result is modest, with the greatest impact seen for ^{133}Ba with a $\sim 1\%$ change (absolute) from the completely uncorrelated analysis to the most correlated. The more appreciable difference between the analyses is in the estimated uncertainties, which increase as additional levels of correlation are accounted for.

Table 2 Uncertainties in weighted mean activities for multi-line nuclides with different analyses for the Mixed Gamma calibration source

Sample nuclide	No correlations (%)	NID correlations (%)	NID and cal. input correlations (%)
⁵⁷ Co	1.6	2.0	2.0
⁶⁰ Co	1.3	1.7	1.9
¹³³ Ba	1.3	1.5	1.5
¹³⁴ Cs	1.0	1.6	1.5
¹⁵² Eu	0.8	1.1	1.1

Table 3 Uncertainties in weighted mean activities for multi-line nuclides with different analyses for the AmBaCsCo calibration source

Sample nuclide	No correlations (%)	NID correlations (%)	NID and cal. input correlations (%)
⁵⁷ Co	3.3	4.5	5.4
⁶⁰ Co	1.7	2.1	2.7
¹³³ Ba	1.1	1.4	2.4
¹³⁴ Cs	1.3	2.2	1.9
¹⁵² Eu	1.1	1.2	1.7

Table 4 Uncertainties in weighted mean activities for multi-line nuclides with different analyses for the AmEu calibration source

Sample nuclide	No correlations (%)	NID correlations (%)	NID and cal. input correlations (%)
⁵⁷ Co	2.8	3.0	3.1
⁶⁰ Co	1.4	1.6	3.1
¹³³ Ba	1.6	1.6	2.5
¹³⁴ Cs	1.1	1.7	3.1
¹⁵² Eu	1.0	1.1	3.0

Table 5 Emission energies of interfering nuclide demonstration set

Nuclide	Emission energies (keV)										
⁷⁵ Se	97	122	136	265	280	400					
⁵⁷ Co		122	136								
¹⁵² Eu		122	245		344	778	867	964	1086	1112	1408

The uncertainties obtained from each analysis are listed in Tables 2–4. For each calibration, the weighted mean activity uncertainties increase as correlations between lines of the sample nuclide are accounted for in each example. Adding in the effects of the correlations in the calibration standard has varied effect, depending on the degree of correlation for that standard. In Table 2, the least correlated calibration standard has little additional effect on the activity uncertainties when the correlations are included. The effect is somewhat more pronounced for the more correlated AmBaCsCo calibration results in Table 3, and even more so for the AmEu source in Table 4. In the AmEu calibration results in Table 4, the uncertainty for the weighted mean ¹⁵²Eu activity for example goes from 1.0% in the uncorrelated analysis, to 3.1% when all correlations are accounted for. This is in accordance with expectation; since the calibration was based on a ¹⁵²Eu source with 3% uncertainty, the activity uncertainties should not generally be smaller than that. This is true for all the activities except

¹³³Ba. This nuclide has strong lines in the region where the influence of the uncorrelated ²⁴¹Am calibration point reduces the degree of covariance in the efficiency curve.

Interference corrections

Interference correction is performed when one or more nuclides identified in the spectrum contribute to one or more common peaks. A least-squares fit optimizes the nuclide activities from the line activity data. The Chi squared function has the form given in (2), with data vector elements $y_i = N_i/(\varepsilon_i T)$, where N_i is the net peak area at energy E_i and ε_i is the efficiency. The basis functions are $F_{\alpha i} = I_{\alpha i}$, the gamma intensity of nuclide α at energy E_i . The parameters a_α are the nuclide activities. The data covariance matrix elements are given by

$$V_{jk} = \frac{N_j N_k}{\varepsilon_j \varepsilon_k T^2} \frac{\text{cov}(\varepsilon_j, \varepsilon_k)}{\varepsilon_j \varepsilon_k}. \quad (10)$$

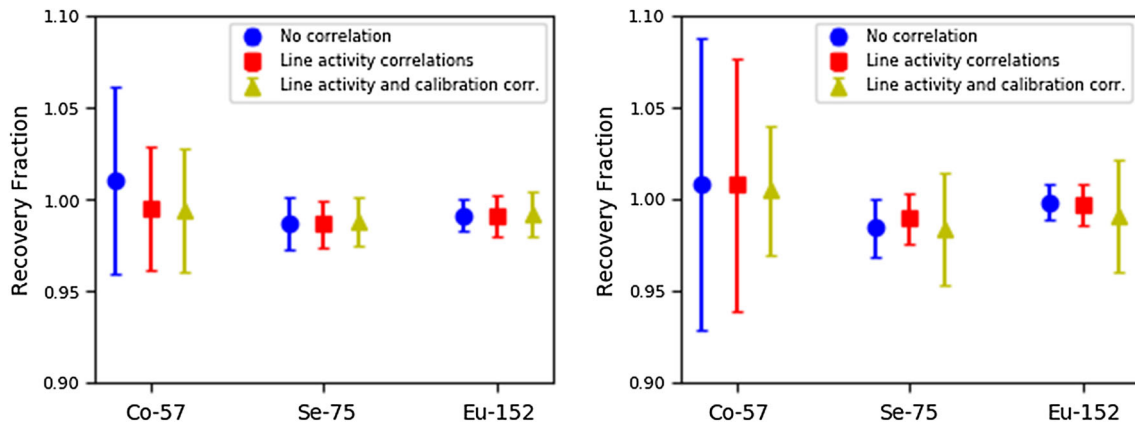


Fig. 4 Changes in recovery fraction and uncertainty when different degrees of correlations are accounted for. Shown are results of the Mixed Gamma (left) and AmEu (right) calibrations

Table 6 Uncertainties in weighted mean activities for multi-line nuclides, with different analyses, for the three calibration sources

Calibration standard	Nuclide	No correlations (%)	NID correlations (%)	NID and cal. input correlations (%)
Mixed Gamma	^{57}Co	5.0	3.4	3.4
	^{75}Se	1.4	1.3	1.3
	^{152}Eu	0.9	1.1	1.2
AmBaCsCo	^{57}Co	10.9	3.4	3.6
	^{75}Se	1.7	1.4	1.9
	^{152}Eu	1.1	1.2	1.7
AmEu	^{57}Co	7.9	6.8	3.5
	^{75}Se	1.6	1.4	3.1
	^{152}Eu	1.0	1.2	3.0

The prototype version of these new algorithms assumes all peak areas are uncorrelated.¹

To demonstrate the effects of correlations on the interference correction, MCNP-CP was used to simulate the spectrum of a sample containing three multi-line nuclides with interfering lines— ^{75}Se , ^{57}Co , and ^{152}Eu —which was then analyzed using the previously described calibrations. The emission energies of the three nuclides are listed in Table 5. Both lines of ^{57}Co are also present in the other nuclides, so that it is only possible to identify and quantify ^{57}Co through the interference correction.

The simulated spectrum was analyzed in three different ways, as described for the multiline nuclides. Recovery fraction results for the Mixed Gamma and AmEu calibrations are plotted in Fig. 4 for each analysis approach.

As observed for the Weighted Mean results without interference corrections, there is little change in the reported activities in most cases and the primary difference

is found in the uncertainties reported on the activities. For the directly observed nuclides ^{75}Se and ^{152}Eu , the error bars increase with the inclusion of additional degrees of correlation in the analysis. Somewhat counterintuitively, the uncertainty on the ^{57}Co activity, quantified by inference in the interference correction, decreases with added correlation. The correlations between line activities and efficiency values actually provide additional constraints on the fit, giving better precision for this activity that is not directly observable.

The relative uncertainties for each nuclide as obtained with all three calibration standards under each of the analysis assumptions described above, are listed in Table 6.

Conclusions

Correlations introduced through the efficiency calibration process can have important effects on the quantification of radionuclide activities and uncertainties. These can be particularly important in high-precision applications, where

¹ In principle, the areas of peaks fit as part of a common multiplet region will be correlated; however, at least in HPGe spectra, this will be a fairly uncommon circumstance for interfering nuclides.

measurement and sample geometry are well controlled and counting statistics do not dominate the uncertainties. Efforts are currently underway to incorporate more rigorous and complete uncertainty quantification in the Genie 2000 software suite by including correct handling of correlations. New spectroscopic analysis engines incorporating these changes have been prototyped to demonstrate the concepts and methodology. In the examples shown here, the prototype engines accurately reproduced the expected nuclide activities. The additional correlation effects from multi line calibration nuclides may affect line activities by as much as a percent or two, in normal applications, and generally will tend to increase the reported uncertainties for line and weighted mean activities. However these effects are somewhat complex and counterintuitive behavior was observed; for instance, the additional constraints provided

by correlation information can reduce the uncertainties in some regions of the calibration curve and for some interference corrected activities.

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