Mirion's Genie™ Spectroscopy Software: Implementation and Impact of Correlations in Efficiency Calibration and Radionuclide Activity Calculation

Introduction

In gamma spectroscopy, accurate reporting of activity results also requires accurate and defendable reporting of uncertainties. The uncertainty calculation includes many components, and one aspect of uncertainty calculations is correlations between quantities used in the calculations. For gamma spectroscopy, the most significant correlations are introduced during the creation of efficiency calibration. These become a topic of interest primarily in high precision measurements and when most other sources of uncertainty are well minimized. With Genie 4.0, the user now has the option to identify correlations during the efficiency calibration process and propagate the effects of these correlations to the reported activity and uncertainty. This application note discusses the impact of correlations on gamma spectroscopy, the changes in Genie 4.0 to support correlations in efficiency calibrations and nuclide activity results, and provides several key examples.

What are correlations and why do they matter?

Correlation is a mutual relationship between two or more things. Two measurements that are correlated will vary in a predictable way. For example, if one of the correlated measurements is higher than the true value, then information is gained about how the other measurement varies from the true value. A positively correlated measurement will also be higher than the true measurement. Conversely an anti-correlated measurement will be lower than the true measurement. Two non-correlated or independent measurements will not provide any additional information about each other.

Example of correlation in gamma spectroscopy: An efficiency calibration may be determined by measurement of the detector efficiency response with source standard of known emission rates, which are determined from the source standard's certificate activity and its stated uncertainties. If a true nuclide activity from the source standard is higher than the certified activity, the true emission rates for each emission lines from that nuclide will each be higher than the expected emission rate calculated from the certified activity. In practice, it is not possible to know if the true value is higher or lower than the certified activity, but it is possible to know how each emission rate calculated from the certified nuclide activity are related to each other.

Knowing whether measurements are independent or correlated matters when these measurements are used together to calculate a result. If the measurements are independent, adding more measurements to the calculation will reduce the uncertainty of the calculated result. For example, measuring the peak efficiency with emission lines from more source standards will improve the knowledge of the true peak efficiency, lowering the efficiency calibration uncertainty and ultimately lowering uncertainty reported radionuclide activities. In some circumstances, this can accurately result in reported radionuclide activity uncertainties even lower than the certificate uncertainty of a calibration source.

However, if the measurements are fully correlated, additional measurements will not improve the knowledge of the true value, and the uncertainty of the calculated value is not reduced. In the case of measurement of peak efficiency, adding additional measurement of fully correlated emission lines will similarly not improve the reported nuclide uncertainty. Correctly identifying and including correlations in gamma spectroscopy analysis can lead to more accurate results reporting.

Overview of efficiency calibration process and radionuclide activity calculation

To understand how correlations in a source standard can affect the results of an activity calculation, it is useful to review the full workflow from efficiency calculation to reporting of nuclide activity:

1. **Identify a source standard to be used for the efficiency calibration.**

   This source standard should be the same geometry as your expected sample and include documentation with a certified activity or gamma emission rate. Using the Genie Certificate File Editor, create a digital certificate that has the documented half-lives, emission rates, and associated uncertainties for the source standard.

2. **Count the source standard on the detector at the same location as your sample.**

   To minimize the impact of statistical counting uncertainty on the final activity results, the standard recommendation is to count to 10,000 counts in each peak of interest.

3. **Perform peak fitting analysis of the calibration spectrum.**

   The Genie software uses these calibration spectrum peak fitting results and the certificate file to calculate the efficiencies and their uncertainties at the calibration energies. True coincidence summing correction should be applied to the efficiency calculation when appropriate. The resulting "energy, efficiency, and efficiency uncertainty" values are often referred to as triplets.

4. **Select an efficiency calibration model type and create the efficiency calibration.**

   Use the triplets to employ a method of determining the efficiency and uncertainty at any energy between the lowest and the highest energy of the calibration source. A common way is to fit one or two calibration functions or models to the efficiency triplets. In Genie software, this is the selection of the Interpolated, Dual, Linear, or Empirical model.

5. **Count and analyze the unknown sample.**

   Ensure the sample is counted in the same geometry as the calibration standard. After performing peak analysis of the unknown sample spectrum, use the calibration model to calculate the efficiency and uncertainty at the energy of the peaks found in the spectrum.

6. **Calculate and report the radionuclide activity and activity uncertainty of the unknown sample.**

   Using the unknown sample peak analysis results, calculated peak efficiencies, library radionuclide decay data, and various other corrections, the Genie Nuclide Identification algorithm will determine the identified nuclides, nuclide activities, and nuclide activity uncertainties for the entire spectrum. It will also report the calculated activities at each radionuclide emission line.

During spectroscopy analysis calculations, inputs provided at each stage propagate to the final radionuclide activity results. Those inputs include the uncertainties and the treatment of those uncertainties from the calibration standard. If uncertainties in the calibration standard are treated as correlated, the uncertainty propagation throughout the entire workflow is affected.

Implementation of correlations in efficiency calibration and activity calculations

Part I: Identification of correlations from source certificate standards
Correlations can first be introduced into a gamma spectroscopy analysis during the efficiency calibration. As discussed earlier, if the emission lines used to generate an efficiency calibration are correlated, this can impact the users' knowledge of the calibration fit for that detector and geometry. Therefore, the user needs a way to indicate which emission lines are correlated for a given efficiency calibration. This is accomplished with an update to the Genie Certificate File Editor.
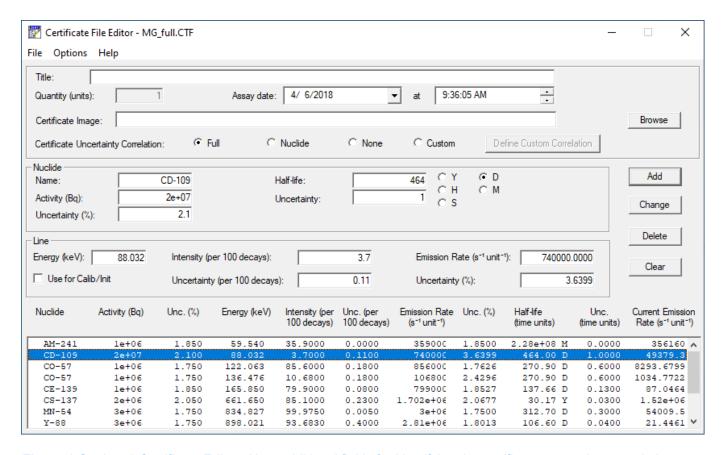
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Figure 1 Genie 4.0 Certificate Editor. Note additional fields for identifying the certificate uncertainty correlation.

The new certificate editor has these additional options and quantities:

- Selection of Certificate Uncertainty Correlation type:
  - o "Full" – This selection will treat the emission rates from all emissions in the source are correlated. This is the most conservative assumption and will give the largest uncertainty for the radionuclide activities calculated using an efficiency calibration made from this certificate file.
  - o "Nuclide" – This selection treats all emissions that originates from the same radionuclide in the calibration source as correlated by the decay-corrected radionuclide activity uncertainty of the source. For example, emissions from Co-60 or Eu-152 are correlated.
  - o "None" – This selection treats all emissions from the calibration source as independent. This option provides similar results to previous versions of Genie software.
  - o "Custom" – This option allows for manually selecting which emissions from the certificate are correlated and their relative correlation strength. This gives large flexibility of defining more complex correlations in the source.
- Activity and activity uncertainty is used as an input for the "nuclide" correlation option.
- Intensity and intensity uncertainty is used for to calculate the emission rate and uncertainty from the activity and activity uncertainty.

- Quantity uncertainty is the uncertainty in the quantity of the calibration source. This is used to as an input into the when the "Full", "Nuclide" or "Custom" correlation type is selected.

In the peak analysis of the calibration spectrum (Step 3 in the Analysis Workflow above), this information is used together with the peak area results to generate a covariance matrix that contains information about the correlations between the input parameters.

Part II: Propagating Correlations in the Efficiency Calibration Fit

To propagate correlations from the calibration source standards all the way to the radionuclide activity results, the algorithms used in Genie software during the Analysis Workflow Steps 4 and 6, creating an efficiency calibration fit and nuclide activity determination, have been updated in Genie 4.0. The model coefficients in the linear and dual efficiency calibration methods are determined using a least-squares optimization. Importantly, the χ^2 used in the fit has been up*dated from an independent equation to a correlated equation:

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$$\chi^2 = \sum_{i=1}^{n} \frac{(y_i - f(x))^2}{\sigma_i^2} \quad \rightarrow \quad \chi^2 = (\bar{y} - f(\bar{x}))^T V^{-1} (\bar{y} - f(\bar{x})) */$$

where y and σ are the efficiencies and efficiency uncertainties, $f(x)$ is the model function, and V is the covariance matrix of the efficiency triplets determined in Step 3.

These additions are used to define the covariance matrix, V, in the formula above used for optimizing the efficiency calibration model. The details of this calculation can be found in the Genie Customizations Tools Manual. The output of the optimization is the values of the coefficients that minimizes the χ^2-equation and another covariance matrix. At this stage, the correlations are between the optimized model coefficients, and Genie software stores this covariance matrix for use in the activity calculation. This covariance matrix is completely defined from the model optimization and there are no further inputs. Even if the efficiency calibration triplets used in the optimization are independent, the optimized coefficients will still be correlated from the model optimization and the covariance matrix will be stored.

Part III: Propagating Correlations in the Nuclide Identification and Activity Calculations

For activity calculations two changes were made to support the correlations from the efficiency calibration. The first change is to the weighted mean activity and uncertainty for multiline radionuclides:

$$A = \frac{\sum_{i=1}^{N} \frac{A_i}{\sigma_{A_i}^2}}{\sum_{i=1}^{N} \frac{1}{\sigma_{A_i}^2}} \rightarrow A = \frac{\sum_{i,j} V_{ij}^{-1} A_i}{\sum_{i,j} V_{ij}^{-1}}$$

$$\sigma_A = \sqrt{\frac{1}{\sum_{i=1}^{N} \frac{1}{\sigma_{A_i}^2}}} \rightarrow \sigma_A = \sqrt{\frac{1}{\sum_{i,j} V_{ij}^{-1}}}$$

Where A and σ_A are the radionuclide activity, A_i and σ_{A_i} are the emission activities for emission i, and V_{ij} is the covariance matrix from the correlation of the efficiency model evaluated at the energies of emissions i and j.

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The second change is to the interference correction algorithm, which uses a least squares optimization of the radionuclide activities to the measured peak areas. Correlations from the efficiency calibration and uncertainties in the intensities of the gamma emissions are now included in the optimization. The new χ^2 equation for interference correction optimization is:

$$\chi^2 = \left(\bar{y} - f(\bar{x}^*)\right)^T V^{-1}\left(\bar{y} - f(\bar{x}^*)\right) + \sum \frac{(x_i^* - x_i)^2}{\sigma_{x_i}^2}$$

where $\bar{y}$ is the ratio of the peak count rates and the efficiencies at the peak energy, x and σ_x are the intensities and intensity uncertainties of the radionuclides in the interference and $f(\bar{x}^*)$ is the sum of the contributions from all radionuclides to the peak count rates.

The new algorithm for activity calculation has been implemented in a new Genie nuclide identification analysis step called "NID with Correlations". Like the "NID with Interference Correction" step. The "NID with Correlations" step also performs interference correction. The effect of these changes will be highlighted in the examples discussed.

Considerations for determining correlations in efficiency calibration sources

A critical new input to the spectroscopy analysis is how the emissions from the efficiency calibration source are correlated. This is the certificate uncertainty correlation type, which is selected in the Genie Certificate File Editor. There are many considerations when deciding this. For a single radionuclide calibration source that emits multiple emission lines, such as Eu-152 or Ba-133, the source certificate typically lists the radionuclide activity and its uncertainty. This means that the emission rate from each of the emissions will be related through the radionuclide activity. In this case, the appropriate certificate uncertainty correlation is Nuclide.

For a calibration source that consists of many radionuclides, the choice of correlation is more complicated and may depend on how the source was manufactured and how the activities or emission rates were determined. The source manufacturer usually provides this information. A typical multi-radionuclide efficiency calibration source may be manufactured from a master solution of radionuclides with optional additional radionuclides. The activity or emission rates from the radionuclides may be determined by different calibration methods. A typical listing of radionuclides and calibration methods is shown in Table 1.

In this case all certificate uncertainty correlation options can be appropriate choices. If one assumes that all emissions from a single source are correlated, then "Full" is the appropriate choice. This is the most conservative choice and consequently, the efficiency uncertainty and the correlations between the efficiencies evaluated at different energies will be the highest of any of the choices. The relative uncertainty of the efficiency calibration fit will not be less than the relative uncertainty of the calibration efficiency triplet with the smallest relative uncertainty in the emission rate. This also means that the relative uncertainty of a radionuclide activity result for a measured unknown sample using this efficiency calibration will not be less than the relative uncertainty of the smallest relative uncertainty of the emission rate.

Another possible assumption is that the radionuclides that are calibrated using the same calibration method are correlated. In this case the "Custom" option should be chosen and the emissions lines with the same calibration method should be selected as correlated in the same group. This option will give lower relative uncertainty for the efficiency calibration than choosing "Full."

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A third option is to select "Nuclide", where the emissions from the same radionuclides are correlated, but each emission lines from different radionuclides are considered independent. In general, this will result in a lower relative uncertainty as compared to a selection of the "Full" or "Custom" method. Finally, "None" can be chosen to treat all emission lines in the calibration certificate as independent. This effectively ignores correlations in efficiency calibration, leading to efficiency uncertainties very close to results generated with previous versions of Genie software. This may also result in anti-correlation effects between efficiencies calculated at different energies and a lower radionuclide activity uncertainty for multiline radionuclides in the new "NID with Correlations" algorithm compared to "NID with Interference Correction" algorithm. The "NID with interference Correction" algorithm does not consider correlations from the efficiency calibration.

Table 1. Typical radionuclides, their calibration methods, and typical relative uncertainties used in multi-radionuclide efficiency calibration sources. This is used in the examples that follow.

Radionuclide	Calibration method	Typical relative uncertainty (%)
Am-241	LSC	1.90
Cd-109	HPGe	2.20
Co-57	HPGe	1.85
Ce-139	HPGe	1.75
Cr-51	Ionization chamber	2.15
Sn-113	HPGe	1.95
Sr-85	Ionization chamber	2.00
Cs-137	HPGe	2.10
Mn-54	Ionization chamber	1.85
Y-88	*HPGe	1.85
Zn-65	Ionization chamber	2.00
Co-60	HPGe	1.75

Examples

The effect of propagating correlations from the calibration source to the measured sample activity and activity uncertainty are best demonstrated using a few examples. These examples compare the activity and relative uncertainties for two methods. The first method is when correlations are included in the calculations, which includes defining correlations between emission lines of the calibration standard and propagation of correlations through the efficiency calibration and nuclide identification process. The second method is for when correlations are not considered; the calibration efficiency triplets are treated as independent, and the efficiencies calculated from the efficiency model at two different energies are also treated as independent.

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Example 1

In the first example the efficiency calibration is performed using an Eu-152 source standard with an activity uncertainty of 2%. The geometry is point-like and positioned at 30 cm from the detector. Then a sample with known activity is measured at the same geometry consisting of Co-60, Cs-134, Cs-137, and Eu-152. The detector used was a 50 cm^2 point-contact planar (BE5030) HPGe detector.

- Genie Certificate file is generated from the Library Extract option using a library that contains Eu-152 and the most recent decay data. The certificate activity uncertainty is set to 2%. For the method with correlated inputs, the Certificate Uncertainty Correlation type set to "Nuclide". For the method with independent inputs, the correlation type is set to "None".
- The calibration source standard is measured at the calibration geometry until statistical uncertainty of the major peaks for Eu-152 was at or below 1%. Efficiency calibration performed in Genie software using the "By Certificate" option.
- Sample with unknown activities is measured in the same geometry as the calibration standard. For the method with correlated inputs, the activities and uncertainties are calculated using the NID with Correlations algorithm. For the method with independent inputs, the NID with Interference Correction algorithm is used.

Table 2, The relative activity uncertainty and comparison of measured activity values of emission lines and radionuclides (in bold) when correlations from the source included and propagated in the calculations versus when all data are treated as independent.

Nuclide	Energy (keV)	Relative Activity uncertainty (%) using correlated inputs	Relative Activity uncertainty (%) using independent inputs	Ratio of measured activities with correlated inputs versus a method with independent inputs
Co-60	1173.2	2.2	1.5	0.997
Co-60	1332.5	2.2	1.5	0.996
Co-60		**2.1**	**1.1**	**0.997**
Cs-134	569.3	13.2	13.2	0.988
Cs-134	604.7	3.6	3.5	0.989
Cs-134	795.9	4.7	4.5	0.993
Cs-134		**3.2**	**2.7**	**0.991**
Cs-137	**661.7**	**2.3**	**2.2**	**0.990**
Eu-152	121.8	2.2	2.2	0.998
Eu-152	244.7	2.3	2.3	1.000
Eu-152	344.3	2.3	1.7	0.991
Eu-152	778.9	2.5	2.1	0.992
Eu-152	964.1	2.5	1.7	0.996
Eu-152	1085.8	2.4	1.8	0.997
Eu-152	1112.1	2.4	1.7	0.997
Eu-152	1408.0	2.3	2.2	0.995
Eu-152		**2.1**	**0.7**	**0.991**

Table 2 shows the ratio of the measured activity as determined by the Genie analysis method incorporating the correlation inputs and algorithms to the measured activity determined by Genie using a method that does not incorporate correlations. It also illustrates the calculated activity uncertainty for each of the two methods.

The first observation is that there is minimal difference between the activities calculated when the correlations are included and when they are treated as independent. The second observation is that the relative uncertainty is always larger when correlations are included in the calculations. This is true for both the activities for a single emission line and for the weighted mean activity of a radionuclide.

The largest difference between the two methods is observed for Co-60 and Eu-152, where the largest contribution to the activity uncertainty is the efficiency calibration. Most notably, the relative uncertainty of the activities for Co-60 and Eu-152 are above the relative uncertainty of the activity of the calibration source when correlations are included in the calibration. When correlations are not included and the calibration data is treated as independent data points, the relative activity uncertainty of these two radionuclide results are significantly below the relative uncertainty of the calibration source. This is consistent with the theory that when correlations in the calibration are included and the efficiency calibration uncertainty is dominating, adding more emission lines to the weighted mean calculation does not further reduce the propagated uncertainty because it can't be reduced below the uncertainty of the calibration source.

For the Cs-134 radionuclide results, the statistical uncertainties from the peak areas are larger than the uncertainty from the efficiency calibration. Calculating the radionuclide activity from the three most intense emission lines reduces the relative uncertainty of the radionuclide activity because the statistical uncertainty is independent and using all three emission lines increases the knowledge of the true activity, reducing the independent part of the overall radionuclide activity uncertainty. This is comparable to increasing the measurement time for a single emission. This effect happens for both methods, although there is a slight increase in the uncertainty for the correlated method.

Example 2

The second example is a 20 ml vial filled with water measured at 10 cm from 40% relative efficiency coaxial (GC40) HPGe detector. The efficiency calibration source is a mixture of the radionuclides shown in Table 1 with the relative uncertainties in the activities from the table. The sample contains known quantities of Co-57, Co-60, Cs-134, Cs-137. This example has three parts to illustrate how different assumptions of the correlations in the calibration source impact the sample activity results.

- Genie Certificate file is generated from the reference source certificate using the relative uncertainties from Table 1. For the method with correlated inputs, the Certificate Uncertainty Correlation is set as discussed in each specific example. For the method with independent inputs, the correlation type is set to "None".

- The calibration source standard is measured at the calibration geometry until statistical uncertainty of the major peaks for the calibration standard as shown in Table 1 was at or below 1%. Efficiency calibration performed in Genie software using the "By Certificate" option.

- Sample with unknown activities is measured in the same geometry as the calibration standard. For the method with correlated inputs, the activities and uncertainties are calculated using the NID with Correlations algorithm. For the method with independent inputs, the NID with Interference Correction algorithm is used.

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Example 2a

In the Example 2a, only the emissions from a common radionuclide in the calibration source are assumed to be correlated, i.e., the two emissions from Co-60 are correlated and the two emissions from Y-88 are correlated. For the correlated method, the Certificate Uncertainty Correlation option is set to "Nuclide."

Table 3, *The relative activity uncertainty and comparison of measured activity values of emission lines and radionuclides (in bold) when correlations from the source included and propagated in the calculations versus when all data are treated as independent. In this example, only emission lines from a common calibration radionuclide are considered to be correlated.*

Nuclide	Energy (keV)	Relative Activity uncertainty (%) using correlated inputs	Relative Activity uncertainty (%) using independent inputs	Ratio of measured activities with correlated inputs versus a method with independent inputs
Co-57	122.1	1.5	1.5	1.000
Co-57	136.5	7.8	7.8	1.000
Co-57		**5.2**	**4.9**	**1.002**
Co-60	1173.2	1.8	1.7	1.001
Co-60	1332.5	1.8	1.7	1.002
Co-60		**1.5**	**1.2**	**1.002**
Cs-134	569.3	12.4	12.4	0.999
Cs-134	604.7	2.8	2.8	0.999
Cs-134	795.9	3.5	3.5	0.999
Cs-134		**2.2**	**2.2**	**1.000**
Cs-137	**661.7**	**3.2**	**3.2**	**0.999**
Eu-152	121.8	1.6	1.6	0.999
Eu-152	244.7	2.9	2.9	1.000
Eu-152	344.3	1.9	1.9	1.000
Eu-152	778.9	3.0	3.0	1.000
Eu-152	964.1	3.1	3.1	0.999
Eu-152	1085.8	3.2	3.2	0.999
Eu-152	1112.1	3.4	3.3	1.000
Eu-152	1408.0	2.4	2.3	1.000
Eu-152		**1.1**	**0.9**	**1.003**

Observations: The radionuclide activities calculated with the two methods are near identical. Additionally, the relative uncertainties for the individual sample emission lines are near identical. This is because when applying the assumption that only the emissions from the same radionuclides are correlated, most of the triplets used for the efficiency calibration are treated as independent. The main difference in this case is that the relative uncertainty of Co-60 result is for the correlated method is higher than for the independent analysis. The relative uncertainty for Co-60 for the correlated method is still lower than the relative uncertainty of Co-60 in the calibration source because the triplets in the efficiency calibration close in energy to Co-60 are independent from the emissions of Co-60.

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Example 2b

The second example is the same configuration as above, but this time correlations in the calibration standard are defined by the calibration method for each radionuclide as indicated on the source certificate. The Certificate Uncertainty Correlation option is set to "Custom." The emissions using HPGe as calibration method are selected in Correlation Group 1 and the emissions with Ionization Chamber as the calibration method are selected in Correlation Group 2. A correlation strength of 1.00 is used for each group.

Table 4, *The relative activity uncertainty and comparison of measured activity values of emission lines and radionuclides (in bold) when correlations from the source included and propagated in the calculations versus when all data are treated as independent. In this example, the correlations are between calibration groups as indicated on the calibration source certificate.*

Nuclide	Energy (keV)	Relative Activity uncertainty (%) using correlated inputs	Relative Activity uncertainty (%) using independent inputs	Ratio of measured activities with correlated inputs versus a method with independent inputs
Co-57	122.1	1.6	1.5	1.006
Co-57	136.5	7.8	7.8	1.006
Co-57		**4.2**	**4.9**	**0.993**
Co-60	1173.2	1.8	1.7	1.003
Co-60	1332.5	1.8	1.7	1.003
Co-60		**1.6**	**1.2**	**1.002**
Cs-134	569.3	12.5	12.4	1.007
Cs-134	604.7	2.9	2.8	1.007
Cs-134	795.9	3.6	3.5	1.005
Cs-134		**2.4**	**2.2**	**1.007**
Cs-137	**661.7**	**3.3**	**3.2**	**1.006**
Eu-152	121.8	1.7	1.6	1.006
Eu-152	244.7	2.8	2.9	1.007
Eu-152	344.3	1.9	1.9	1.007
Eu-152	778.9	3.1	3.0	1.006
Eu-152	964.1	3.2	3.1	1.004
Eu-152	1085.8	3.3	3.2	1.004
Eu-152	1112.1	3.4	3.3	1.003
Eu-152	1408.0	2.4	2.3	1.003
Eu-152		**1.5**	**0.9**	**1.006**

In this example, there are more emissions that are correlated, and the correlated emissions are spanning a larger energy range. As before, the impact on the calculated sample activity is minimal. The largest increase in the relative uncertainty is for Eu-152 which has emissions with energies in almost the entire energy range of the efficiency calibration. For Co-57 the radionuclide activity uncertainty is lower with the correlations method because the main emission at 122 keV is interfered by Eu-152. This can cause the uncertainty to be lower compared to the independent method.

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Example 2c

The last example presents the same scenario as the previous two examples, except with the assumption that all emissions from the calibration source are fully correlated. The correlated method has the Certificate Uncertainty Correlation option is set to "Full."

Table 5, *The relative activity uncertainty and comparison of measured activity values of emission lines and radionuclides (in bold) when correlations from the source included and propagated in the calculations versus when all data are treated as independent. In this example, all emission lines from the calibration source is considered to be fully correlated.*

Nuclide	Energy (keV)	Relative Activity uncertainty (%) using correlated inputs	Relative Activity uncertainty (%) using independent inputs	Ratio of measured activities with correlated inputs versus a method with independent inputs
Co-57	122.1	2.0	1.5	1.006
Co-57	136.5	7.9	7.8	1.004
Co-57		**4.2**	**4.9**	**0.999**
Co-60	1173.2	2.2	1.7	1.000
Co-60	1332.5	2.2	1.7	0.999
Co-60		**2.0**	**1.2**	**0.998**
Cs-134	569.3	12.5	12.4	1.006
Cs-134	604.7	3.2	2.8	1.006
Cs-134	795.9	3.8	3.5	1.004
Cs-134		**2.7**	**2.2**	**1.006**
Cs-137	**661.7**	**3.6**	**3.2**	**1.005**
Eu-152	121.8	2.0	1.6	1.006
Eu-152	244.7	3.1	2.9	1.001
Eu-152	344.3	2.3	1.9	1.003
Eu-152	778.9	3.4	3.0	1.004
Eu-152	964.1	3.5	3.1	1.002
Eu-152	1085.8	3.5	3.2	1.001
Eu-152	1112.1	3.7	3.3	1.001
Eu-152	1408.0	2.7	2.3	0.999
Eu-152		**2.0**	**0.9**	**1.001**

When all emissions in the calibration standard are correlated, we expect that the relative uncertainty of the sample activity of all the emission lines and corresponding radionuclides to be larger than the lowest relative uncertainty emission in the calibration source. **Table 5** shows that the relative uncertainty for the correlated method does not go below 2.0%, which is larger than the lowest relative uncertainty of the calibration source. As before, it is worth noting that the relative uncertainty of Co-57 for the correlated method, which has its most intense emission interfering with Eu-152, has a lower relative uncertainty when using the correlated method than for the analysis without correlations.

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Additional considerations
Correlations when using ISOCS™/LabSOCS™ generated efficiency calibrations
When using ISOCS or LabSOCS generated efficiency calibrations, Mirion recommends using the interpolated efficiency model and calculating the efficiencies at the energies of the peaks using the Efficiency Correction by ISOCS step. Alternately, one can include use an energy list which calculates the efficiencies at enough energy points so that all features of the efficiency shape are captured. The NID with Correlations algorithm treats all ISOCS and LabSOCS efficiencies as correlated. For multi-line radionuclide samples, the calculated relative activity uncertainty will not be below the lowest relative efficiency uncertainty for a calibration emission line.

Interference correction algorithm and uncertainties in intensities
For interference correction, the uncertainties in the intensities have been included in the calculation of radionuclide activities and uncertainties. This is important when the radionuclide decay data is not well known and all emissions from a radionuclide have large uncertainties or when there are interferences with low intensity emissions from radionuclides with high activities. The low intensity emissions often have large relative intensity uncertainties. One current limitation of the implementation is that the uncertainties in the intensities are treated as independent. This may not be accurate when the uncertainties originate from the uncertainty of the normalization factor from relative intensities to the intensities per 100 decays of the parent radionuclide. This limitation may be addressed in a future version of Genie software.

Inconsistent data
When emission line activities for a common radionuclide are inconsistent with each other, a weighted mean calculation or a least square fitting with correlations calculation can result in an activity result that is outside the range of the individual emission line activities. In gamma spectrometry, this can lead to radionuclide activities that are lower than the lowest non-interfered emission activity. Common causes of inconsistencies for radionuclide activity calculations are missing interferences, non-optimal peak area calculations, or incorrect shape of the efficiency calibration. The NID with Correlations algorithm disregards emission lines in the weighted mean calculation and interference correction that are inconsistent with the other emission lines from the radionuclide or interference set. The disregarded lines are marked on the report and the user is encouraged to investigate the cause of the inconsistency.

Efficiency calibrations with multiple source measurements
Sometimes it is desirable to perform the efficiency calibration using measurements of more than one efficiency calibration source, for example when extending the energy range of an efficiency calibration or increasing the number of efficiency triplets in an energy region. When this technique is used in Genie software, the efficiency triplets from all measurements are used to optimize the efficiency model parameters. Correlations between triplets from the same measurement are determined from the user selection of Certificate Uncertainty Correlation type. Efficiency triplets from different measurements are treated as independent. Using more than one source measurement where the energy range of the emissions from the sources overlap can be used to reduce the uncertainty of the efficiency calibration.

ADDITIONAL READING

Genie Customizations Tools Manual – Mirion Technologies

Genie Operations Manual – Mirion Technologies

Correlation effects in gamma spectroscopy efficiency calibrations and their impact on activity and uncertainty quantification – Journal of Radioanalytical and Nuclear Chemistry (2018) 318:641–647 https://doi.org/10.1007/s10967-018-6087-7

Effects of efficiency correlations and intensity uncertainties on interference correction for gamma spectrometry – Journal of Radioanalytical and Nuclear Chemistry https://doi.org/10.1007/s10967-022-08447-2

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