Efficiency Optimization Employing Random and Smart Search Using Multiple Counts and Line Activity Consistency Benchmarks - 11398

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ABSTRACT

The ISOCS mathematical method developed by Canberra Industries is a well established technique for computing High Purity Germanium (HPGe) detector efficiencies for a wide variety of source shapes and sizes. In the ISOCS method, the user inputs the source dimensions, matrix composition and density, along with the source-to-detector distance. In many applications, the source dimensions, the matrix material and density may not be well known. Under such circumstances, the efficiencies may not be very accurate since the modeled source geometry may not be very representative of the measured geometry. Canberra is developing a customized efficiency optimization software, which is an extension of the ISOCS, known as "Advanced ISOCS" that varies the uncertain parameters and determines the optimal efficiency shape and magnitude based on available benchmarks in the measured spectra. The benchmarks could be results from isotopic codes such as MGAU, MGA, or FRAM, activities from multi-line nuclides, and multiple counts of the same item taken in different geometries (from the side, bottom, top etc). The efficiency optimization is carried out using either a random search based on standard probability distributions, or using numerical techniques that carry out a more directed (smart) search. Measurements were carried out using representative source geometries and radionuclide distributions. Benchmark data from multiple geometry counts of the same item, and activities from multi-line nuclides was used in efficiency optimization. Results of optimizations carried out using the random search method are presented in this paper. The radionuclide activities were determined using the optimized efficiency and compared against the true activities.

INTRODUCTION

Measurements with reference gamma sources have long been the preferred method for the efficiency calibration of a counting system. This method, however, had a number of disadvantages. It was often not possible to obtain a physical source that would represent the actual counting geometry. In addition sources can be expensive to obtain, maintain and remove from service. While it is often impractical to emulate the actual measurement geometry with a calibration source, mathematical modeling can be a reasonable alternative. Mathematically modeled geometries can be fitted to a particular measurement scenario and the geometry parameters can be rapidly adjusted if necessary to optimize performance.

The quality of mathematical efficiency calibrations depends on the accuracy of the modeled measurement geometry. This modeled geometry includes a source model, a detector model, and perhaps a collimator model. The detector model is used with MCNP and validated by performing a set of measurements with calibration sources over the full range of energies of interest to create a detector characterization file. The characterization file is used with Canberra's In Situ Object Counting System (ISOCS) [1 - 3] and a wide set of sample shape templates for efficiency calibration. The ISOCS software [4] can quickly calculate the gamma-ray efficiency for a particular geometry based on input parameters (e.g. sample dimensions, densities, etc.) provided by the user. Given accurate geometry dimensions, efficiency calibrations can then produce results with uncertainties ranging from 4% at energies above 300 keV to 15% at 10 keV. In some cases, however, parameters of the counting geometry may not be well known, thus increasing the uncertainty. This can be the case when the unknown source is located inside a container, or source matrix and compositions are not precisely known. This type of conditions is often encountered during safeguards measurements of special nuclear materials, when counting radioactive waste or performing measurements following a radioactive accident. In such cases, an Adaptive Mathematical Efficiency Calibration [5, 6] implemented in the Advanced ISOCS software, which is an extension of the ISOCS method, that can use additional data obtained directly from the measured spectrum can provide a more accurate physical model of the counting geometry, and, as a result, improves the efficiency calibration.

ADVANCED ISOCS DESCRIPTION

The Advanced ISOCS program is used to optimize geometry parameters that describe an item containing radioactive material. These parameters are then used in ISOCS to generate the absolute efficiency curve for a given counting geometry. The software uses spectral data, e.g., line activities for the isotopes present in the spectrum, or information obtained by some other means, for example, isotopic analysis in case of uranium or plutonium items, to determine the best set of geometry parameters.

The geometry optimization process consists of several steps. First, one or more gamma ray spectra of a given item must be acquired and the standard ISOCS efficiency calibration performed using a reasonable initial set of geometry parameters. The nuclide activity results obtained using this initial efficiency curve would often be used as a final answer. In some cases, an expert user could review data, adjust the geometry parameters, and, if necessary, regenerate the efficiency curve in order to improve the final result. This approach can often be impractical and time consuming and also strongly relies on the user expertise, which may not always be available depending on a particular situation. Under such circumstances, an automated geometry optimization with minimal user input and decision making can help to improve the measurement results.

Several optimization benchmarks are available in Advanced ISOCS including the following:

- MGAU/MGA: based on the relative efficiency data provided by MGA and MGAU isotopic codes
- FRAM: based on the relative efficiency data provided by FRAM isotopic code
- Line Activity Consistency Evaluator (LACE): based on consistency of the line activities for multiple energy nuclides
- User Defined Isotopics: based on the known enrichment information provided by the user for uranium items
- U or Pu Mass: based on consistency between the modeled and measured U or Pu mass
- Multiple Count: based on consistency between multiple counts of the same item from different location

The benchmarks produce Figures of Merit (FOM), and these two terms are used interchangeably in this paper. Each of the benchmarks can be used either individually or in combination with any other FOM(s).

Optimization can be performed either by using the Best Random Fit or Smart methods. In the first case, a large number of random geometries within the range of uncertainty is automatically created by the software and evaluated against the selected benchmark(s). Geometries that best satisfy the optimization criteria are then used to generate the optimized efficiency curve. In case of the Smart method geometries are not randomly generated, but rather iteratively defined each time using results from the previous optimization step, thus reducing the overall number of generated models and shortening the optimization time.

During the optimization the most critical geometry parameters affecting the efficiency curve, such as density, heavy elements weight%, fill height, etc..., are varied. When using the Best Random Fit several best ISOCS efficiencies are found that provide results consistent with the benchmark. The optimized average efficiency is then calculated based on these best ISOCS models. The five best models were arbitrarily chosen in this study for use during every optimization run. This number proved to provide reasonable results, which are described in this paper. The number of best models may be refined by the user, as more experience will be gained from using this software on real counting scenarios.

During the optimization a relative standard deviation (%RSD) of the absolute efficiencies for the current set of best ISOCS models is determined and used as a convergence criterion. Once the user defined limit for the %RSD is reached the optimization stops and the average efficiency is calculated. In this study the target %RSD was set to 5% in all cases when the Best Random Fit method was used.

In case of the Smart methods, where the optimization process only yields a single best efficiency curve a different convergence criterion was implemented.

OPTIMIZATION BENCHMARKS

Several optimization benchmarks are implemented in Advanced ISOCS. Each benchmark can either be used alone or in combination with any other benchmark during the optimization. For example, when measuring uranium items

it is possible to find an optimum solution (geometry) that would satisfy U Mass, MGAU, and FRAM benchmarks, all at the same time.

Isotopic Codes benchmark

Three isotopic codes – FRAM [7], MGA [8], and MGAU [9] were considered to be used for the optimization. These codes use similar methodology to determine the isotopic abundances ratios, which includes the generation of the relative efficiency curve. The shape of the relative efficiency curve generated by isotopic codes is largely dependent on the counting geometry and therefore was chosen to be used to optimize ISOCS geometry. During the optimization various methods can be used to find a geometry whose absolute ISOCS efficiency curve would have a shape that is most consistent with the relative efficiency curve produced by isotopic codes.

User Defined Isotopics benchmark

This is an independent benchmark method, which is quite similar to any of the isotopics methods. It is currently implemented for uranium analysis only. The idea behind this method is to build a relative efficiency curve based on a user entered uranium enrichment value. In this case the peak areas of uranium gamma lines are calculated for a measured spectrum using Genie2k gamma-spectroscopic software. The peak areas along with appropriate nuclear data are used in Advanced ISOCS to build a benchmark relative efficiency curve, which is then used during the optimization in a similar way as the MGAU or FRAM relative efficiency curves.

U or Pu Mass benchmark

This optimization method is based on the comparison of the measured uranium or plutonium mass and source mass modeled in ISOCS. In the ideal case the modeled U or Pu mass should correspond to the measured mass. The "measured" U or Pu mass is automatically obtained using measured peak areas, the gamma ray intensities and the ISOCS calculated peak efficiencies at uranium or plutonium gamma ray energies of interest. Activity is converted to mass by using the specific activities of the nuclides.

In case of uranium it is possible to calculate the ^{235}U and ^{238}U masses separately, as activity information is usually available for both nuclides. Masses of each individual isotope are added together to get a total uranium mass.

In case of plutonium this approach will not work, simply because it is not possible to calculate activities for each of the individual plutonium isotopes (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu), e.g., ^{242}Pu usually cannot be directly measured and its content is therefore determined by using a correlation formula. Thus, in order to determine total plutonium mass it is necessary to use isotopics information provided by isotopic codes, such as MGA or FRAM. Generally either ^{239}Pu or ^{238}Pu (for heat sources) activity in a sample can be easily estimated from the spectral data using corresponding peak areas. This nuclide activity is then converted into the nuclide mass, which may be used to calculate total plutonium mass if the weight % of ^{239}Pu or ^{238}Pu is known. The weight % of ^{239}Pu or ^{238}Pu (heat source) obtained using MGA or FRAM generally have the smallest uncertainties, and therefore these plutonium isotopes were chosen to calculate total plutonium mass.

LACE benchmark

This optimization benchmark is based on selecting the optimum geometry parameters, which, if used in ISOCS to perform an efficiency calibration, would result in a consistent activity for all gamma lines from a given nuclide.

In order for the LACE analysis to be performed the nuclide of interest has to have at least two gamma lines. To enhance the range and power of the LACE analysis, it is recommended to use nuclides whose individual gamma lines cover a wide energy range. An appropriate nuclide can be selected after reviewing the Nuclide Identification Analysis results for a given spectrum.

Similar to the optimization methods utilizing isotopic codes, this method is also based on the efficiency shape optimization. As a result this method is generally insensitive to the changes in the source to detector distance.

Multiple Counts benchmark

The Advanced ISOCS supports optimization based on multiple counts. By performing multiple measurements of an item from different perspectives, a better optimization of the efficiencies may be possible, especially on "thick" or "near-infinite" samples where the efficiency shape is not changing very much.

During the optimization, the weighted average activity measured for a selected nuclide(s) is compared for each individual count. The multi-count optimization is implemented by requiring that the weighted average activity of all counts of the item should be as close as possible to each other.

Although several measurements may be required to either pinpoint an exact source location or define the source shape, even two measurements of the same item taken from different perspectives help narrow down the solution space where the optimum geometry parameters can be found.

OPTIMIZATION METHODS

Several optimization methods were implemented in Advanced ISOCS, including a robust, but time consuming Best Random Fit method, as well as several smart methods utilizing various techniques that are used to guide the optimization process.

Best Random Fit Method

The Best Random Fit method is used to randomly generate a large number of ISOCS models. These models are automatically created by randomly picking the geometry parameters from within the predefined bounds specified by the user. The user has the ability in the software to specify a certain range for any geometry parameter used in ISOCS template to generate the absolute efficiency curve, e.g., that the container wall thickness varies from *X* mm to *Y* mm, source matrix density varies from *A* g/cc to *B* g/cc, fill height varies from *C* cm to *D* cm, etc… Often these bounds are not only reasonable estimates of the likely value ranges, but may also represent physical limitations, such as preventing negative densities, or exceeding the container dimensions/weight limit. The random models are used to generate the corresponding efficiency curves, which are then tested against the selected benchmarks. Either a single or several best models are chosen and used to generate the optimized efficiency curve.

Smart Methods

Although a FOM may indicate the quality of a given model, it does not directly indicate how to change the free model parameters to optimize the ISOCS geometry. A variety of automated numerical optimization techniques are possible. Several candidate optimization methods are available for this particular application [10] and five of them were considered for the optimization process:

- Particle Swarm Method
- Quasi Newton Method
- Sequential Method
- Simplex Method
- Marquardt Method

EXPERIMENTAL AND DISCUSSION

Multiple measurements were used to validate the optimization routines. These included measurements with uranium and plutonium items, as well as measurements with mixed gamma sources under various counting conditions. Although the Smart methods were also investigated, in this paper we only present results obtained with several sets of uranium standards using the Best Random Fit optimization method. We are still in the process of evaluating results obtained using the Smart methods, and therefore they will be presented elsewhere.

For each optimization case presented in this paper the total uranium mass was obtained using MGAU + U Mass, FRAM + U Mass, LACE + U Mass, User Defined Isotopics + U Mass FOMs, and ALL FOMs applied together.

Uranyl Nitrate solution standards

The measurements in this case were done with high density polyethylene (HDPE) bottles of 58 mm internal diameter and 100 mm in height. Each bottle was filled with a U+HNO_3 (3-Molar) solution with various concentration and filled heights. The bottles were placed 7 cm from the detector with the detector end-cap axis looking at the mid point of the bottles. The measurements were performed at the IAEA and the spectra were made available to Canberra. The optimization results for seven such samples (Uranium 1, Uranium 2, Uranium 3,

Uranium 4, Uranium 5, Uranium 6, and Uranium 7), with total uranium mass ranging from 18 to 30 grams are presented in this study.

The following geometry parameters were varied during the optimization process:

- Fill height: 20 to 100 mm
- Uranium wt%: wide range of values specific to each particular case (e.g., 1 to 10wt% for Uranium 1 and 5.5 to 30wt% for Uranium 7)
- Solution density: 0.1 to 3 g/cc

The optimization results showed that for most of the Uranyl Nitrate measurements the expected mass falls within 1σ to 2σ from the optimized value (see Fig.1 below).

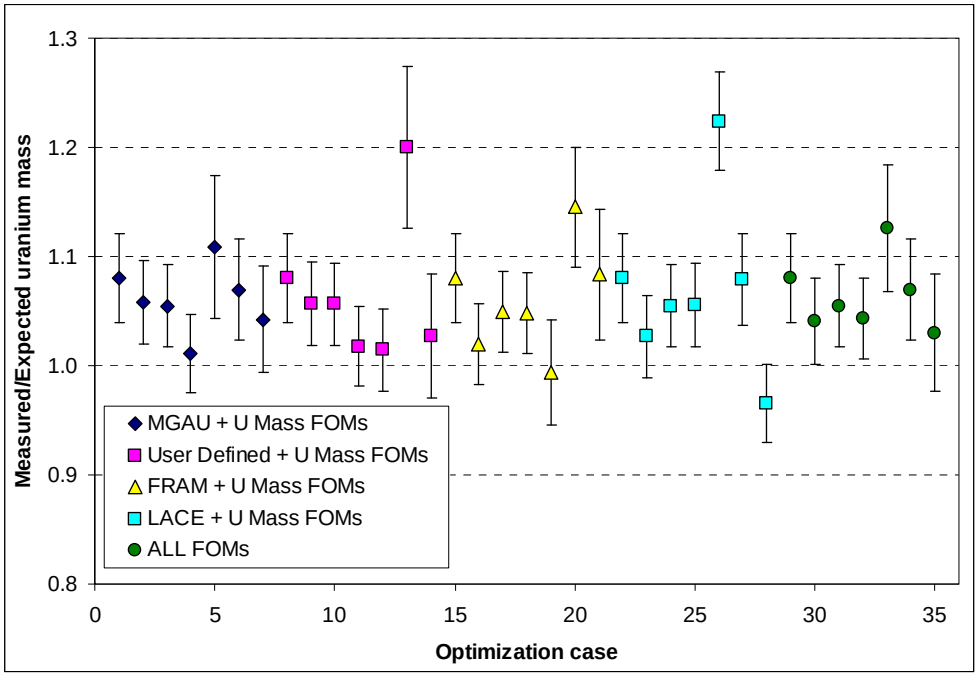

Fig. 1 Measured to Expected uranium mass ratio shown for Uranyl Nitrate solutions obtained using different optimization benchmarks. The dots on the plot are representing results obtained for each of the seven counting scenarios using different FOM combinations, i.e. first dot in each subset of datapoints corresponds to Uranium 1 data, second dot corresponds to Uranium 2 results, etc… Error bars shown represent a 1 sigma uncertainty.

Overall all benchmark methods performed well with most of the results being within 1 – 2 sigma from the expected uranium mass. The optimization time varied from 10 – 15 minutes in some cases when the convergence was quickly reached, to about 2 hrs total time when the target %RSD of 5% could not be reached even after all 2000 models have been run.

A slight positive bias (~ 6% on average) was observed in the optimization results for all cases. The reason for that is yet to be understood. Initial investigation has shown however that a similar bias is observed even when the best known geometry parameters, which were measured at the time of measurement and sample preparation, are used for ISOCS efficiency calibration (see Fig. 2 below). Possible reasons for this bias include discrepancy in the measured geometry parameters (e.g., source-to-detector distance, uranium mass/concentration), background fluctuations (although a background subtraction was performed in all cases, it should be noted that the background measurement was done a few days later than the actual measurements), bias due to uncertainty in ISOCS characterization for the particular detector used in this measurement campaign.

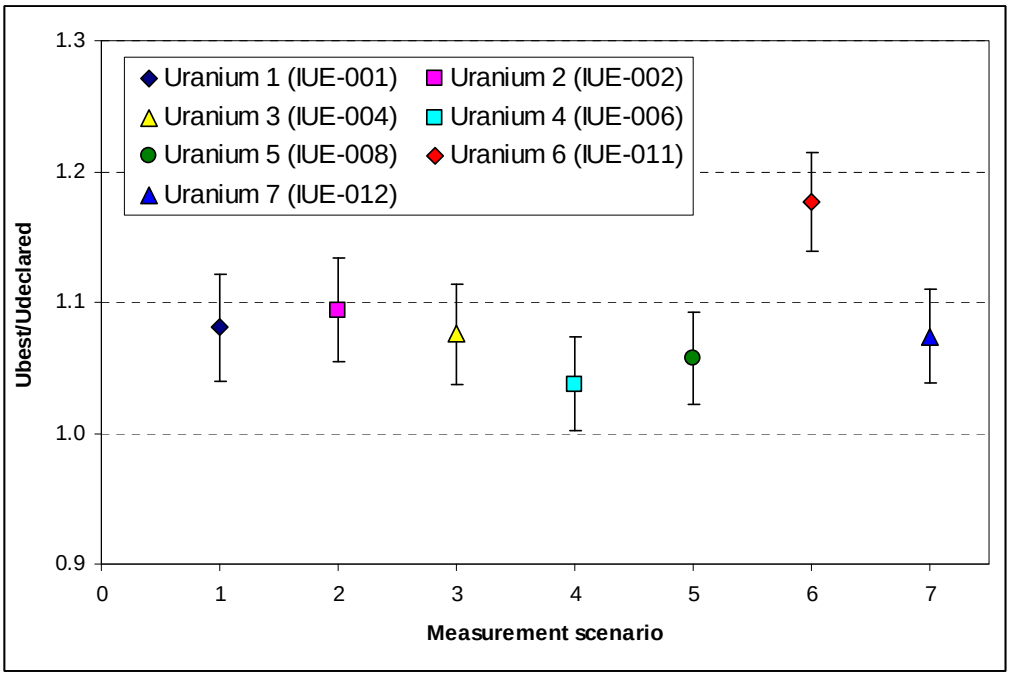

Fig. 2 Measured to Declared uranium mass ratio shown for Uranyl Nitrate solutions obtained using the best known geometry parameters during ISOCS efficiency calibration.

It should be noted that although this counting geometry was quite well defined, the variable geometry parameters, such as fill height, uranium wt%, and solution density, were chosen to cover a significantly wide range during optimization. As it has been mentioned above, in some cases using a wide range of geometry parameters resulted in the targeted %RSD of 5% not being reached with the maximum of 2000 models. This is an expected behavior of the code, as a large number of variations in random models require more models to be run overall to reach the same target %RSD, compared to the case when the geometry parameters are varied only in a narrow range.

When the geometry parameters are not well known, optimization can be performed in several steps, starting from a wide range of variation and then progressively narrowing down the range. In the first step a wide range is used for geometry all geometry parameters, as well as a large target %RSD. Each of the geometry parameters can then be observed for the best models, to find whether they all come from a certain part of the initial wide range. Optimization can then be repeated using a narrower ranges for the variable geometry parameters and also smaller target %RSD value. These steps can be automated in order to accelerate and ease the optimization process.

CBNM and SU-135 uranium standards

This series of measurements with uranium standards was done at the IAEA and the spectra were provided to Canberra. Some of measurements were performed at the Canberra's facility. Two types of uranium standards were used for these measurements – CBNM and SU.

Each CBNM standard contains about 200 g of U_3O_8 of a given enrichment encased in aluminum can. Measurements were performed with several such standards. The outer can diameter is 80 mm and the can height is 89 mm. The base has a thickness of 2 mm and serves as a lightly attenuating window for emitted gamma-radiation. The internal diameter of the can is 70 mm, and the fill height for the U_3O_8 powder is ~21 mm for all samples except for the 4.46 wt% enriched standard for which the height is ~16 mm. With these dimensions an approximate U_3O_8 density is 2.6 g/cc. Uranium standards of two different enrichments 2.95wt% and 4.46wt% were used in this study.

The SU type uranium standard consists of an aluminum can filled with about 1000 g of U_3O_8. The internal diameter of the can is 149 mm with a side wall thickness of 12.5 mm. The base of the aluminum can is only 2 mm thick. Uranium enrichment of SU standard is 3.105wt%

Each standard was measured at a number of different counting geometries, which included both measurements from the side and from the bottom, as well as measurements performed with additional absorbers. These counting

scenarios were named as Uranium 8, Uranium 9, Uranium 10, Uranium 11, Uranium 12, Uranium 13, and Uranium 14.

Although the chemical composition of the matrix was assumed to be known (U_3O_8, i.e. ~ 85wt% of U), the uranium weight % was treated as an unknown and varied within a wide range during each optimization run.

The following parameters were assumed to be not well known, and varied during the optimization:

- Fill height: 1 to 70 mm
- Uranium wt%: varied from 70.5 to 95 wt%
- Density: 0.5 to 3.5 g/cc

Not considering optimization results for Uranium 13 (point number six in each subset of datapoints presented in Fig.3) for the reasons given below, most of the measurements with uranium standards (CBNM and SU) yielded acceptable results, i.e. most of the optimized results were within 10 – 15% from the expected value (see Fig. 3 below). The optimization time varied from 17 minutes for Uranium 14 and up to about 9 hours in case of Uranium 13 using the Best Random Fit method.

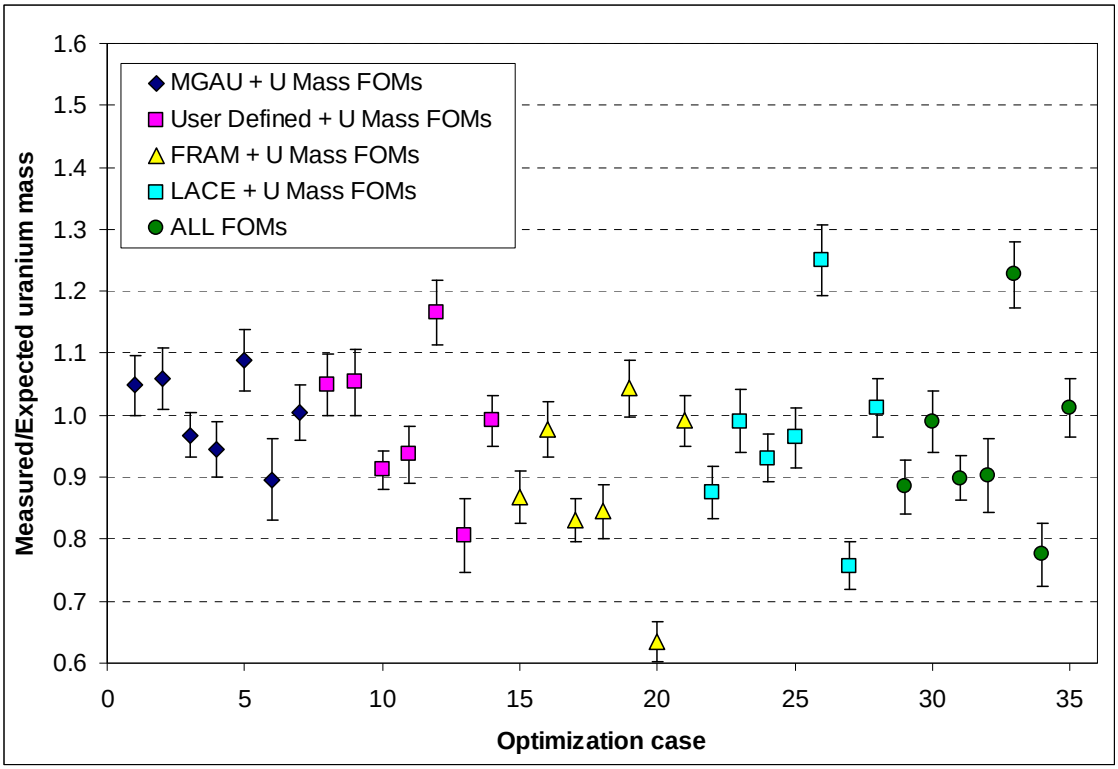

Fig. 3 Measured to Expected uranium mass ratio shown for Uranium Standards obtained using different optimization benchmarks. The dots on the plot are representing results obtained for each of the seven counting scenarios using different FOM combinations, i.e. first dot in each subset of datapoints corresponds to Uranium 8 data, second dot corresponds to Uranium 9 results, etc… Error bars shown represent a 1 sigma uncertainty.

In three cases (Uranium 8, Uranium 11, and Uranium 13) the enrichment results obtained with FRAM were significantly lower (by 10 – 20%) the declared values, while MGAU produced acceptable isotopics results. The 258 keV line in these spectra has a relatively low counting statistics (about 10% uncertainty), however, it is a key gamma line for the parameter set used by FRAM in this study. This line plays an important role when binding the low- and the high-energy regions of the relative efficiency curve and therefore has major effect on the enrichment results. MGAU, on the other hand, is not using the 258 keV line during the enrichment analysis, thus often producing better results when the counting statistics is low.

Uranium mass for the SU type standard measured from the side (Uranium 13) deviates by about 20% from the expected value. This could be related to the fact that the source material of the SU type standard is not tightly packed and may have shifted inside the container during the measurement making the counting geometry very uncertain. This did not seem to affect the optimization results for the measurement with the same uranium standard but performed from the bottom (Uranium 14), due to the uranium source being "infinitely" thick for uranium gamma-radiation.

It should be noted that for cylindrical items measured from the side optimization required significantly more time compared to the same item measured from the bottom, e.g., compare Uranium 8 measured from the side and Uranium 9 measured from the bottom (4 hours and 1 hours respectively), or Uranium 13 measured from the side and Uranium 14 measured from the bottom (9 hours and 17 minutes respectively). In all cases the same variable geometry parameters, including the fill height, were used, which covered the same wide range. However, since the uranium standards used in this study can be considered "infinitely" thick for uranium gamma lines, the measurements taken from the side are more sensitive to the change in the fill height. As a result, these geometries have ended up with significantly varied efficiency curves for the random models, and, therefore, a larger number of models had to be run in order to reach the target %RSD.

Multiple Count

Additional measurements were performed with uranium standards measured from two different angles. The first set of measurements was performed using a single 4.46wt% enriched CBNM standard containing about 200 g of uranium oxide encased in a cylindrical aluminum can. Two measurements were done from the side and the bottom as shown in Fig. 4 below. A similar set of measurements was also done with a larger 3.105wt% enriched uranium standard (SU-136-5), containing 1000 g of U_3O_8.

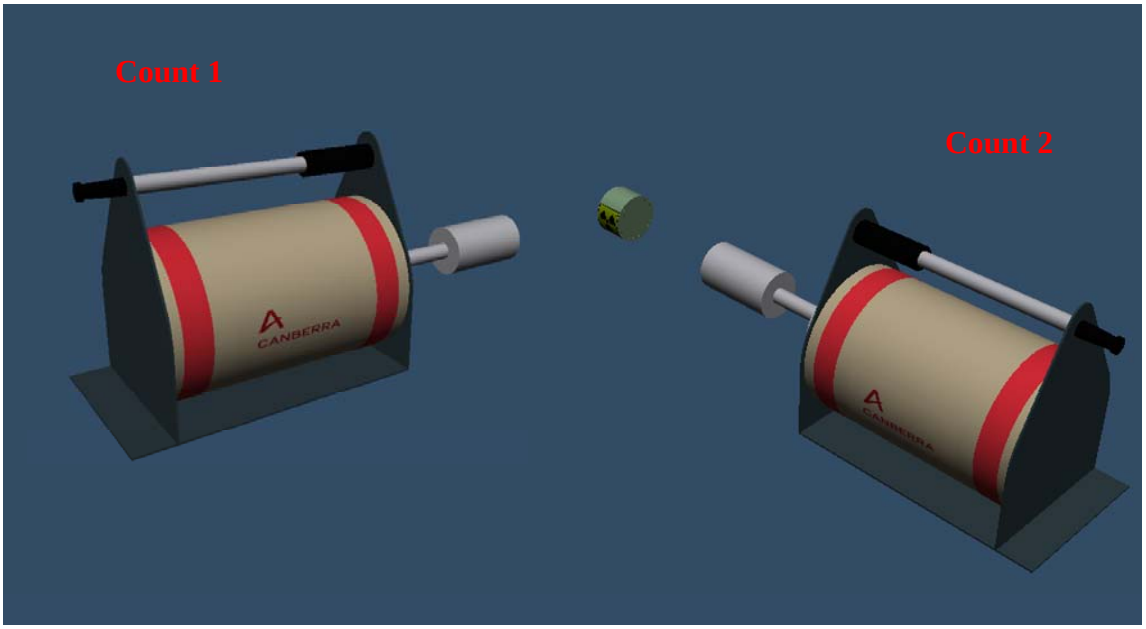

Fig. 4 3-D visualization of the multiple count geometry with CBNM standard

For each set of measurements two counts were initially treated independently, i.e. a total uranium mass was obtained for each count individually using MGAU + U Mass, FRAM + U Mass, LACE + U Mass, User Defined Isotopics + U Mass FOMs, and ALL FOMs one at a time. Then both counts were used simultaneously during the optimization, i.e. with the "Multiple Count" FOM added to each of the individual FOM combinations used in a single count mode. The purpose of the test was to investigate whether the results with individual counts can be improved, by using data from multiple measurements of the same item simultaneously during optimization. In addition, the Multiple Count FOM was also used alone as an independent optimization method.

The following parameters were assumed to be not well known during the optimization:

- Fill height: 1 to 70 mm (for CBNM standard) or 80 mm (for SU standard)

- Uranium wt%: varied from 70.5 to 95 wt%
- Density: 0.5 to 3.5 g/cc
- Aluminum container thickness also varied from 2 to 5 mm for CBNM and 1 to 15 mm for SU standard

Overall, the optimization based on multiple counts improved the results when compared to the data obtained for single measurements (Fig. 5).

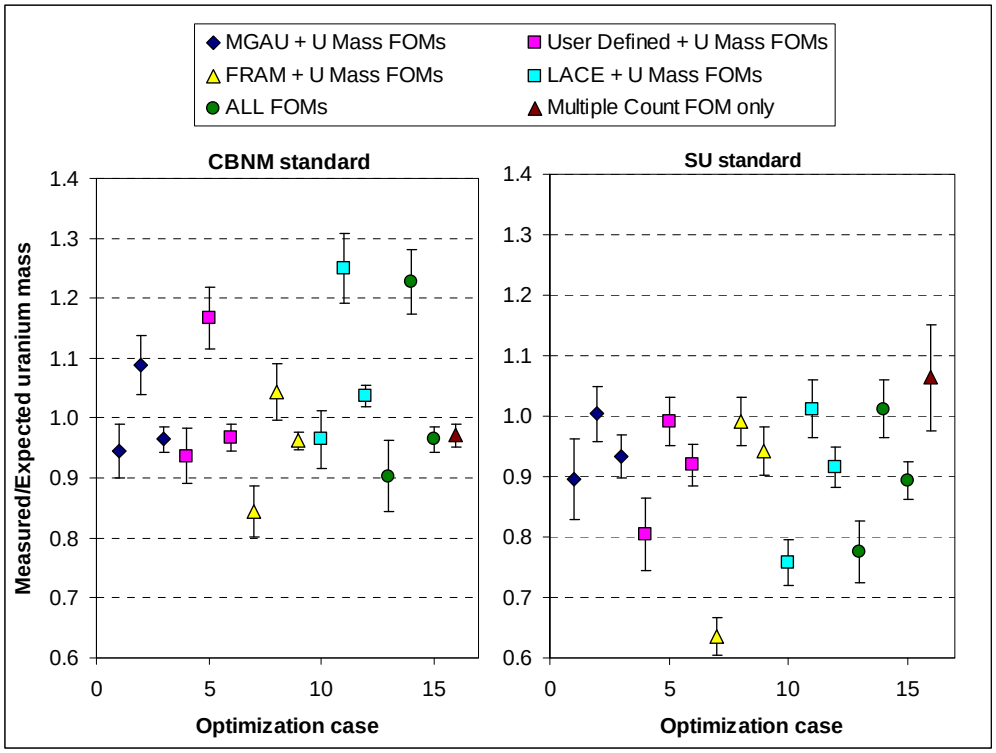

Fig. 5 Measured to Expected uranium mass ratio shown for uranium standards obtained using different optimization benchmarks. For each subset of datapoints obtained using different FOM combinations, the first two dots correspond to two single counts (measurements from side and bottom respectively); while the third datapoint represents results obtained when both single counts were used simultaneously during optimization. Results for the Multicount also include data obtained using the Multiple Count FOM only (last datapoint on each graph). Error bars shown represent a 1 sigma uncertainty.

In case of the SU standard the multi count results have a slight negative bias (~ 6-8%) compared to the expected value. This may be due to the reasons already described, i.e. due to the uranium material not being tightly packed inside a container. This seems to result in a negative bias in the total optimized uranium mass.

Another source of bias for both sets of measurements, and especially in case of the SU standard, could come from a limitation in the “Complex Cylinder” ISOCS template that was used to model these particular counting geometries. This template requires a container wall to be equally thick from all sides (side, top and bottom). While this may be a good assumption for some general type storage containers, in case of uranium standards the bottom thickness of an item is typically made smaller compared to the side wall, e.g., 5 mm wall vs 2 mm bottom for CBNM or 12 mm wall vs 2 mm bottom for SU standard. The container wall thickness in both cases was set to be varied within some interval (2 to 5 mm – CBNM, 1 to 15 mm – SU standard). This condition along with the ISOCS template requirement regarding the side wall thickness being be the same as the bottom thickness may have introduced an additional bias in the results.

In a realistic situation the natural choice when using two independent measurements would be to take an average result for both counts. It should be noted in this respect, that although the multi count results are biased low, they are still better compared to the average result for Count 1 and Count 2.

In some cases, e.g., when counting a single-line nuclide such as ^{137}Cs, the Multiple Count FOM may be the only method available for the optimization. Therefore, an additional test has been performed by using only the Multiple Count FOM during the optimization. As the results of this test show, this type of the optimization by itself demonstrated quite a good performance in both cases providing an excellent agreement between the optimized and expected uranium masses.

CONCLUSION

Mathematical efficiency calibration is a promising approach with applicability in many fields. The use of this calibration method can be improved by implementing an advanced efficiency optimization techniques, which utilize spectral information obtained directly from the measured spectrum. Development of the method and its implementation is currently underway. Preliminary data obtained using a new Advanced ISOCS application showed promising results proving the validity of the optimization approaches.

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