Waste Characterization Using Gamma Ray Spectrometry with Automated Efficiency Optimization – 13404

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ABSTRACT

Gamma ray spectrometry using High Purity Germanium (HPGe) detectors is commonly employed in assaying radioactive waste streams from a variety of sources: nuclear power plants, Department of Energy (DOE) laboratories, medical facilities, decontamination and decommissioning activities etc. The radioactive material is typically packaged in boxes or drums (for e.g. B-25 boxes or 208 liter drums) and assayed to identify and quantify radionuclides. Depending on the origin of the waste stream, the radionuclides could be special nuclear materials (SNM), fission products, or activation products. Efficiency calibration of the measurement geometry is a critical step in the achieving accurate quantification of radionuclide content. Due to the large size of the waste items, it is impractical and expensive to manufacture gamma ray standard sources for performing a measurement based calibration. For well over a decade, mathematical efficiency methods such as those in Canberra's In Situ Object Counting System (ISOCS) have been successfully employed in the efficiency calibration of gamma based waste assay systems. In the traditional ISOCS based calibrations, the user provides input data such as the dimensions of the waste item, the average density and fill height of the matrix, and matrix composition. As in measurement based calibrations, the user typically defines a homogeneous matrix with a uniform distribution of radioactivity. Actual waste containers can be quite non-uniform, however. Such simplifying assumptions in the efficiency calibration could lead to a large Total Measurement Uncertainty (TMU), thus limiting the amount of waste that can be disposed of as intermediate or low activity level waste. To improve the accuracy of radionuclide quantification, and reduce the TMU, Canberra has developed the capability to optimize the efficiency calibration using the ISOCS method. The optimization is based on benchmarking the efficiency shape and magnitude to the data available in the analyzed gamma ray spectra. Data from measurements of a given item in multiple counting geometries are among the powerful benchmarks that could be used in the optimization. Also, while assaying a waste stream with fission products and activation products emitting gamma lines of multiple energies, optimizing the efficiency on the basis of line activity consistency is very effective. In the present paper, the ISOCS- based optimization methodology is applied to measurement scenarios involving multiple counting geometries, and multi-gamma-line radionuclides. Results will be presented along with accuracy and precision estimates for each measurement.

INTRODUCTION

Mathematical methods such as ISOCS [1,2] are being increasingly employed for determining gamma ray efficiencies in radioactive waste assay applications, and in D&D related activities. The ISOCS method uses the intrinsic response characterization grid for the specific gamma ray detector, and a ray tracing code for determining photon attenuation through absorbers internal and external to the source geometry. The response grid spans a radius of up to 500 meters, and an energy range of 10 keV to 7 MeV. The ISOCS software enables the user to model the measurement geometry including the source, collimator, shielding, and any intervening absorber material. The ISOCS method also includes the capability to estimate uncertainties in efficiencies due to not-well -known input geometry parameters. For uncertainty estimation, the user indicates the variable parameters in the ISOCS Uncertainty Estimator (IUE) utility in the ISOCS software [3], the range of variation of each variable parameter, and a probability distribution function for selecting the input parameter value. The IUE then generates a number of input geometry models, computes efficiencies at each of the gamma ray energies indicated by the user, and then determines the average and standard deviation of efficiency values. A recent innovation in ISOCS enables the optimization of efficiencies in applications where very limited information is available regarding the radioactive source item that is to be assayed. The optimization methodology uses the data available from the measured gamma ray spectra, and automatically determines the best geometry model that yields results consistent with the measurements.

BENCHMARKS AND ROUTINES USED FOR OPTIMIZATION

When performing waste assay, a good strategy is to measure a given radioactive item in various source to detector configurations; for example, pointing the detector at different sides of the item, at different angles etc., and determining the source model that yields consistent results for all measurement geometries. Another useful strategy is to take advantage of data that may be available from multiple gamma energy lines emitted by the same nuclide. Many of the activation products and fission products do emit multiple gamma energy lines, and the efficiency optimization can therefore be performed by line activity consistency evaluation (LACE). Figures of Merit (FOM) were defined for the Multiple Count and the LACE benchmarks. Measurements were conducted to validate the performance of the efficiency optimization using Multiple Count and LACE FOM.

Two different optimization routines were used in the current validation campaign; the Best Random Fit (BRF), and a Smart method. In the Best Random Fit method, a large number of random models are created for the measurement geometry and evaluated against the selected benchmark(s). Models that best satisfy the optimization criteria are then used to generate the optimized efficiency curve. In case of the Smart method, models 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.

The smart method showcased in the present work is the Downhill Simplex [4]. It involves continuously improving the FOMs of models represented by points in the solution space at the vertices of a multidimensional form, or simplex. An initial simplex is established with one vertex more than the number of free parameters, and all of these point models are evaluated. The points are sequentially improved by simultaneously adjusting all of the free parameters in the point with the worst FOM. After the worst point is improved and is no longer the worst point, the new worst point is improved. Improvements are performed by reflecting, expanding or contracting the worst point through the centroid of the other points. If none of these three trials improves the worst point to better than the second worst point, all of the points are contracted halfway towards the point with the current best FOM. The Simplex method maintains all sampled points inside the parameter bounds by truncating any parameter values attempting to extend beyond the bounds. The vertices are initialized with one point at the center of each parameter range and the other points randomly located.

EXPERIMENTAL SETUP

Two sets of measurements were performed with standard 208 liter drums filled with different matrices. The first set of measurements was done with a drum filled with softboard matrix having an average density of 0.4 g/cm^3. For the second set of measurements a higher density sand matrix with a density of 1.65 g/cm^3 was used to fill a drum. During measurements a ^{152}Eu gamma source (Source #1) was randomly placed inside a drum and counted for several hours using a High Purity Germanium (HPGe) detector to ensure good counting statistics. The certified gamma source activity is given in Table I.

TABLE I. Certified Eu gamma source activity at the time of measurement

	Isotope	**Activity, μCi**	**Uncertainty, 1 sigma**
Source #1	Eu-152	3.95	+/- 0.05
Source #2	Eu-152	3.62	+/- 0.05

Each set of measurements consisted of two individual counts that were performed with the detector pointing at a drum from two opposite directions as shown in Fig. 1. The same set of measurement was repeated with two Eu-152 sources (Source #1 and Source #2) simultaneously placed inside a drum. This represented a special and more difficult case of a drum containing two hot spots.

The resulting spectra were analyzed individually as well as all together to evaluate the measured gamma-source activity and compare it to the expected value.

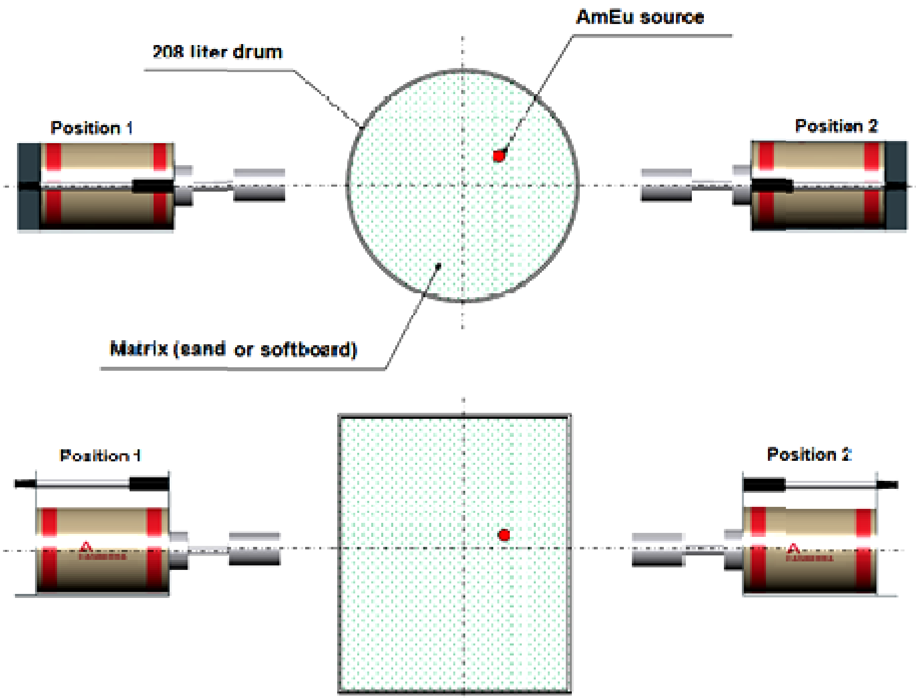

Fig. 1. Counting geometry (top and side view).

DATA ANALYSIS

Several methods were used to analyze the measured spectra. The most basic general approach, which is typically used during waste measurements, assumes that the source is uniformly distributed within a drum. The corresponding efficiency was generated using ISOCS and used to obtain Eu-152 activity from the measured spectra. Although this approach is routinely used in the field it often does not fully represent the actual counting geometry and therefore can produce a biased result.

In order to overcome the limitations of the uniform source distribution approach and utilize the information that is available from the measurement itself, the same data were analyzed using the optimized efficiency. The optimized efficiencies were obtained using the LACE and Multiple Count FOMs. The LACE FOM 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. During the optimization using the Multiple Count FOM, 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. These FOMs were used both individually and in combination with each other. In this particular study we used the Eu-152 activity for all benchmarks.

During the optimization it was assumed that the point source, representing one or two hotspots, could be located anywhere within a drum volume. The matrix density could also change within some pre-defined limits, and for the softboard drum it was assumed to vary between 0.3 and 0.5 g/cm^3, while for the sand drum it was set to vary between a 1.55 to 1.75 g/cm^3 range.

With the Best Random Fit optimization method 1000 random ISOCS models were created and their efficiencies calculated. These efficiencies were evaluated against the selected benchmarks, and then the top five models were used to generate the optimized efficiency. The total optimization time in case of the Best Random Fit optimization was on the order of 15 minutes. In case of the Simplex method the optimized efficiency was generally obtained with less than 100 iterations (ISOCS models), and therefore the overall optimization time was considerably shorter (about an order of magnitude lower) when compared to the Best Random Fit.

It should be noted that in case of two hotspots only the Best Random Fit method was used during the optimization. This is because the Simplex method requires a special handling when optimizing discrete values, such as the number of hotspots in a drum, and these feature was not yet implemented in the optimization routine.

RESULTS

Table II below presents the results obtained for the Eu-152 gamma source measured inside the softboard drum. The first rows in the table shows the measured activity obtained using a basic analysis approach, i.e. uniform source distribution. In this case a significant difference was observed between the measured activities obtained with each individual detector. Although the average activity of the two counts was only about 30% higher than expected value, the measured uncertainty was extremely high due a large scatter between the individual results.

The best result in the case of the softboard drum was obtained when a combination of the LACE and Multiple Count FOMs was used during the optimization. Both methods, Best Random Fit and Simplex, showed similar results with the optimized Eu-152 activity within just a few percent from the expected value. Note that even when individual FOMs were used separately in the optimization process, the final results were still considerably better than the ones obtained using a general calibration approach.

The data presented in Table II also shows that the Simplex optimization method works well for individual FOMs. For example, when this method was used to optimize the counting geometry based on the Multiple Count FOM, the resulting efficiencies, when used with the measured spectra, produced almost identical activity results (~4.36 μCi of Eu-152) for the measurements performed at Position 1 and Position 2.

Fig. 2 below shows a comparison between the LACE analysis results obtained for the uniform source distribution case and for the geometry optimized based on the LACE and Multiple Count

FOMs used together. In the ideal case, the LACE curve should represent a horizontal flat line, which indicates that the line activities for all individual gamma lines from the same nuclide are the same within the estimated uncertainty limits. From the LACE curves shown in Fig. 2, it is evident that the assumption that was used to generate the efficiency curves representing a uniform source distribution is not valid, while the optimized results obtained with the Best Random Fit and the Simplex methods show that the line activity data is consistent.

TABLE II. Measured Eu-152 activity in a softboard drum obtained with different efficiency models

Efficiency model	**Activity for Position 1, µCi**		**Activity for Position 2, µCi**		**Average activity +/- stdev, µCi**		**Measured/ Expected**
Uniform source distribution	9.20	+/- 0.17	1.22	+/- 0.03	5.21	+/- 5.64	1.320
BRF (LACE)	3.70	+/- 0.06	4.42	+/- 0.14	4.06	+/- 0.51	1.030
BRF (Multi)	3.88	+/- 0.13	3.78	+/- 0.13	3.83	+/- 0.07	0.970
BRF (Multi + LACE)	**3.73**	**+/- 0.04**	**4.19**	**+/- 0.04**	**3.96**	**+/- 0.33**	**1.003**
Simplex (LACE)	3.50	+/- 0.07	4.60	+/- 0.09	4.05	+/- 0.78	1.026
Simplex (Multi)	4.36	+/- 0.08	4.36	+/- 0.09	4.36	+/- 0.002	1.106
Simplex (Multi + LACE)	**4.09**	**+/- 0.08**	**4.04**	**+/- 0.08**	**4.06**	**+/- 0.03**	**1.029**

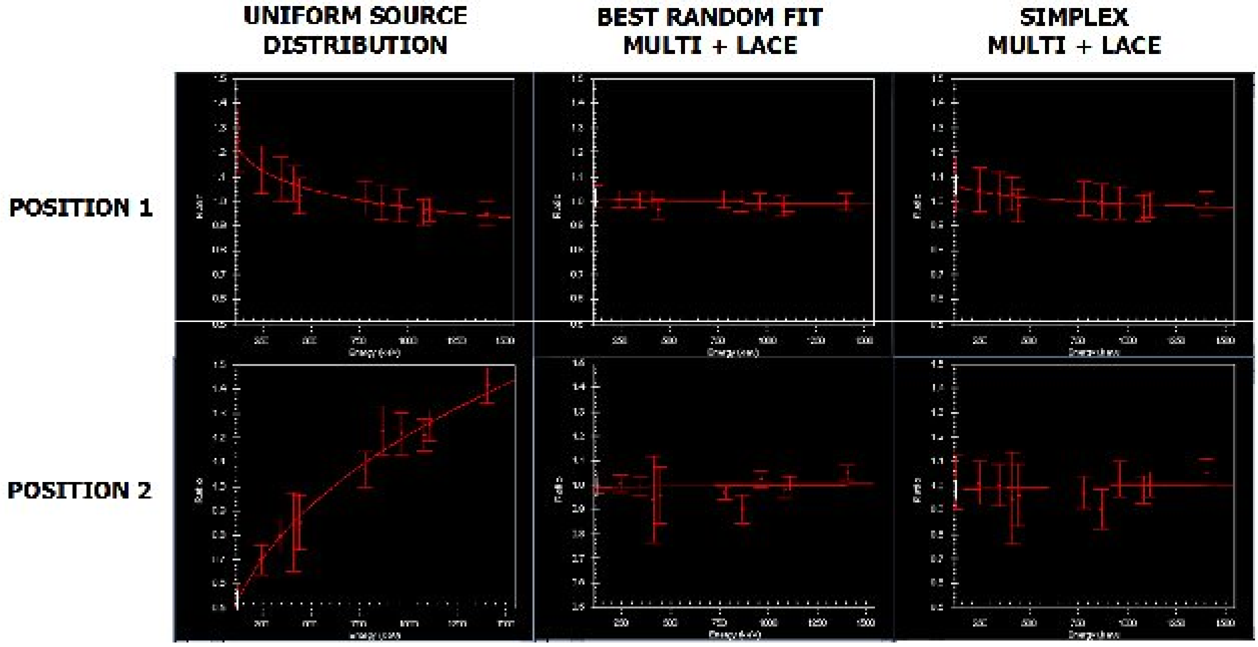

Fig. 2. Comparison of the LACE curves for the softboard drum.

As it is shown in Table III below, similar analysis results were observed in case of the sand drum. Once again, the best result was achieved when using a combination of LACE and Multi Count FOMs, although the optimized results using the individual FOMs still showed quite satisfactory performance for this highly attenuating geometry. Both, the Best Random Fit and Simplex optimization using a combination of the LACE and Multiple Count FOMs produced the Eu-152 activity result, which was within just a few percent from the expected value.

TABLE III. Measured Eu-152 activity in the sand drum obtained with different efficiency models

Efficiency model	**Activity for Position 1, µCi**		**Activity for Position 2, µCi**		**Average activity +/- stdev, µCi**		**Measured/ Expected**
Uniform source distribution	0.27	+/- 0.01	0.76	+/- 0.03	0.51	+/- 0.34	0.130
BRF (LACE)	2.68	+/- 0.06	5.29	+/- 0.23	3.99	+/- 1.85	1.011
BRF (Multi)	6.89	+/- 0.59	7.35	+/- 0.54	7.12	+/- 0.32	1.805
BRF (Multi + LACE)	**3.90**	**+/- 0.19**	**4.18**	**+/- 0.10**	**4.04**	**+/- 0.19**	**1.024**
Simplex (LACE)	2.93	+/- 0.08	3.19	+/- 0.07	3.06	+/- 0.18	0.776
Simplex (Multi)	5.49	+/- 0.15	5.52	+/- 0.13	5.51	+/- 0.02	1.396
Simplex (Multi + LACE)	**4.09**	**+/- 0.12**	**4.03**	**+/- 0.09**	**4.06**	**+/- 0.04**	**1.029**

Table IV below shows the results that were obtained with the Best Random Fit method for the two hot spots geometry. It can be seen from the data that the optimization process allowed a much better agreement between the expected and measured total activity for two hot spots, and also much lower measurement uncertainty when compared to the traditional uniform source distribution approach.

Overall, a significant improvement over the routine waste measurement approach was observed in the analysis result for all cases where the geometry optimization process was utilized. While the standard analysis methodology, assuming a uniform source distribution, produced results which were significantly different from the expected value and/or had large uncertainty, the analysis results that were additionally optimized based on the measured data obtained directly from the spectrum were generally much closer to the expected value. It was found that when several benchmarks are combined together during the optimization, the final result can be as close as a few percent to the expected activity value.

TABLE IV. Measured Eu-152 activity in the softboard drum obtained for two hot spots

Efficiency model	**Activity for Position 1, µCi**		**Activity for Position 2, µCi**		**Activity for Position 3, µCi**		**Activity for Position 4, µCi**		**Average activity +/- stdev, µCi**		**Measured/ Expected**
Uniform source distribution	4.43	+/- 0.12	3.62	+/- 0.09	7.55	+/- 0.15	9.25	+/- 0.22	6.21	+/- 2.64	0.821
BRF (LACE only)	8.70	+/- 0.49	8.63	+/- 0.35	6.52	+/- 0.15	8.27	+/- 0.54	8.03	+/- 1.02	1.060
BRF (Multi only)	8.07	+/- 0.80	7.70	+/- 0.60	7.70	+/- 0.51	7.52	+/- 0.53	7.75	+/- 0.23	1.023
BRF (Multi + LACE)	**9.30**	**+/- 0.37**	**7.83**	**+/- 0.60**	**6.19**	**+/- 0.54**	**8.34**	**+/- 0.49**	**7.92**	**+/- 1.30**	**1.045**

CONCLUSIONS

Optimization of gamma ray efficiencies based on the ISOCS is a powerful method to reduce bias and improve accuracy in the reported nuclide activity results. This method, which can be run on a personal computer, utilizes spectral data obtained directly from the measured spectrum in order to determine the optimal ISOCS geometry for the efficiency calibration. Two optimization approaches were investigated in this study. One of them requires generating a large number of efficiencies and then selecting the best ones that satisfy the benchmark criteria, i.e. Best Random Fit method. The other method uses a numerical routine, namely Downhill Simplex, and does a focused search to optimize based on the benchmark data. The results presented in this paper showed that both optimization methodologies offer a significant improvement in the accuracy of the activity determination for an unknown geometry.

FUTURE WORK

As part of the future work aimed at improving the optimization process we are now working on enhancing the Simplex method capabilities that would allow it to optimize geometries containing variable discrete parameters. This will significantly expand the area where this smart optimization routine can be used.

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