An Advanced ISOCS In Situ Gamma Spectrometry Services Tool to Reduce Uncertainties in Waste Characterization Projects - 16311

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ABSTRACT

CANBERRA's In Situ Object Counting System (ISOCS) is an established tool used for gamma spectrometry allowing a physical representation of complex geometries and mathematical calculation of the calibration function. It avoids the need for radioisotope standards which can be costly and prohibitively difficult to produce.

Uncertainties in the sample – detector geometry lead to uncertainties in the calibrations. CANBERRA's ISOCS Uncertainty Estimator (IUE) tool offers an automated means of accommodating known model geometry uncertainties, with calculation of robust defensible calibration uncertainties. However these uncertainties may be large leading to potential over-estimation of under-estimation of activity. This can be equated directly to waste disposal costs if one is using ISOCS for waste sentencing to classify wastes as (for example) Transuranic Waste (TRU) or Low Level Waste (LLW).

An Advanced ISOCS *in-situ* Gamma Spectrometry (AIGS) services tool has been developed to reduce the uncertainties. This system is based on generating and comparing geometry models that yield Figures of Merit (FoM) indicative of improved consistency between modelled data and available diverse measurement data.

This paper describes the development and features of AIGS, focusing on recent testing with radioisotope sources in a 200 litre drum filled with a simulated waste matrix. The results demonstrate a significant advantage for Am-241 where uncertainties can otherwise amount to a factor of 2 or more. We show how our specialist teams can develop procedures for deployment of standard ISOCS hardware, adapting measurement geometries and procedures through careful planning, to provide data that can be used to greatly reduce the uncertainties in the measured activities. Lastly, we comment on the range of potential applications for this new technique and the wider benefits for the decommissioning industry.

INTRODUCTION

Model-based physics methods are employed [1] to develop calibrations for Non-Destructive Assay measurements used to characterize materials contaminated with Plutonium, Uranium, and other radioactive isotopes. CANBERRA's In Situ Object Counting System (ISOCS) is an established tool that is used for field gamma spectrometry measurements allowing a mathematical representation of physical complex source – sample geometries and calculation of the detection efficiency function. This avoids the need to use representative radioisotope standards and this can be beneficial to users as such standards can be costly and difficult to produce in a way that ensures the activity is representative of activity profiles expected in reality. This tool can be used to develop calibrations both for plant-installed assay systems that may need to be reconfigured to measure different container types, and to develop special calibrations for a range of waste item types for field measurements. As such, it provides a powerful, flexible means of delivering waste characterisation projects offering time and cost savings for plant operators.

ISOCS offers a simple, powerful and flexible method of performing calibrations for field gamma spectrometry measurements where the sample geometries can be variable. However, sample – detector geometry (container size, dimensions, waste matrix type and homogeneity, activity spatial distribution and physical form, source – detector coupling) uncertainties can lead to high uncertainties in the calibrations and hence the determined activities. To allow ISOCS to produce robust uncertainty estimates in such cases, CANBERRA has extended the normal ISOCS software to include the ISOCS Uncertainty Estimator (IUE) tool and this is included in the standard release of the software. This offers an automated means of incorporating known model geometry uncertainties (parameters and population distributions for the possible values), and calculating their effects on the uncertainties in the efficiency function. This allows for the expressed uncertainty to follow prescribed formalism (distribution type, confidence interval), and hence provides robust defensible data. However, correct use of IUE can only model a measurement based on accurate known data for the source – sample geometry and the source distribution [2]. Therefore although the uncertainties will be reliable, they may be large depending on the geometry uncertainty especially if the geometry is not fully optimized for the specific application and sample type.

Recent trends in the nuclear industry towards planned minimization of waste disposal costs, in the light of budgetary constraints and limited repository capacity, is driving the improvement of accuracy for waste activity measurements when used for characterization or sentencing applications.

CANBERRA has developed an advanced ISOCS software tool specifically to improve the accuracy of measurements. Originally developed in response to safeguards needs for improved flexibility of modelling and adaptation to varied fissile material measurement scenarios, this tool is now being developed for waste assay applications in response to the industry driver for improved accuracy. This tool is known as the Advanced ISOCS *in-situ* Gamma Spectrometry (AIGS) services tool. Demonstration measurements have been carried out under factory conditions [3, 4] to show how Total Measurement Uncertainty (TMU) can be reduced for measurements of activity in 200 litre waste drums. In [5] it was shown how this can be used for real-world waste sentencing applications. CANBERRA now offers AIGS services through the specialist deployment of these tools. Recent efforts have been devoted to the demonstration of these techniques for a benchmark set of simulated waste drums. This work is designed to show how the TMU can be improved for 200 litre drum measurements based on realistic waste matrix densities and scenarios for heterogeneous activity distribution (unknown point sources within the drum compared to the normal default reference assumption of uniform activity throughout the drum) and to develop an operational work flow that can be used for real-world applications.

In this paper we describe the recent 200 litre drum measurement programme and discuss the results, showing how this represents an improvement compared to what is possible using traditional techniques. We comment on potential applications and how the AIGS tool can be used as an advanced planning tool, to optimize measurement programmes to provide beneficial TMU improvements.

The motivation for improving TMU can be to:

- Improve accuracy of waste stream characterisation
- Improve efficiency of segregation of wastes
- Reduce proportion of wastes pessimistically sentenced to higher category
- Optimise storage of containers in facilities
- Reduce waste storage costs
- Reduce waste disposal costs

Figure 1 illustrates the benefits of improved accuracy for waste sentencing applications. The waste drums labelled “savings” can be sentenced to the lower waste category by virtue of the lower TMU that is possible using the AIGS tool. When the waste storage and disposal costs can be much higher for the higher category, the potential benefits are large.

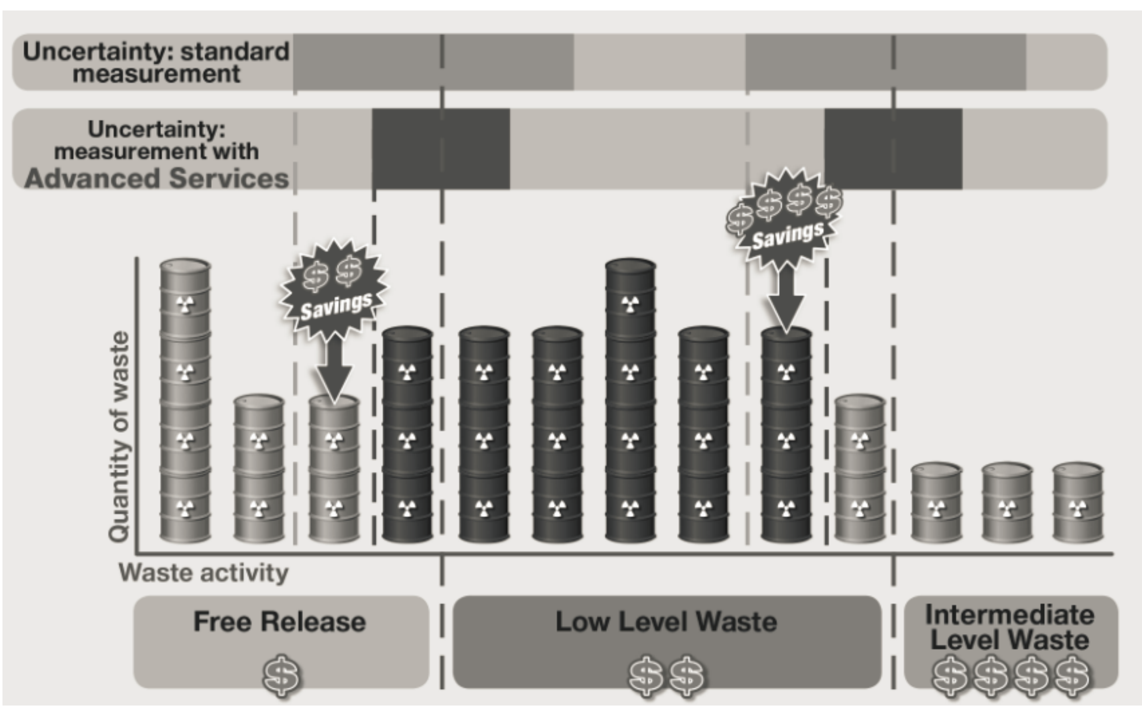

Figure 1. Benefits of improved TMU for lowering waste disposal costs.

Our studies for small low-density waste packages (typical of laundry bags) of TRU-bearing soft waste (densities of approximately 0.1 – 0.2 g.cm^{-3}) show that a response variation equivalent to approximately a factor of 2 can result from normal ISOCS "single-shot" measurements (based on Am-241 assay). Figure 2 shows typical data for a range of densities and 2 different types of matrix in a 200 litre drum. This shows how severe the effect can become, especially for high density, metallic matrices.

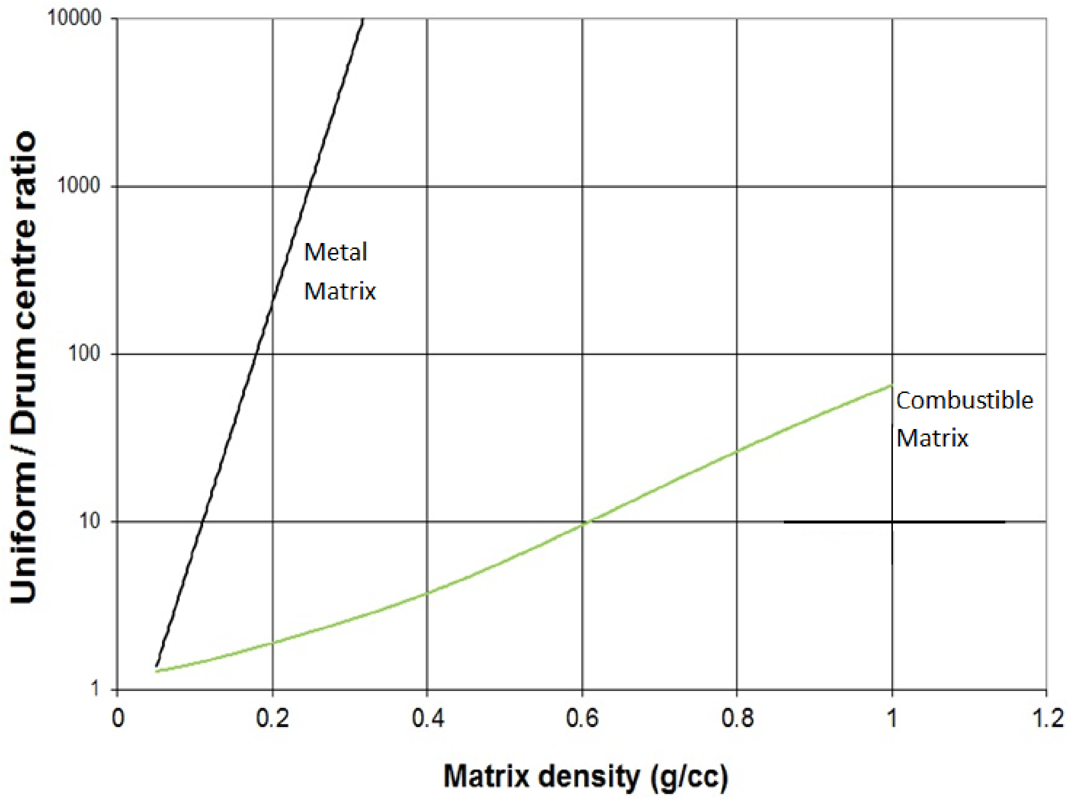

Figure 2. Potential uncertainty for TRU measurements (based on Am-241 assay) of a 200 litre drum with a single point source at unknown location.

These examples illustrate the motivation for reducing the TMU for a range of waste container types.

ADVANCED ISOCS IN-SITU GAMMA SPECTROMETRY SERVICES TOOL OVERVIEW

The philosophy is to utilize all of the available measurement data, and to adjust the model parameters that are subject to uncertainty in order to obtain a best-fit model which gives results most closely matching the measurement data. Several benchmark measurements can be utilized including the following [3]:

- MGA / MGAU or FRAM. The relative efficiency curve provided by these Pu & U isotopics analysis codes are used.

- User-defined isotopics. If the Pu or U isotopics are known then this can be provided as input data.
- U or Pu mass. A consistency evaluation is performed between measured and modelled Pu or U mass.
- Line Activity Consistency Evaluator (LACE). This method performs a comparison between the activity evaluated using different energy gamma lines from the same nuclide. The correct geometry model is indicated by the same activity obtained from each line.
- Multiple counts. This method performs a consistency check between the activity measured for the same item but for different item – detector geometries. By comparing the results of measurements of the same item from different orientations it is possible to correct for non-uniform source distributions.

To assess the measurements, figures of merit are used. The two approaches available are the best random fit (BRF) method and smart optimisation. The BRF technique consists of running a large number of models to fill the parameters space (spanning the uncertainties of the various parameters) and seeking the model that best matches the measurement data. This approach yields unambiguous optimized results but requires the most computing time. Smart methods consist of a focused search for the best-fit parameters. In developing the software, CANBERRA physicists considered and evaluated several alternative candidate algorithms including Simplex, Quasi-Newton, Particle Swarm, Sequential optimization and Marquardt optimization. The “down-hill” simplex technique was selected for detailed performance evaluation.

MEASUREMENTS AND ANALYSIS

Early measurements [2, 3] were conducted studying a single or two point sources of Eu-152 at various locations inside a 200 litre drum of homogeneous softboard (density 0.3 – 0.5 $g.cm^{-3}$, representative of typical low density waste) and of homogenous sand (density 1.55 – 1.75 $g.cm^{-3}$, representative of typical decommissioning bulk rubble waste). The case of a single point source represents the worst-case scenario because the range of efficiency is at its most extreme (if multiple hotspots can be assumed to be present then the averaged efficiency has a lower statistical spread than for a single source randomly distributed). Multi-count (4 detectors at different azimuthal locations around the drum) and LACE benchmark measurements were performed and Figures Of Merit were evaluated using both techniques separately and together. For a single point source, the use of a conventional “uniform activity” calibration resulted in measurement bias (measured / declared activity ratio) values of approximately 1.30 and 0.13 for softboard and sand matrices respectively. This indicates the potential under-estimation that can occur for the sand matrix. The use of the AIGS tool was found to produce measured / declared activity ratios within a few % of unity, indicating the benefits of the technique. In all cases the “corrected” activity was within 10% of the known value. The simplex technique was found to be useful giving similar results to the

BRF technique and in some cases the use of LACE and multi-count together was found to lead to small improvements. The results provided confidence that the technique could be successfully used for improving accuracy of ISOCS measurements of waste drums.

For the most recent measurements (reported in this paper), the following homogeneous matrix was studied, filling a 200 litre drum; the results are presented below.

- Mineral Wool - $\mathbf{0.111}\ \boldsymbol{g.cm^{-3}}$

Reference calibration line-sources of Ba-133, Cs-137, Eu-152 and Am-241 were studied.

Each source has a length approximately 1/3 of the drum height, representing a typical “package” of activity located at a position inside the drum. A complete set of measurements was conducted with each source located (in turn) at each of three heights (spanning the height of the drum) and 3 radial locations spanning from the centre to the edge of the drum, providing 9 source configurations for each matrix. The measurements provided a rich data set that could be used to evaluate the performance of the AIGS tool for point sources combinations at any position(s) and compare with a traditional “default” ISOCS approach assuming uniform activity. The drums used are reference Segmented Gamma Scanner (SGS) calibration drums and contain well-known inactive matrix materials. The available source positions are selected from the typical SGS helical tube positions. Measurements were conducted using a BEGE detector with 180 degree collimator (for maximum counting efficiency) and positioned horizontally at each of three different heights centred on three axial drum segments. The detector – drum surface offset distance was chosen (approximately 70 cm) as a typical distance for this type of drum measurement. In order to simulate the effect of multiple azimuthal detector measurements (necessary to obtain information on the radial distribution of activity within the drum), the drum was stepped on a turntable through 8 angles to provide effectively 8 different detector locations. For each source configuration (9 in total), there are 24 measurements comprising the 3 different detector heights and 8 detector angles. The measurement geometry is illustrated in Figures 3, 4 and 5. To simplify the analysis, the 360 degree rotation was represented using 4 of the available measurements (with 90 degree spacings) at each height. Multi-count optimization was deployed using these 4 x 3 = 12 measurements. Figure 5 (ISOCS model) depicts a typical measurement geometry. It is planned to conduct analysis later to investigate the value of deploying finer azimuthal segmentation (using all 8 data sets). It was decided to focus on three source positions, denoted 1a, 2c and 3b, as defined in Figure 6. These span the range of response throughout the drum. Position 1a corresponds to “top, centre”, position 2c corresponds to “mid-height, outer-radial” and position 3b corresponds to “bottom, mid-radial”.

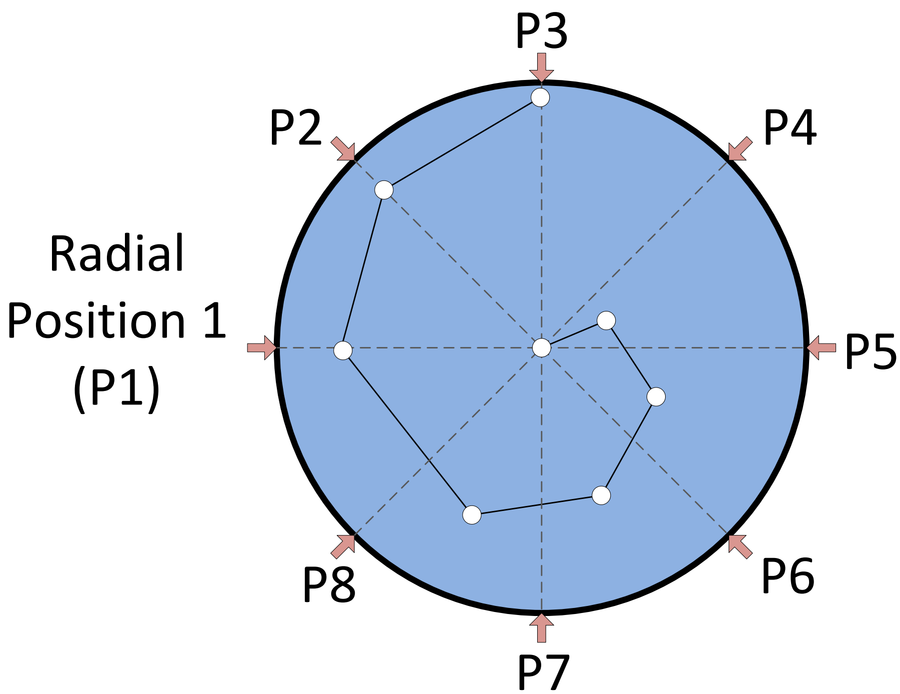

Figure 3. Test drum source positions (radial) and azimuthal detector positions. There are 8 different detector positions (P1 – P8) and 3 chosen radial source positions corresponding to the central 0th tube, 3rd and 7th tubes measured radially outwards.

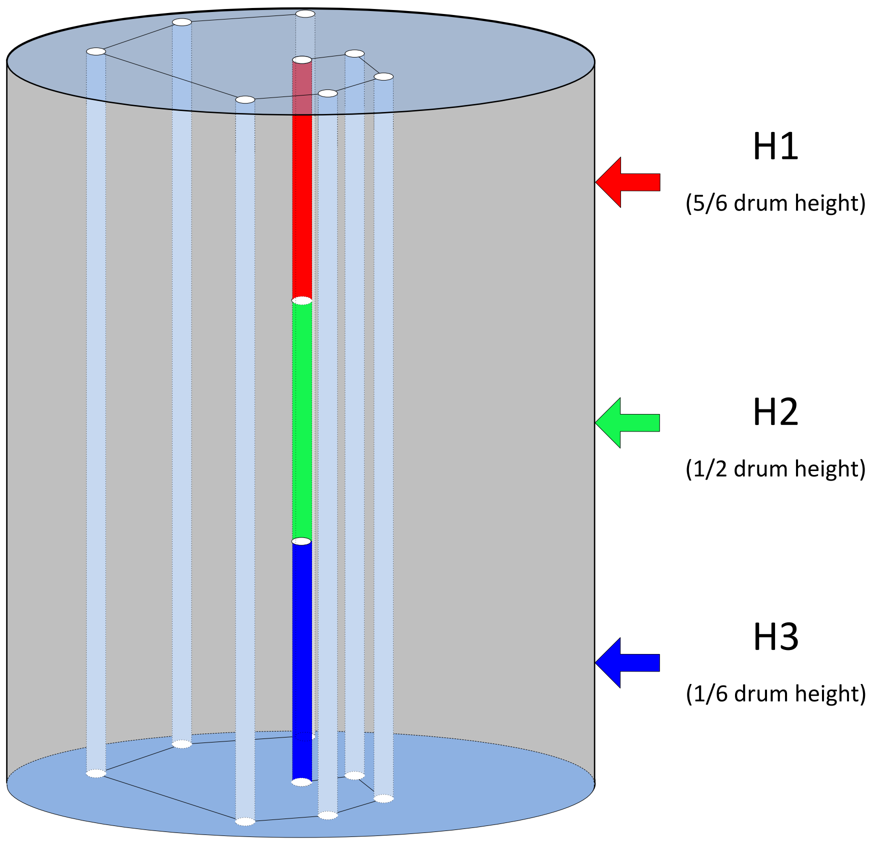

Figure 4. Test drum source positions (vertical) and azimuthal detector heights. There are 3 different source heights and detector heights.

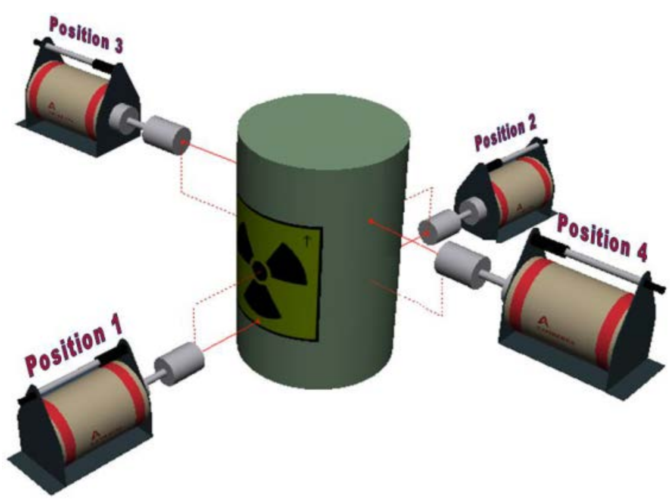

Figure 5. A typical ISOCS measurement geometry showing 4 detectors at mid-height.

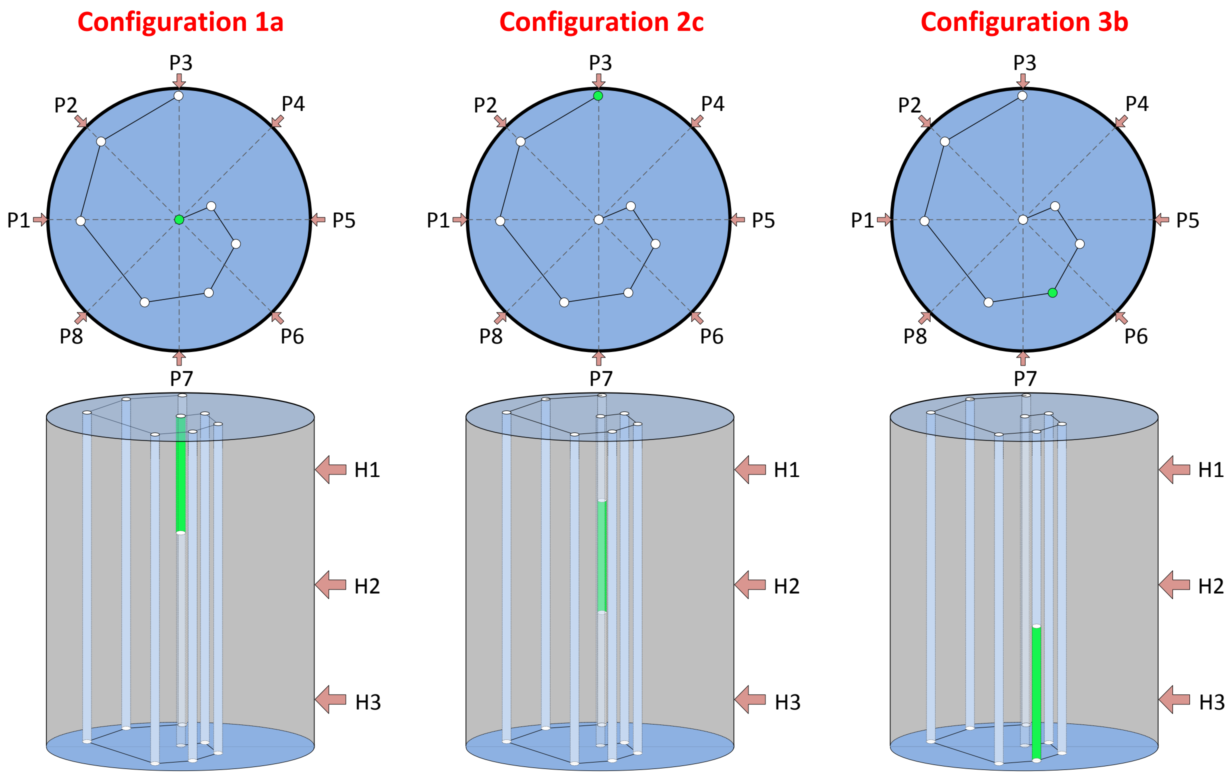

Figure 6. Diagram to show the three source configurations chosen for this study.

Analysis was conducted in “single nuclide” mode in which each nuclide was considered to have an independant spatial distribution (that is, different for each nuclide). Additional analysis was performed for Eu-152 and Ba-133 using a “mixed nuclide” analysis approach in which all the nuclides are assumed to be co-located. This provides an interesting benchmark that may be relevant for some waste assay applications.

Each measurement of the mineral wool matrix was conducted for a duration of approximately 1000 seconds. Activities were derived for each nuclide, using both the new AIGS tool (averaging the activity over the 4 x 3 = 12 chosen detector positions for each source configuration) and for a traditional ISOCS method assuming uniform activity (again averaging over the same 12 chosen detector positions). This comparison is instructive as it allows the value of the new technique to be assessed, taking into account both the assay bias that is to be expected due to assuming a uniform activity distribution, and the performance of the new method in performing a correction for source non-uniformity. Multi-count optimization was performed using the 4 detectors at different azimuthal positions, and also at 3 separate heights. The counting statistics uncertainty on each activity result was between 5 and 10% (at 1 standard deviation).

RESULTS

Results for the “mineral wool” matrix are summarized in Table I, Table II and Table III for source configurations 1a, 2c and 3b respectively.

TABLE I. Measured / Declared activity ratio results for source configuration 1a.

Analysis Method	Optimisation Method	Am-241	Cs-137	Eu-152	Ba-133
AIGS – Single nuclide	BRF	0.79	1.04		
AIGS – Single nuclide	Simplex	0.79	1.07		
AIGS – Mixed nuclides	BRF	0.88	1.13	1.14	1.11
AIGS – Mixed nuclides	Simplex	0.86	1.11	1.12	1.08
Traditional ISOCS	None	0.64	0.91	0.92	0.87

TABLE II. Measured / Declared activity ratio results for source configuration 2c.

Analysis Method	Optimisation Method	Am-241	Cs-137	Eu-152	Ba-133
AIGS – Single nuclide	BRF	0.70	1.00		
AIGS – Single nuclide	Simplex	0.77	1.03		
AIGS – Mixed nuclides	BRF	0.77	1.04	1.04	1.00
AIGS – Mixed nuclides	Simplex	0.78	1.00	1.00	0.96
Traditional ISOCS	None	0.92	1.12	1.10	1.02

TABLE III. Measured / Declared activity ratio results for source configuration 3b.

Analysis Method	Optimisation Method	Am-241	Cs-137	Eu-152	Ba-133
AIGS – Single nuclide	BRF	0.80	0.98		
AIGS – Single nuclide	Simplex	0.77	0.98		
AIGS – Mixed nuclides	BRF	0.71	1.02	1.03	0.96
AIGS – Mixed nuclides	Simplex	0.75	1.04	1.05	0.98
Traditional ISOCS	None	0.68	0.95	0.96	0.89

The results show modest matrix effects, reflecting the low matrix density, especially for Eu-152 and Ba-133 (for which the relatively high gamma energies result in modest gamma attenuation), therefore even with the traditional ISOCS method, matrix effects of no more than approximately 10 % are seen. However, the various AIGS methods do give a slight improvement in some cases, resulting in deviations from the true activity of only a few %. For Cs-137 the use of AIGS again leads to measured activities within a few % of the known activity (a slight improvement compared to the traditional ISOCS method).

In the case of Am-241 the improvement is significant and clear for the most "extreme" source position where the potential bias is pronounced (position 1a where the measured / declared ratio is 0.64 for the traditional ISOCS method), as expected due to the higher matrix attenuation of the 59.5 keV gamma ray. However, for many other positions, the matrix effect is rather small and this means that the benefits of AIGS are not obvious as the measured / declared ratio is already close to unity, even for the traditional ISOCS method, and therefore not easily discernible within the constraints of the counting statistics. In any case the trend in the results is as expected, note for example that position 1a (closest to the drum centre, therefore subject to maximum average gamma attenuation) gives the lowest measured / declared ratio, 0.64, for the traditional ISOCS method whereas position 2c (closest to the outside of the drum) gives a corresponding ratio of 0.92. With the exception of position 2c, the results demonstrate an improvement with the use of AIGS compared to "Traditional ISOCS", leading to measured / declared activity ratios that are closer to unity. This is especially the case for the "worst case" position 1a for which the improvement is from 0.64 to approximately 0.8 – 0.9 (depending on the optimization technique used).

Considering the inherent uncertainties, due principally to propagation of counting statistics for the measurements, of up to approximately 10%, these results are considered very encouraging. These results provide confidence in the value of the AIGS technique and also in the measurement and analysis work-flow that has been developed as part of the project.

Feedback from the recent work has allowed development of a procedure for future project implementation including the following key steps:

1. Identify optimum measurement geometry
2. Setup baseline ISOCS model
3. Specify parameter uncertainty range
4. Setup ISOCS models and specify benchmark and optimisation types
5. Perform multiple measurements
6. Run ISOCS models (automated within AIGS) to generate calibration files
7. Associate measured spectra with appropriate ISOCS calibration models
8. Run AIGS optimisation
9. Calculate final activity values.

SUMMARY AND CONCLUSIONS

CANBERRA's AIGS tool offers a powerful technique to drastically reduce the measurement uncertainties associated with waste measurements using model-based in-situ gamma spectroscopy. Recent experience has demonstrated the performance that can be achieved and the experience gained has allowed development of a procedure to allow robust implementation for projects.

The AIGS analysis technique offers highly flexible measurements with the major advantages that the same hardware as deployed for traditional ISOCS measurements, can be used with no or little modification, and the analysis is based on standard ISOCS software that has been extensively field-proven and validated. A typical AIGS analysis simply requires that measurements are performed using a prescribed sample – detector geometry but this can be achieved using widely available simple hardware based on turntables and detector lifts.

CANBERRA's recommended approach to take advantage of the substantial accuracy improvements that AIGS makes possible, is to engage experienced ISOCS physicists to assess each specific measurement challenge. This will allow an assessment to be made of the accuracy required by the project team, resulting in an assessment of whether and how existing ISOCS procedures can be adapted to allow AIGS analysis, or specification of bespoke field measurement procedures. Typically, AIGS should be deployed by specialists, for use as a pre-planning tool in this manner. Through exploratory AIGS (using the IUE features) modelling, an optimized measurement geometry and sequence can be developed, and predictions can be made as to the potential accuracy improvements that can be realized using AIGS to address the project objectives. This will include assessment of the measurement and analysis time and costs, so that the project can determine whether the additional investment in advanced AIGS analysis (more measurements possibly with extended counting times, and additional analysis costs), compared to "traditional" techniques (which use punitive assumptions), is worthwhile. Typically, it is expected that the investment costs are likely to be far outweighed by the cost savings associated with more efficient waste disposal (see Figure 1) if there are even a small number of containers that have radionuclide inventories close to the boundaries between two adjacent waste categories (particularly between LLW and ILW (TRU) for which the differences in disposal / storage costs are large).

Applications that can benefit from AIGS services are expected to include:

- Improved segregation of wastes (bags, drums, crates) in containers subject to waste matrix / source uncertainty: PCM (TRU) vs. LLW, LLW vs. VLLW, VLLW vs. Free Release, etc.

- Measurement of challenging bulk wastes with uncertain and variable packing methods (drums, ISO containers).

- Improved characterisation of legacy wastes (heterogeneous matrices, poorly defined radionuclide fingerprints).

- Safeguards or other field measurement projects (nuclear material items, waste items or in-situ plant measurements for decommissioning pipework, walls, building structures, etc) where limited sample data is available and / or limited time is available for measurements.

REFERENCES

1. J. Wachter, S.Stanfield, S.Croft, L.Bourva, M.Villani, R.Venkataraman, P.Chard, J.Harvill,
   "Model-Based Approaches to Calibration and Certification of Nondestructive Assay Systems", *Proceedings of the 41st Waste Management Symposia,* Phoenix, March 2015.

2. F. Bronson, V. Atrashkevich, G. Gerukov, B. Young, "Probabilistic Uncertainty Estimator for Gamma-Spectroscopy Measurements", J. of Rad. and Nucl. Chem., Vol 276, Pp 589-594.

3. A.Bosko. N.Menaa, T.Spillane, F,Bronson, R.Venkataraman, W.Russ, W.Mueller, V.Nizhnik,
   "Efficiency Optimization Employing Random and Smart Search Using Multiple Counts and Line Activity Consistency Benchmarks", *Proceedings of the 37th Waste Management Symposia,* Phoenix, February-March 2011.

4. A Bosko. N.Menaa, T.Spillane, F,Bronson, R.Venkataraman, W.Russ, W.Mueller, V.Nizhnik,
   "Waste Characterization Using Gamma Ray Spectrometry with Automated

Efficiency Optimization", *Proceedings of the 39th Waste Management Symposia,* Phoenix, February 2013.

5. P. Chard, "Radiometric challenges and approaches for the UK decommissioning programme", *3rd UK Decommissioning and Waste Management conference*, Penrith, UK, 10-11 July 2013