The Efficiency Calibration of Non-Destructive Gamma Assay Systems Using Semi-Analytical Mathematical Approaches - 10497

D. Nakazawa, F. Bronson, S. Croft, R. McElroy, W. F. Mueller and R. Venkataraman Canberra Industries Inc., 800 Research Parkway, Meriden, CT, 06450, USA.

ABSTRACT

A large variety of non-destructive assay (NDA) systems have been developed for the analysis of waste containers by passive gamma-ray spectroscopy. Examples of such systems include Segmented Gamma Scanners (SGS), Q^2 -style drum counters, Large Box Counters, and far-field measurement systems. The calibration of these systems typically involves the purchase and construction of "calibration" containers that are to varying degree representative of the items to be assayed. Typically these "calibration" containers represent idealizations of the waste stream with homogenized matrix materials and pseudo-uniform source distributions. But, for complete and correct calibrations, these containers must span the operational range of the system, and consequently include extreme and difficult configurations of matrix, density and source distribution. The NDA systems themselves typically have multiple configurations to deal with different kinds of items, and each configuration must be calibrated separately, often with different calibration sources. If the system is changed or modified, it must subsequently be recalibrated experimentally. The accuracy of such a calibration is typically estimated by performing a limited number of "worst-case" scenarios to establish the bounds of the Total Measurement Uncertainty (TMU). In its entirety, the calibration and uncertainty estimation of an NDA system represents a significant up-front expense.

Alternatives exist to the source-based calibrations of NDA systems. Among these alternatives is the mathematical modeling of the efficiency response of a given system to a particular waste configuration. In this situation, the system is calibrated using physically-relevant models of the gamma-ray response, with a relatively small number of validation measurements performed to demonstrate proper operation of the system and establish traceability to national standards. Due to the complexity of NDA systems and waste streams, purely analytically models are typically not feasible, but semi-analytical Monte-Carlo approaches based on the fundamental principles of gamma-ray interactions have proven to be very successful. These semi-analytical approaches are sufficiently versatile to allow the modeling of general waste stream configurations. Systems can be quickly adapted to changing waste streams or detector configurations. Not only can the measurement limits of the TMU be accurately estimated with semi-analytical techniques, but its functional form can be properly modeled to allow a more accurate assessment of the entire waste stream.

In this paper, we will present a survey of instruments including SGS, Box Counter, and far-field systems that have been calibrated using semi-analytical mathematical models. We will discuss experiences in calibration techniques, modeling of the TMU, and recent advances that further enhance the capabilities of such approaches.

INTRODUCTION

Gamma-ray spectroscopy is commonly employed to assay nondestructively nuclear waste containers of all shapes and sizes [1,2]. A central aspect of accurate quantification of nuclear material is the efficiency calibration of a given NDA gamma system. The efficiency, the number of events that are registered by the system per gamma ray emitted, is dependent on the energy of the gamma-ray, the geometry of the entire setup, and the composition of any material in the path of the item and the detector. Ideally, a

measured calibration is performed with known sources, distributed within representative packages identical to the unknown items to be measured during operation [1,2]. Important aspects of the measurement include similar waste containers, matrix materials, and radionuclides of interest between the calibration and unknown assay. There are many standards that outline the construction of calibration assay items, and many of them are site-specific [3-6]. Monte Carlo and semi-analytical methods that simulate radiation transport provide a tool to address virtually any aspect of the NDA measurement that departs from the ideal case.

The degree of departure from the calibration standards spans the range from minimal (a situation in which simulation is only used for feasibility or design) to entire (the calibration is completely modeled, then validated with measurement) [7]. At the smallest level, the use of simulations in designing systems requires that the application of the modeling has been benchmarked with prior measurements. More direct involvement as part of an efficiency calibration is the interpolation or extrapolation of the measured efficiency data points. An example of this would be when the production radionuclides differ from the calibration radionuclides. In this case, simulating intermediate energies or those outside the measured range can reduce the residual errors when fitting calibration curves. Other parameters, whose values could deviate from the calibration standards can be and are not limited to the following: container wall thickness, source-to-detector distance, matrix composition, matrix density, detector parameters (e.g. Ge dead layers), and sample attributes (isotopics, distribution). Finally, the efficiency calibration can be entirely simulated. This may be necessary for instances where a measured calibration is not feasible. No standards exist or are available, prohibitively large containers, and insufficient source activity in the case of highly collimated or attenuated geometries: these are a few examples. When the system efficiency is simulated, validation of the intrinsic efficiency of the detector and the modeling methods is highly desired and performed with measurement conditions that are available during calibration. Regardless of the involvement of simulation within an efficiency calibration, rigorous benchmarking and comparisons to measurement are essential to bolster confidence in the accuracy in the final assay results.

Another key aspect of any NDA gamma system is a strong understanding of its total measurement uncertainty (TMU). The purpose of the TMU is to assign a reasonable confidence band around the assay result to account for deviations from the calibration conditions used to quantify the radionuclides present. The TMU includes and is propagated from random and systematic uncertainties. The primary sources of uncertainties most likely to contribute to the TMU of a gamma NDA system include the following (in arbitrary order):

- Counting statistics
- Calibration uncertainties
- Rate loss correction factors
- Matrix inhomogeneity
- Non-uniform source distribution within the matrix
- Background fluctuations
- Interferences from other radionuclides present

- Self attenuation in 'lumps' of special nuclear material
- Isotopic measurement uncertainty
- Fill height uncertainty
- System stability
- Source-detector distance uncertainty
- Multi-curve efficiency calibration
- Transmission correction calibration

Other potential sources of measurement uncertainty are typically quite small in comparison to the ones listed above, and indeed not all those listed are significant in most circumstances. Of these sources of error, typically the matrix inhomogeneity and non-uniform source distribution tend to contribute the most to the TMU [8-10]. This is especially true for nuclides emitting gamma-rays of low energy (< 300 keV). To quantify the magnitude of these contributions, simulations provide a simple and cost-effective tool for

evaluation. The amount of labor, cost, and materials needed to estimate experimentally these contributions for a particular NDA system can be very high.

System design and feasibility studies comprise another large sector where the ability to simulate system responses is paramount. Monte Carlo and semi-analytical approaches to model radiation transport have contributed greatly in predicting system responses and planning the appropriate measurement solution. With respect to nuclear waste management and NDA measurements, assay items tend to vary greatly from site to site. Achieving the optimum system performance often requires both modification to existing or legacy counter designs and creating entirely new measurement modalities. Simulations and semi-analytical mathematical modeling of production scenarios allow for the exploration of a measurement solution, saving cost and time.

There exist several NDA gamma system archetypes that allow for the wide range of nuclear waste characterization. Far-field or hold-up measurement systems allow for large-area surveys and possibly the widest range of applicability. Shielded systems (e.g. Canberra's Q² or Ortec-Antech's QED) provide low-level waste characterization and free release determination. Segmented systems, such as the Segmented Gamma Scanner (SGS), Box Counters, and Tomographic Gamma Scanners (TGS) provide radionuclide characterization for a wide range of container sizes, waste streams, and source/matrix distributions. Monte Carlo and semi-analytical modeling of the efficiency response of a given system has proven to be crucial in all aspects of all of the forementioned NDA gamma system modalities.

The first section of this paper will briefly describe the tools and practices used with these systems. The second section will outline several case studies that highlight the major applications of simulations in NDA gamma systems. The final section will provide a summary and offer concluding remarks.

SIMULATION TOOLS AND METHODS

Modeling radiation transport with computer simulations has a rich history and is widely used in physics and engineering [11]. Of these, there exist several simulation tools that lend themselves to gamma NDA waste assay systems: MCNP, MCBEND, ISOCS, ISOTOPIC / ISOPLUS, SNAP, EGS, PENELOPE, and GEANT [7]. Monte Carlo and semi-analytical (deterministic) codes comprise the two main types of modeling, and each package has varying degrees of usability (i.e. learning curve), methodologies, and computer load.

The case studies presented here were primarily performed with MCNP and Canberra's ISOCS efficiency calibration software and its ISOCS Uncertainty Estimator (IUE) feature [12,13]. ISOCS (InSitu Object Counting Software) is deterministic and is based on ray-tracing to calculate attenuations due to source, container, environment, and detector materials. The intrinsic detector photo-peak efficiency is based upon a detector-specific MCNP model that is validated in the factory with a series of source geometries and gamma-ray energies. The IUE portion of ISOCS allows for every parameter (e.g. matrix density) within the template to be sampled from a given distribution. Efficiency calculations are performed repeatedly with new parameter values, facilitating the execution of sensitivity and TMU studies. Both programs have an extensive history of validation and verification, and more detail can be found in [12,13] and references contained therein.

Difficult geometries or highly specific measurement conditions that will be presented in the next section have been evaluated with MCNP where noted. Although this study focuses primarily on systems that use high purity Germanium detectors (HPGe) due to their excellent energy resolution and nuclide identification performance, the principles are also relevant to NaI and LaBr₃ detector systems. In all of the case studies with large waste assay systems, Canberra's NDA2000 software package was used for

data collection and analysis [14]. NDA2000 contains many of the algorithms and correction factor calculations that are either based upon or use directly the results of the presented simulations.

CASE STUDIES

Every gamma waste assay system modality has had some aspect of its calibration modeled by simulation. This section is aimed at providing some specific examples of where modeling is used in NDA waste assays. The particular examples given have all been constructed and deployed by Canberra Industries. The section is not intended to be an exhaustive list or a prescription of analysis methods.

Correction Factors / Analysis Methods

The use of simulations to provide a correction factor to an existing measured calibration is commonly used in NDA systems. The approximations and assumptions that simulations inherently introduce are minimal if the correction factor is a small percentage of the final assay result. One example is determining a correction factor for effects of container wall thickness in hold-up Uranium enrichment systems [15]. The underlying calibration constants that determine U-235 enrichment as a function of the peak area of the 186 keV line are determined with measurements. With low resolution detectors, such as NaI(Tl), it has been shown [16] with MCNP that if the wall thicknesses of calibration and assay items differ (e.g. UF_6 cylinders of varying sizes), the peak area of the 186 keV line will vary significantly when no collimator is used. Based upon MCNP simulations, an empirical correction factor was proposed.

A second example is source activity correction factors. In many cases, the information provided within the calibration source certificate indicates the contained activity of a given nuclide, not taking into account any source cladding or source material attenuation. When direct comparisons between certificate values and those using an efficiency calibration are made, the self-attenuation of the source materials must be calculated. Table I is taken from an SGS calibration report [17] in which the Aluminum cladding surrounding the epoxy source was 2 mm thick. The diameter of the active volume of the rod source is 5.49 mm, and the total length is 809 mm. The energy-dependent correction factors were calculated by taking the ratio of the ISOCS efficiencies without the Aluminum cladding to those with the Aluminum. Extensive research has been performed on the self-attenuating effect of Pu for waste and safeguards where simulations can provide correction factors [18,19]. Determining the geometrical factors for attenuation correction due to waste matrices using transmission calibrations has also been investigated with simulations; an example will be given for a large container assay system in the Full Calibration case study section [18].

Energy (keV)	Nuclide	Correction Factor
59.54	Am-241	1.142
81	Ba-133	1.108
276	Ba-133	1.071
303	Ba-133	1.069
356	Ba-133	1.065
383	Ba-133	1.063
662	Cs-137	1.052
1173	Co-60	1.040
1332.5	Co-60	1.037

Table I. Correction factors to the activity due to aluminum cladding surrounding the active portion of the rod sources. The data is taken from Ref. [17].

Another example is correcting for cascade summing loses from nuclides that emit cascades of gammarays (e.g. Co-60 or Eu-152) rather than single lines. In these cases, true-coincidences occur for gammaray emitted faster than the resolving time of the data acquisition system, and counts fall out of the full energy photo-peak. The amount of the losses is dependent upon nuclear data, source geometry, the peak efficiency, and the total efficiency of the detector. Analysis engines that apply a correction factor to the peak areas are used in Canberra's NDA2000 and Genie-2000 software. The cascade summing correction factor is derived from the ISOCS peak and total calculations [21]. A situation where this could occur in waste assay systems would be if the efficiency points and the resulting calibration curve were obtained from single-line gamma emitters, such as Cs-137, K-40, and Am-241, the source-to-detector distance was small (~tens of centimeters), and nuclides with cascade summing were measured (e.g. Co-60, Cs-134, and Y-88).

Interpolation / Extrapolation

Many NDA gamma calibrations employ polynomial curve fits to calculate efficiency as a function of gamma-ray energy and bulk density of the object. This collection of fits is often called a multi-curve efficiency and is used when there is a lack of transmission data for matrix correction [14]. First, containers with densities spanning the range of weights and typical compositions are assayed with radionuclides that have energies close to the production item energies. At each density, a measured efficiency curve versus energy is created. At a large number of the fitted energies, the efficiency is then fit over the measured density values. When production items are weighed and their densities are determined, the appropriate efficiency is calculated using these curve-fits. Errors from the fitting approximation are minimized when the original data points span the operation range. Any significant gaps in energy of density in the measurements can be supplemented with benchmarked simulations.

A good example of extrapolation of efficiency values using simulation is the calibration of tomographic gamma scanners (TGS). TGS systems [22] consisted of highly collimated HPGe detectors that scan waste drums in three dimensions. During an assay, the drum is divided into vertical segments, horizontally translated, and rotated. A transmission scan is performed with a transmission beam (typically Eu-152 due to its wide range of gamma-ray lines) to generate a matrix attenuation mapping of volume elements. A subsequent emission scan is acquired with the transmission source heavily collimated. A typical mapping contains emission and transmission data for 16 vertical slices, and 100 volume elements, or voxels, per slice. The efficiency value at each gamma-ray energy is comprised of the sum of all the emission peak areas in each voxel, compensated by the transmission map, divided by the activity of the calibration sources. The sum of the attenuation-corrected emission voxel responses is also referred to as the TGS number.

To generate the emission and transmission maps, TGS systems acquire spectra lasting only a few seconds in length, as the drum is rotating and translating. Due to the short length of acquisition, a Region-of-Interest (ROI) peak analysis is performed at known gamma-ray energies. Typical systems are calibrated with Ba-133, Cs-137, and Co-60. In order to obtain calibration factors for other nuclides at intermediate gamma-ray energies, linear interpolation is used [22]. For energies outside the range of the calibration energies (e.g. 303 keV from Ba-133 and 1333 keV from Co-60 for the low and high end, respectively), ISOCS is used. The detector and collimator efficiency is calculated using a point source, 25 centimeters away at all the energies of interest outside the calibration range. The TGS numbers per gamma per second (gps) at low energies *E*, are then obtained by scaling to 303 keV [1333 keV for the high energy regime] with the efficiencies, ε , simulated with ISOCS:

$$\frac{TGS\#}{gps}(E) = \frac{TGS\#}{gps}(303keV) * \frac{\varepsilon_{ISOCS}(303keV)}{\varepsilon_{ISOCS}(E)}$$
 Eq. 1.

An empirical fit is performed using the measured values (303, 356, 383, 667, 1173, 1333 keV from Ba-133, Cs-137, and Co-60), the interpolated values (e.g. 898 keV from Y-88), and extrapolated values (e.g. 1898 keV from Y-88). Figure 1 displays an example of such values from which the production calibration factors (TGS #/gps) are taken.



TGS # per Gammas per sec Vs Energy for 55 gallon drums

Fig. 1. Efficiency versus energy for a tomographic gamma scanner. The efficiency is labeled as TGS # / gps, where the TGS # is the attenuation-corrected emission data. The blue squares comprise the interpolated points in between 303 keV and 1333 keV, the green diamond and red triangles are the low and high extrapolations, respectively. The measured values are the open circles. The inset is a picture of the system. The data is taken from the system described Ref. [23].

In Fig. 1, the measured (open circles), interpolated (blue squares), and the extrapolated points (green diamond and red triangles) comprise calibration data taken from a TGS system built in 2009 by Canberra [23]. Although only the extrapolated points are directly scaled by ISOCS efficiencies, the production calibration factors use all of data points when generating the final curve fit.

Another illustration of the extrapolation to high gamma-ray energies is a Q^2 system [24] built in 2004. In this example, the Q^2 system was comprised of four HPGe detectors [25]. Three of the HPGe detectors are stacked vertically through the ports in the shielding, spanning the height of the waste drums. The gain was set such that energy range went to 1.5 MeV. A fourth detector mounted on another one of the sides was set for a gain of 5 MeV. The measured calibration was performed using Am-241 and Eu-152 rod sources. For energies above 1408 keV, the entire drum and detector geometry was simulated with ISOCS up to 7 MeV. For the fourth detector, measured and ISOCS efficiencies were used for the generation of the multi-curve. If benchmarked and validated properly, combining measured and simulated efficiencies can extend the capabilities of a given NDA gamma system.

Full Calibration

Complete simulated efficiency calibrations are needed when calibration standards are not available due to their size, complexity, activity, or cost. As discussed in Ref. [7], the utmost care, documentation, benchmarking, and validation must be adhered to when using a simulated calibration. As much measured data should be obtained with what is available, even though the validation geometries of the calibration will never be used in production. This section provides a few examples of NDA gamma systems that the calibration was completely simulated.

Segmented gamma scanner (SGS) systems are primarily designed to measure 55 and 83 gallon waste drums [1,26]. Many vendors only stock these containers sizes to perform standard, measured calibrations of the systems. For high dose rate situations, slotted collimators and/or layers of attenuating material will be placed in front of the detector. Calibration sources that are able to penetrate these attenuators and thick concrete container walls are not typically maintained by vendors. Table II includes examples of two SGS counters and the containers for which ISOCS was used for their calibrations. In all cases, a full measured validation of the 55 gallon drum geometry was performed. Far-field assay systems that do not scan the assay item also require calibration using simulation if container sizes differ from available containers [27].

Table II. A few examples different segmented gamma systems and the containers for which ISOCS was employed for efficiency calibration.

High Activity SGS (2005)	Integrated Crate Interrogation System (ICIS) counter (2007)	SGS (2009)
 400 Liter with concrete liner 400 Liter with concrete and steel liners AX3 container (steel and concrete lined) 	 Standard Waste Box Standard Large Box – II Ten Drum Overpack 	 Four types of concrete containers of varying wall thicknesses

A key example of a simulated calibration is the box segmented gamma scanner (BSGS) [28]. Typical counters have towers of multiple HPGe detectors on either side of a moving platform (Fig 2a). Waste containers can be as large as 6700 liters (e.g. the Standard Large Box – II) where measuring calibration standards are not feasible. The box counter shown in Fig. 2 (Integrated Crate Interrogation System) used MCNP and ISOCS in almost every aspect of calibration and TMU estimation [29]. Aspects of the TMU analysis will be discussed in the next section.

The multi-curve efficiency calibration of ICIS was performed with ISOCS for all containers listed in the middle column of Table II at densities up to 1.5 g/cc, which is limited by the maximum weight capacity of the containers. The only measured values used in the calibration were the empty container efficiencies which were used to normalize the empty ISOCS efficiencies. As one can see from Table III, the agreement for the empty containers is approximately 4% over all energies from 60 to 1333 keV.

Table III. Comparison of the measured efficiencies (corrected for line source self-attenuation) and the ISOCS efficiencies for the empty containers for the ICIS Gamma Box Counter. The relative uncertainty (unc.) is shown at one standard deviation. The data is taken from calibration report is the system described in Ref. [29].

	Efficiency Ratio – Measured/ISOCS							
Energy (keV)	S	WB	SLB-2		TDOP			
	Ratio	Unc.	Ratio	Unc.	Ratio	Unc.		
59.5	1.08	12.50%	1.04	12.60%	1.02	12.76%		
81.0	0.97	10.50%	0.93	10.46%	1.00	10.48%		
122.1	0.92	9.55%	0.89	9.51%	0.95	9.52%		
276.4	0.97	8.61%	0.95	8.57%	1.00	8.58%		
302.9	0.96	8.61%	0.94	8.57%	0.99	8.58%		
356.0	0.96	7.69%	0.94	7.64%	0.99	7.66%		
383.8	0.95	7.69%	0.94	7.64%	0.98	7.66%		
661.7	0.96	6.81%	0.94	6.74%	0.98	6.76%		
1173.2	0.97	5.14%	0.97	5.04%	1.00	5.08%		
1332.5	0.97	5.14%	0.97	5.04%	0.99	5.08%		

For the validation of loaded containers, sample matrices were procured and constructed. Holes were drilled into the matrices, and numerous measurements were performed shuffling the rod sources amongst every position. For example, the SWB container had 55 evenly spaced holes to cover the entire volume. The measurements using 8 rods at a time were then summed together to get as close to a uniform distribution as possible. Table IV shows excellent agreement between measured and expected activities once a non-uniform distribution correction was performed for the SWB and SLB-II containers. For energies above 122 keV (Co-57), the average agreement is 7%. When Am-241 (60 keV) is included, the average agreement is 12%. More specifically, the summed rod source measurements were taken to be rectangular plane sources corresponding to the rod source spacing shown in Fig. 2b. The final calibration assumes a uniform source distribution.



Fig. 2. (a) ICIS Gamma counter with a Standard Waste Box loaded on the conveyor. (b) Example of the plane source distribution for the measured validation of ISOCS efficiency calibrations of the Standard

Large Box – II container. The view is from above, and the red bars indicate the source material. The detector positions at the 5 scanning positions are also shown.

Table IV. ISOCS calibration validation results from the ICIS counter for the Standard Waste Box (SWB) and the Standard Large Box – II (SLB-II). The values shown are the ratio of the measured activity to the expected activity with a non-uniform source distribution correction. The relative uncertainty is shown at one standard deviation. The data is taken from the calibration report of the system described in Ref. [29].

Container	Nominal Density (g·cm ⁻³)	⁵⁷ Co	⁶⁰ Co	¹³³ Ba	¹³⁷ Cs	²⁴¹ Am
SWB	0.152	$0.92\pm9.11\%$	$0.98 \pm 4.00\%$	$0.91\pm7.98\%$	$0.95\pm6.01\%$	$1.20\pm14.92\%$
	0.297	$0.99\pm9.09\%$	$0.98\pm4.01\%$	$0.92\pm7.98\%$	$0.95\pm6.01\%$	$1.62 \pm 14.19\%$
	0.580	$1.01 \pm 9.15\%$	$0.99\pm4.01\%$	$0.91\pm7.98\%$	$0.97\pm6.01\%$	$1.57 \pm 15.53\%$
	0.780	$1.15 \pm 9.15\%$	$1.05\pm4.01\%$	$0.99\pm7.98\%$	$1.06\pm6.01\%$	$1.49 \pm 15.68\%$
SLB-2	0.027	$0.90\pm9.05\%$	$0.97\pm4.00\%$	$0.92\pm7.98\%$	$0.95\pm6.00\%$	$0.96 \pm 14.10\%$
	0.064	$0.85\pm9.07\%$	$0.95\pm4.00\%$	$0.88\pm7.98\%$	$0.92\pm6.01\%$	$0.82\pm14.09\%$
	0.152	$0.87\pm9.09\%$	$0.96\pm4.00\%$	$0.90\pm7.98\%$	$0.94\pm6.01\%$	$1.12 \pm 14.62\%$
	0.565	$0.92\pm9.13\%$	$0.97\pm4.01\%$	$0.91\pm7.98\%$	$0.95\pm6.01\%$	$1.34 \pm 14.58\%$

The ICIS gamma counter also has the capability to perform transmission corrections to account for matrix attenuation. The analysis used to calculate the matrix attenuation requires a parameter, κ , that is dependent on the geometry of the container [1,20]. Equation 2 is an example of a form of the correction factor *CF*, applied to the empty matrix efficiency calibration:

$$CF_{Matrix} = \frac{\kappa \cdot x}{1 - e^{-\kappa \cdot x}}$$
 Eq. 2,

where κ is a geometry dependent factor, *x* is equal to ln(1/T), and T is the measured transmission of a collimated beam through the assay item. For drums, an approximate value of 0.80 is generally used for κ . For ICIS, MCNP was used to simulate the transmission sources and detectors at various matrix densities, and the correction factors were calculated with ISOCS to numerically solve for the κ values for the larger containers [20]. Benchmark measurements were performed for the 55 gallon drums, SWB, and SLB-II containers with a range of densities, and agreement between simulation and measurements for the transmission values was within 10%.

Total Measurement Uncertainty

The TMU is a critical specification of any waste assay system. Significant contributions to the TMU of non-destructive assays that use quantitative gamma-ray spectroscopy stem from the characteristics of the attenuating container matrices, such as matrix inhomogeneities and non-uniform source distributions. Because the extent of these effects is typically unknown for a given container, waste assay systems generally assume a uniform matrix and source distribution in determining the container activity.

Accurate knowledge of the bulk density is necessary for using multi-curve efficiency calibrations for gamma counters. One study by Bosko et al. [30] used ISOCS to simulate the effects of differences in the tare weights calibration drums and production drums with an SGS counter. Comparing both analytical methods, ISOCS simulations, and measurements, it was determined that uncertainties on the order of 3-

5% above 100 keV were to be expected for different drum weights. The differences in tare weight resulting from either matrix material or drum wall thickness were found to be the same order of magnitude. The ability of simulations to quantify these contributors in isolation adds significant value.

Due to the size of box counters, it is often the case that the radioactive material is not uniformly distributed within the waste container. To quantity this uncertainty ICIS box counter, five randomly located point sources were simulated using the ISOCS Uncertainty Estimator (IUE) [10]. The ratio of the efficiency of the five sources and a uniformly distributed of the same activity was binned as a function of energy and density for all containers. By estimating the maximum and minimum deviations of the system response, empirical curve-fits were generated as a function of transmission, and one-sigma uncertainty values were estimated. Table V shows an example of the results for the SLB-II container. With uniform matrices, five point sources were measured with the ICIS counter to benchmark this uncertainty estimation. Fig. 3 shows the results for one container for Co-60. Fifteen different measurements were performed, and the agreement with the ISOCS efficiency and the IUE simulation is excellent. For much higher degrees of non-uniformity, such as a single source in the center of the container, the deviations from the uniform calibration are more severe.

Table V. One standard deviation uncer	tainty estimations due t	o non-uniform source distr	ibutions for ICIS
box counter and the SLB-II container.	The values are in (%).	The data is taken from Real	f. [10].

Energy (keV)> Density (g/cc)	59	100	129	186	414	662	1173	1408	2000	4000
Empty	9	7	7	7	7	7	7	7	7	7
0.15	35	28	27	25	21	18	16	15	14	12
0.3	50	45	43	41	34	30	25	24	21	17
0.6	73	64	63	62	56	50	41	39	34	26
0.9	78	71	69	68	62	58	50	48	43	35
1.2	93	86	85	81	70	67	57	54	50	41
1.5	111	95	92	91	84	79	72	69	64	53



Fig. 3. Fifteen different measurements of five randomly distributed point sources within an SLB-II (uniform cardboard matrix) container using the ICIS gamma box counter. The efficiency calibration used to determine the Co-60 activity assumed a uniform source distribution. The 1-sigma uncertainty value from simulations is shown by the dotted lines, and the solid line is the average measured activity. The data is taken from Ref. [10].

Design and Feasibility

New measurement solutions to waste, safeguard, accountability, and safety challenges have simulated efficiency calibrations at their core of their design and feasibility stage. From developing the next generation portal monitor to determining shielding effectiveness, simulation offers a great tool to explore new system modalities.

Recent examples of using MCNP and ISOCS for design and feasibility of gamma systems at Canberra have included systems for fuel pin assembly safeguards, remote monitoring systems (RMS) sensitivity studies for nuclear power plants, meeting NDA system shielding specifications, and segmented scanners for small, can-sized objects [11]. One study recently investigated the sensitivity of a NDA in-situ system for the decommissioning of large hot cells. The assessment of detector type, counting times, efficiency calibrations, scanning length of embedded pipes: all of these aspects were investigated in the use a thin-window HPGe detector to measure the X-rays from alpha-emitters. Fig. 4 shows examples of some of the ISOCS calculations performed in the study to estimate scanning length [31]. Pictured are surface sources (orange) of a 103 mm pipe in increasing length and the corresponding efficiencies of the detector (grey).



ISOCS Efficiencies for 40% REGe Detector for a surface source in a 103 mm pipe with varying source length



Fig. 4. ISOCS simulations to assess the scanning length for surface contamination NDA gamma system and the resulting efficiency results versus energy. The pictures and graphs are taken from Ref. [31].

A few recent studies have exercised the IUE utility to optimize counting geometries for waste applications and to compare sampling methods vs. in-toto measurements [32]. By permuting several operational parameters (number of non-uniform hot spots, size of hot spots, number of detectors, detector positions, etc.) and comparing the deviations to a uniform standard, many useful conclusions were found when assaying 55 gallon drums. For example, detector-to-drum distance is a trade-off with respect to non-uniform source distributions. Larger distances decrease deviation but lower detection efficiency, increasing count times. Rotation of the drum greatly decreases the deviation, while vertical scanning does not. A second detector on the opposite face of the drum also helps, and this can also be achieved by rotating the drum 180 degrees halfway through the assay. Having the ability to quickly simulate efficiencies with a wide range of adjustable parameters not only saves time and cost, but allows for the creation of improved NDA gamma systems.

DISCUSSION AND CONCLUDING REMARKS

Recent developments [33] have been shown to use ISOCS to dynamically adapt a calibration where knowledge of assay item is sparse or in question. In the case of uranium and plutonium, the relative efficiencies of peaks with respect to other can be determined using isotopic codes (e.g. MGA, FRAM). Ongoing work has shown that parameters such as container wall thickness or matrix density can be varied dynamically to obtain a more accurate efficiency calibration [34]. Adaptive efficiency correction is one example of future work and development that simulations can contribute to improving NDA accuracy and confidence.

A vast majority of the instruments use semi-analytical efficiency calibrations (e.g. ISOCS) for far-field waste characterization [11,12]. Many large items, such as large casks or whole rooms, do not easily lend themselves to calibration standards. In addition to cost and time savings, the ability to simulate the efficiency of virtually any item-detector configuration increases a system's versatility. Situations could arise in which one or two items within a waste stream are oddly shaped or exceedingly high in radioactivity. Having a previous validated model can provide more confidence that a newly generated calibration with an additional attenuator in front of detector will yield accurate results.

This study is not intended to be a complete or exhaustive survey of all examples in which semi-analytical methods and simulations are used in NDA gamma systems. Specific instances of using simulations in the varying degrees of aiding measured calibrations have been addressed. With increasing vigilance on safeguarding all forms of nuclear material, sourceless efficiency calibrations have become desirable. Saving cost and time by assaying large containers prevents the need to repackage waste into smaller forms. Other savings include upfront and upkeep costs of maintaining an inventory of radioactive material for calibration purposes. Computations and numerical methods are becoming increasingly faster with the improvements in processors. Extensive design studies and adaptive optimization have been shown to improve accuracy and reduce the TMU of NDA waste assay systems. Extending the operational energy and density range are sometimes only possible with simulated efficiency calculations. This is also true for many aspects of the TMU of a system. Coupled with benchmarks and measured validations, simulated system efficiencies provide a useful tool to tackle many challenges facing gamma waste characterization.

REFERENCES

- 1. D. REILLY, N. ENSSLIN, and H. SMITH, Editors, Passive Nondestructive Assay of Nuclear Materials, LA-UR-90-732, Los Alamos National Laboratory, 1991.
- 2. P. MCCLELLAND and V. LEWIS, Editors, Measurement Good Practice Guide No. 34, Radiometric Non-Destructive Assay, 2003.
- 3. Guide to Calibrating Nondestructive Assay Systems, ANSI N15.20-1975, 1975.
- 4. Standard Test Method for Nondestructive Assay of Special Nuclear Material in Low-Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning, ASTM C1133-03, 2003.
- 5. H. SMITH, J. STEWART, and W. RUHTER, Design of standards for Nondestructive Assay of Special Nuclear Material, LA-UR-97-766, 1997.
- 6. American National Standard for Calibration of Germanium Detectors for In-Situ Gamma-Ray Measurements, ANSI N42.28-2002, 2002.
- 7. P. CHARD, Editor, A Good Practice Guide for the use of Modelling Codes in Non Destructive Assay of Nuclear Materials, IAEA Issue 2.0, 2009.
- 8. B. YOUNG, S. CROFT, and H. ZHU, The Influence of Source and Matrix Nonuniformity on TMU and Bias of Large Container Gamma-Ray Assay Results, Presented at the 47th INMM Annual Meeting, 2006.

- 9. S. CROFT, R. MCELROY, B. YOUNG, and R. VENKATARAMAN, Quantifying SGS Matrix Correction Factor Uncertainties for Non-Uniform Source Distributions, Presented at the 45th INMM Annual Meeting, 2004.
- 10. D. NAKAZAWA, S. CROFT, M. HENRY, W. MUELLER, R. VENKATARAMAN, M. VILLANI, and H. ZHU, Determination of Non-Uniform Source Distribution Uncertainty of Waste Assay Systems using ISOCS Uncertainty Estimator, Presented at the ISRSM Conference, 2007.
- F. BRONSON, R. MCELROY, S. PHILIPS, W. RUSS, and S. CROFT, The Application of Mathematical Modeling for Commercial Nuclear Instrument Design, Development, and Calibration, Presented at the 1st ANIMMA International Conference, 2009.
- R. VENKATARAMAN, F. BRONSON, V. ATRASHKEVICH, B.M. YOUNG, and M. FIELD, Validation of in-situ object counting system (ISOCS) mathematical efficiency calibration software, Nucl. Instr. and Meth. A 422 (1999) 450.
- F. BRONSON, G. GEURKOV, and B. YOUNG, Probabilistic uncertainty estimator for gammaspectroscopy measurements, Journal of Radioanalytical and Nuclear Chemistry, Vol. 276, No. 3 (2008) 589-594.
- 14. NDA-2000 Technical Reference Manual, (Canberra DOC #9231595D), Canberra Industries, Inc.
- 15. S. PHILIPS, S. CROFT, and A. BOSKO, Enhancements to the IMCA Software, Presented at ESARDA meeting, 2007.
- H. SCHWENN, S. CROFT, A. BOSKO, S. PHILIPS, and R. VENKATARAMAN, Simulation of the Effects of Small Angle Scattering When Applying the Enrichment Meter Principle, Presented and 50th INMM Meeting, 2009.
- 17. SGS Calibration Report, Canberra Industries, Inc., 2009.
- S. CROFT, R. MCELROY, S. PHILIPS, R. VENKATARAMAN, and D. CURTIS, A Review of recent developments in Self-Attenuation Correction leading to a Simple Dual Lump Correction Model for Use in the Quantitative Gamma Assay of Plutonium, J. Radioanal. Nucl. Chem. 276 (2008) 667.
- 19. T. MILLER, Lump Correction for Radioactive Waste Assay, J. Radiol. Prot. 29 (2009) 385.
- H. ZHU, S. CROFT, R. VENKATARAMAN, and S. PHILIPS, An MCNP Based Method to Determine the Matrix Attenuation Correction Factors for a Gamma Box Counter, Presented at 48th INMM Meeting, 2007.
- H. ZHU, K. MORRIS, W. MUELLER, M. FIELD, R. VENKATARAMAN, J. LAMONTAGNE, F. BRONSON, and A. BERLIZOV, Validation of true coincidence summing correction in Genie 2000 V3.2, J. Radioanal. Nucl. Chem. 282 (2009) 205.
- 22. S. CROFT, D. BRACKEN, S. KANE, R. VENKATARAMAN, AND R. ESTEP, Bibliography of Tomographic Gamma Scanning Methods Applied to Waste Assay and Nuclear Fuel Measurements, Presented at the 47th INMM Meeting, 2006.
- N. MENAA, D. NAKAZAWA, H. YANG, S. SMITH, D. PETROKA, and M. VILLANI, Tomographic and Segmented Gamma Scanning for Nuclear Power Plant Applications, Presented at WM2010, 2010.
- 24. F. BRONSON, Q² A Very Low Level Quantitative and Qualitative Waste Assay and Release Certification, Presented at WM1990, 1990.
- 25. Automated Q² Calibration Report, Canberra Industries, Inc. 2004.
- 26. E. MARTIN, D. JONES, and J. PARKER, Gamma-Ray Measurements with the Segmented Gamma Scanner, LA-7059-M, 1977.
- 27. S. HALLIWELL and S. JEONG, Design of a Facility for the Receipt Inspection and Characterization of L/ILW Using an Integrated System of Non-Destructive Examination and Non-Destructive Assay Techniques, Presented at WM2010, 2010.
- 28. R. MCELROY, S. CROFT, and B. YOUNG, Non Destructive Assay Box Counter, Presented at the the 46th INMM Meeting, 2005.

- 29. S. PHILIPS, A. BOSKO, S. CROFT, R. MCELROY, D. MOSCATO, R. MOWRY, W. MUELLER, D. NAKAZAWA, D. PETROKA, R. VENKATARAMAN, M. VILLANI, B. YOUNG, and H. ZHU, An Overview of the Integrated Crate Interrogation System (ICIS) for Use at the Savannah River Site, Presented at WM2008, 2008.
- 30. A. BOSKO, S. CROFT, and E. GULBRANSEN, Estimated Uncertainty in Segmented Gamma Scanner Assay Results due to the Variation in Drum Tare Weights, Presented at WM2009, 2009.
- 31. D. Nakazawa, Technical Description of an In-Situ HPGe System for the Detection of Alpha-Emitting Nuclides for Dansk Dekommissionering, Internal Report, Canberra Industries Inc., 2009.
- 32. F. BRONSON, To Sample or Not to Sample: An Investigation into the Comparative Benefits of Sampling Followed by Laboratory Analysis Versus In-Toto Gamma Spectroscopy for Situations of Non-Uniform Radioactivity Distribution, Presented at WM2008, 2008.
- 33. W. RUSS, N. MENAA, D. NAKAZAWA, A. BOSKO, R. VENKATARAMAN, and F. BRONSON, Evaluation of Numerical Techniques for Optimization of ISOCS Modeled Detector Measurement Geometries, Presented at the International Conf. on Mathematics, Computational Methods, and Reactor Physics, 2009.
- 34. W. RUSS, N. MENAA, D. NAKAZAWA, A. BOSKO, R. VENKATARAMAN, and F. BRONSON, Private Communication, 2010.