An Alternate Approach to the Add-A-Source Matrix Calibration for SLB-2 Containers in the Box Neutron Assay System - 9339

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ABSTRACT

The Box Neutron Assay System (BNAS) provides the passive neutron counting capability of the Integrated Crate Interrogation System (ICIS) installed at the Savannah River Site. The system is designed to assay transuranic waste in large containers up to and including the Standard Large Box 2 (SLB-2). A multi-position californium Add-A-Source (AAS) capability is built into the system to determine gross matrix correction factors for the moderating effects of waste containers over a wide range of matrix materials and densities. The AAS correction factor is based on a calibration curve for the Reals (i.e. coincidence) rate for a given container, with the calibration curve being derived from Volume Weighted Average (VWA) rates and AAS perturbation factors for different homogeneous matrices. For the SLB-2 container the large volume poses difficulties in the calibration and verification measurements, particularly for matrices with high concentrations of hydrogen where steep spatial gradients are observed in the response.

The VWA rates for AAS calibrations are typically obtained by measuring the response of a Cf source at several distinct locations within the matrix, and then applying a numerical integration based on proportionate weighting factors. The technique has been proven to work well for small containers. In the case of the SLB-2 container with high density matrices, however, the calibration was found to be highly sensitive to the numerical integration technique. This sensitivity results in a large contribution to the overall Total Measurement Uncertainty (TMU) for the end measurement. In this work a Monte Carlo approach was taken in order to better constrain the AAS calibration curve, and reduce the TMU in the final application.

An MCNP model for the BNAS was first benchmarked to the point-source measurements made in the various calibration matrices (the empty container reference case was included in the benchmark). The model then provided the numerical integration necessary to obtain the response from a uniformly distributed source for each matrix, which was otherwise limited by the number of measurements required to cover the full volume of the matrix. Once the model was tailored to match the data at an acceptable level, it also allowed for the possibility of expanding the calibration that was otherwise limited by the number of available calibration matrices. This required the generation of the AAS perturbation response as well, which was also benchmarked to the calibration data. With the model suitably benchmarked, additional calibration points were generated to constrain the shape of the calibration curve. This alternate approach results in a better constrained AAS correction factor over a wide range of matrix densities, and results in a reduced uncertainty contribution to the TMU. Since the model can be further modified to study other possible variations in the measurement scenario, such as partially filled containers or heterogeneous matrices, it can also be used when needed as a tool to guide expert analysis.

INTRODUCTION

The Box Neutron Assay System (BNAS) [1] has been installed at the Savannah River Site (SRS) and is intended to quantify Pu-240-effective mass and provide total alpha activity for transuranic (TRU) waste that is to be shipped to the Waste Isolation Pilot Plant (WIPP). As the neutron complement of a combined gamma-neutron system, the neutron assay also improves the accuracy of the result that is otherwise limited by matrix or self-absorption effects.

An Add-A-Source (AAS) Matrix Correction assembly has been incorporated into the counter using a built-in interrogation source. The AAS calibration procedure is intended to provide a correction for the moderating effects of waste containers. The correction for matrix moderation is based on a multi-position AAS technique that has been proven to work well in the case of smaller containers; e.g. 208L drums [2, 3]. The BNAS, however, is designed to assay containers as large as the Standard Large Box 2 (SLB-2) which has (inner) dimensions of ~259cm in length, ~160cm in breadth, and ~168cm in height (8.5' x 5.3' x 5.5'), with a capacity ~30 times the volume of a 208L drum. This sets the scale of the counter which comprises 320 He-3 filled proportional counters arranged in a 4 geometry around the assay cavity. Figure 1 shows the BNAS with an SLB-2 container loaded on the conveyor system in preparation for an assay.



Fig.1. Box Neutron Assay System (shown with SLB-2 on the load conveyor).

The AAS correction technique is based on a correlation between the perturbative effects of a matrix on an external interrogation source (the Cf-252 AAS), and the volume perturbation response for that matrix that has been pre-determined by calibration. During the assay of a given container a small fraction of the assay time is reserved for the insertion of the Cf-252 AAS into the assay chamber in order to perform the AAS perturbation measurement. (When not in use the Cf-252 AAS is stored outside the assay chamber in a high-density PE shield in order to minimize the background.) The measured perturbation response then needs to be correlated with the previously defined AAS calibration in order to obtain an AAS correction factor that is then applied to the passive neutron measurement result.

The AAS calibration is made at the factory prior to shipment of the counter and is intended to span a range of matrix densities and hydrogenous content. A set of calibration matrices therefore need to be constructed in order to establish the correlation between the AAS perturbation and the volume perturbation response as a function of matrix. The volume perturbation response is obtained by measuring a point source at multiple spatial locations within each container matrix thereby generating a map of the variation in moderating effects over the entire volume of the container. A Cf-252 source is typically used for this purpose as well as it also serves as a surrogate for Pu-240. Since the reference calibration condition is for activity uniformly distributed throughout a container that is completely filled with a homogeneous matrix, equal volumes of the container can be assumed to hold equal matrix mass of equal activity. In order to extend the point-source activity measurements to a distributed activity over the entire matrix, a weighting scheme is used to account for the representative volume associated with each spatial location of the point source. A Volume Weighted Average (VWA) activity can then be extracted from the measurements. This approach will potentially depend on the number and choice of the point source spatial locations within the matrix, as well as on the weighting scheme itself, to a degree which depends on the size of the container.

With increasing container size the impact of moderating materials can result in under-reporting of the fissile mass as much as an order of magnitude, and perhaps more. The extension of the application of the AAS technique to large containers is therefore extremely important.

PREVIOUS WORK ON THE BNAS AAS CALIBRATION

A description of the existing BNAS AAS calibration [4] is summarized here in order to provide context to the new approach. In addition to the much greater impact of the increased volume of moderation material, for the large containers a greater degree of heterogeneity is anticipated in the contents as well. Consequently a more varied impact is expected on the measurement result from the distribution of the moderating matrix within the container. To mitigate the impact of this variation, the AAS was designed to allow for multiple positions within the assay chamber, and the number of positions and location of each position was pre-programmed as a function of container. In the BNAS the Cf-252 point AAS is attached to the end of a steel Teleflex cable and runs inside a U-shaped steel track at the bottom of the assay chamber, with each branch of the "U" being equidistant from the lengthwise symmetry axis of the counter. The position of the AAS can be programmed to stop at any point along the U-shaped track. For the SLB-2 container ten AAS positions were defined with five equally spaced positions on each branch of the "U". The perturbation measurement thus comprises ten sub-measurements (of equal time) which are then averaged to give the AAS perturbation for the entire container. (In previous applications for 208L drums the use of 1-3 AAS positions along the length of the drum has been shown to be sufficient.) In order to minimize the count time (and maximize throughput) the strength of the Cf-252 AAS is chosen accordingly, and for the BNAS a 3.2×10^5 n/s source was used so that for on-site assays each submeasurement would only be 1 minute long giving a total perturbation measurement time of 10 minutes out of a 60-minute nominal total assay time. During the calibration process the measurement time for each AAS position was chosen to be 10 minutes (giving a total perturbation measurement time of 100 minutes per matrix), so that the statistical uncertainty in each measurement would not be a limiting factor in the calibration. Denoting R_a and R as the averaged AAS Reals rates for the empty SLB-2 container and SLB-2 with a given matrix respectively, the AAS perturbation is defined by

$$x = \frac{R_o}{R} - 1 \tag{Eq. 1}$$

For the volume perturbation measurements required for the calibration, a much more imposing expansion in scale is required when going from measuring 208L drums to SLB-2 containers. Typically 20 spatial locations are used within a 208L drum with any two adjacent locations no more than 20-25cm apart. A mapping of the SLB-2 container with similar spacing would require over 500 measurements so that even for low-density matrices where a reasonable count time might be 5 minutes, approximately 40 hours of assay time would be required per matrix. Additional time for re-positioning the source between measurements would then add considerably to the overall measurement time. One approach to reducing the calibration time was to take advantage of the 4 coverage and axial symmetry of the BNAS and only make measurements in one quadrant of each matrix. Due to the presence of the conveyor rollers at the bottom of the assay cavity full symmetry along the vertical axis was not assumed, and measurement locations were chosen along the full height of the matrix. In addition the distance between source positions was increased to 40cm (~16"), but the impact on the determination of the appropriate volume weighting factors was expected to be minimized using analytical or Monte Carlo methods. Denoting R_o^V and R^V as the volume-averaged Reals rates for the empty container and container with matrix respectively, the volume perturbation is defined by

$$y = \frac{R_o^V}{R^V} - 1 \tag{Eq. 2}$$

The objective of the AAS calibration effort is to supply a matrix correction calibration curve for the Reals rate [2] in the form given by the following equation:

$$y = a_o + a_1 x + a_2 x^2 + a_3 x^3$$
 (Eq. 3)

The AAS correction factor *CF* that is applied to the passive neutron measurement result is then defined by:

$$CF = 1 + y \tag{Eq. 4}$$

To establish the AAS calibration for the SLB-2 containers, large matrix modules containing a mixture of plywood, cardboard, and polyethylene were used with pre-fabricated channels for positioning of a source. The choices were designed to span a reasonably broad H_2 concentration using materials that are readily available in bulk and can be fabricated into self-supporting modules. The list of matrices used for the SLB-2 AAS calibrations is provided in Table I below.

Material	Nominal Densities (g/cc)		
Reference (Empty)	0		
100% polyethylene foam	~0.027		
100% polyethylene foam	~0.064		
100% corrugated cardboard	~0.17		
100% plywood	~0.50		

 Table I. SLB-2 Matrices used for AAS Calibration

Figure 2 shows the source locations chosen for the volume perturbation measurements which were performed in one quadrant of the matrix at ~40cm intervals in all three dimensions. Measurements with the source indented on the surface of the matrix were used to ensure that the full extent of variation was represented in the data. The VWA was obtained by averaging the Reals rate at each measurement location with an appropriate weighting scheme to account for the variation in response over the entire matrix. Several weighting methods were evaluated and the method applied was based on a weighting factor that was inversely proportional to the volume of a shell, where the total volume of the matrix was sub-divided into concentric shells. The shell boundaries also shown in Fig. 2, and all points within a given shell share the same weight.



Fig. 2. Top and Cross-section views of SLB-2 showing shell boundaries (solid lines) and source measurement positions (full circles).

The investigation of several alternate weighting schemes showed that with increasing moderation the resulting AAS correction factor was indeed quite sensitive to the details of the approach. This was not unexpected since a highly accurate simulation of a uniform source distribution in a matrix of the extent of an SLB-2 container would require thousands of point-source measurements. For the most dense plywood matrix, the correction factor CF ranged from 6.2 to 18 depending on the weighting approach chosen, but the shell weighting approach resulted in a CF of 12.8. Since the gross matrix correction assumes the matrix is homogeneous and that the activity is uniformly distributed in the matrix, heterogeneity and non-uniformity are accounted for in the Total Measurement Uncertainty (TMU). An additional systematic (method) uncertainty was included in the TMU to account for any uncertainty associated with the volume weighting approach. The TMU analysis results for the SLB-2 container presented elsewhere [5] suggested that a contribution to the TMU due to the AAS calibration weighting may be as high as 50% for the most severe matrix.

In order to reduce the uncertainty associated with the AAS weighting approach and to reliably extend the range of application, a modified weighting approach has been examined using MCNP [6] modeling.

NEW APPROACH FOR AAS CALIBRATION

The new approach for the AAS calibration is still based on the measured data, but the data are now supplemented with MCNP modeling. The new approach for the AAS calibration consisted of the steps given below:

- Creation of the representative MCNP model of the neutron counter loaded with an SLB-2 container and filled with different matrix materials; the model is benchmarked to the Reals rates measured at the 60 locations within each matrix during the calibration process.
- Use of the MCNP model to estimate the Reals rate that could be expected from a neutron source uniformly distributed within each of the matrix materials used during the calibration.
- MCNP modeling of the AAS assembly built into the neutron counter using the measured AAS rates as benchmark data.

The modeling of the AAS perturbation values allowed for the generation of additional 'matrix data points' that could be used to better constrain the AAS calibration curve at the density range for which measured data were not available. For this purpose two additional matrices were modeled to evaluate the impact of the moderating material at intermediate densities.

Benchmarking the SLB-2 model to the measured data

Using an MCNP model of the BNAS counter a point Cf-252 neutron source was modeled at each of the 60 measurement locations within the SLB-2 for all matrices used during the calibration. The physical model and transport solution from MCNP was found to generate a spatial dependence in the Reals rates which broadly tracked the experimental data. For each matrix the ratio between the measured and MCNP modeled rates was determined at each of the measured source locations, and a weighted average of these ratios was used as a gauge of how well the MCNP model represented the measurement. The weighting factor for each source location was a function of the measured Reals rate, so that locations with the lowest measured rates contributed less to the average ratio. The weighted average measured-to-MCNP ratio of the Reals rates is given for each matrix in Table II below.

Table II. Measured-to-MCNP ratios of the Reals rates for different matrix materials

Material	Meas./MCNP	Wt. Ave. Dev.

Reference (Empty)	0.992	0.026
100% polyethylene foam, 0.027 g/cc	0.994	0.068
100% polyethylene foam, 0.064 g/cc	0.891	0.111
100% corrugated cardboard	0.905	0.102
100% plywood	0.832	0.170

The ratios in Table II were used to adjust the counter and the matrix models until the minimum attainable deviation was obtained. The deviations in the observed Reals rates can be attributed to the following factors:

- Differences in material composition between the actual and modeled matrices including moisture content, density, size, and void fraction, especially for the engineered wood products. (For instance plywood was modeled as dry cellulose, yet adhesives were present in the actual material.) The matrices as a whole were made slightly smaller than the container and were made in sections so they could be more easily lifted into place (using straps), so some streaming paths were inevitable.
- Uncertainties in the location of the source placement within the SLB-2 including tolerances in the matrix channels and the placement of the SLB-2 container within the chamber.
- Uncertainties due to the model of the counter including the actual positions of the He-3 proportional counters, variations in the HDPE moderator density, and a simplified homogenized model of the conveyor system.

Once the minimum deviations had been obtained, an MCNP model for each matrix was used to generate the volume average response for a uniformly distributed source as required by the AAS calibration. While the measured-to-MCNP ratios were not used directly in the calibration other than to benchmark the models, the weighted average of the point-to-point deviations were used as the 1-sigma (standard deviation) uncertainty on the MCNP generated uniform volume response. In other words since the average deviation reflects the extent to which the MCNP model does not fully mimic the data, it is used as the uncertainty (over and above typical statistical uncertainties) for the quantity of interest generated purely from the model. Based on an ideal measured-to-MCNP ratio of 1, the uncertainty assigned to the MCNP-generated uniform volume response ranges from 2.6% for the reference (empty) matrix to 17% for the plywood matrix as can be seen from the weighted-average deviations in Table II.

Modeling of the Add-A-Source assembly

The AAS assembly was also modeled in MCNP. The Reals rates were calculated for each of the ten AAS positions located below the SLB-2 container and averaged to produce an AAS rate for a given matrix. The result was then compared with the measured AAS rate during the calibration and the measured-to-MCNP ratio was used as a guide to refining the model. For the different matrices the final measured-to-MCNP ratios for the AAS Reals rates are given in Table III.

Material	Meas./MCNP		
Reference (Empty)	1.09		
100% polyethylene foam, 0.027 g/cc	1.08		
100% polyethylene foam, 0.064 g/cc	0.98		
100% corrugated cardboard	0.98		
100% plywood	0.90		

Table III. Measured-to-MCNP ratios of the Reals rates for different matrix materials

The MCNP generated AAS rates shown in Table III were only used to benchmark the MCNP model. For the AAS calibration the actual measured data obtained during the calibration process were still used for all calibration matrices. MCNP was only used to generate the AAS Real rates for two additional matrices of intermediate densities for which the measured data were not available; namely cellulose with density 0.25 g/cc and cellulose with density 0.35 g/cc. These matrices were chosen so that the perturbation was between that of the most moderating matrix (100% plywood which was also at the weight limit of the container) and the matrix subsequent to this (100% corrugated cardboard). In order to assign uncertainties to the MCNP generated AAS rates, an average of the measured-to-MCNP deviations observed for the cardboard and plywood matrices was used. This gave a $\pm 6\%$ uncertainty in the AAS rate for the MCNP generated matrices which was used in the generation of the AAS calibration curve.

Enhanced AAS calibration curve using MCNP data

The new uniform full-matrix AAS calibration was based on the five measured matrices (including the empty container) as well as the two matrices generated using MCNP. For all the matrices the VWA Reals rates were obtained using MCNP to model a uniformly distributed source. For the AAS rates the measured rates were used for all but the two MCNP generated matrices, because modeling the AAS accurately is highly sensitive to modeling the structure of the floor, conveyor, the steel guide tube, and the relative distances. Uncertainties were assigned as discussed previously. The volume and AAS perturbations determined for each matrix are presented in Table IV. As described previously the uncertainty in the volume perturbation is assigned based on the weighted-average deviations previously shown in Table II, and ranges from 6.9% at the 0.027 g/cc density material to 17% at the highest moderating material of 100% plywood.

Matrix	AAS Perturbation		Volume Perturbation		Fit	
Reference (Empty)	0.00	± 0.002	0.00	± 0.00	0.00	± 0.01
100% polyethylene foam, 0.027 g/cc	0.39	± 0.003	0.58	± 0.04	0.58	± 0.03
100% polyethylene foam, 0.064 g/cc	0.96	± 0.018	1.35	± 0.15	1.47	± 0.08
100% corrugated cardboard	0.93	± 0.008	1.52	± 0.16	1.37	± 0.07
MCNP (Cellulose, 0.25 g/cc)	1.29	± 0.229	3.52	± 0.49	3.55	± 0.46
MCNP (Cellulose, 0.35 g/cc)	1.57	± 0.257	6.66	± 0.93	7.00	± 0.93
100% plywood	2.00	± 0.034	17.16	± 2.96	16.61	± 1.75

Table IV. Add-A-Source calibration results for SLB-2

The data given in the table were fitted to a third order polynomial according to the form presented in Eq. 1 taking into account the uncertainties in both x and y, and the resulting equation for the calibration curve is shown below:

 $y = 2.948 \cdot x - 5.299 \cdot x^2 + 4.003 \cdot x^3 \tag{5}$

Volume perturbation values calculated from the fit given in Eq. 5 are also shown in Table IV along with the associated uncertainty which is based on the fit parameters (and parameter uncertainties and covariances) and the measured AAS perturbation value (x) and uncertainty.

The AAS calibration curve is shown graphically in Fig. 3, along with the calibration curve used in the previous approach. As seen in Fig. 3, above an AAS perturbation of \sim 1.5 the volume perturbation (y in

Eq. 2) obtained from the new approach and consequently the AAS correction factor (CF in Eq. 4), steadily increases. The most significant change is for the plywood matrix which is at the upper limit of application of the AAS correction. This underscores the previous choice of 1.5 as the AAS perturbation value above which measurements are flagged for expert review. Below this value the new approach yields results that are lower than in the previous approach with a reversal taking place once more past the data point with the lowest perturbation value.



Fig.3. Reals Add-a-Source (AAS) calibration curves obtained using two different methods shown with data points from AAS calibration for SLB-2. NB: the AAS correction factor CF = 1 + y, is obtained from 'y' the volume perturbation in this plot.

In practice, since the AAS correction factor (CF = 1 + y) is obtained from the calibration curve (Eq. 5), the uncertainty in CF is also calculated from the fit parameters (and associated uncertainties and covariances) and the measured AAS perturbation value (x) and uncertainty.

Fig. 4. shows the AAS CF over the range of application along with the uncertainty band that would be expected from the fit. The uncertainty in the AAS perturbation value was conservatively assumed to be no less than the highest uncertainty measured during the calibration (i.e. > 0.034\2.00; see Table IV), and was further amplified by a factor of $\sqrt{10}$. This additional factor is applied since during the calibration measurements the count time at each AAS position was 10 minutes, whereas in the typical application the count time at each AAS position is 1 minute. This gives a 5-6% uncertainty on the AAS perturbation value across the entire range. With the additional uncertainties associated with the fit the resulting uncertainty on CF varies from 2% to 11% at the upper end of the application. This uncertainty is then carried through to the TMU and is separate from the additional uncertainty that might be added based on the weighting approach used in the calibration itself.



Fig. 4. AAS Correction Factor CF for the SLB-2 as a function of AAS perturbation. Also shown is the uncertainty band obtained from the fit.

CONCLUSIONS

A new approach to the AAS calibration for large containers has been investigated with an intended application to the SLB-2 container for the Box Neutron Assay System installed at the Savannah River Site. When the traditional AAS calibration approach (originally conceived for 200L drums) was applied to the large SLB-2 containers, the approach was found to be highly sensitive to the weighting scheme applied to the calibration data particularly for the more severely moderating matrices. To account for this sensitivity a systematic uncertainty was introduced to the total measurement uncertainty (TMU) over and above the typical uncertainty contribution obtained through the AAS calibration. At the upper end of the range of application for SLB-2 containers (AAS perturbation of 2.0) this systematic uncertainty was of the order of 50%. The objective of the new approach was to better bound the range of uncertainty, thereby reducing the uncertainty contribution from the AAS calibration to the TMU.

Using the new approach it was found that beyond an AAS perturbation of 1.5 (the typical threshold used to flag expert review), the previous approach underestimated the AAS correction factor with increasing effect for increasingly moderating material. At an AAS perturbation of 2.0, the deviation between the approaches was at the boundary of the uncertainty contribution of 50%. In the new approach the systematic uncertainty contribution to the TMU for the highly moderating plywood matrix can be significantly reduced to ~17% by constraining the new approach to the original calibration data. (For the lower density matrices this uncertainty approaches values on the order of a few percent.) Since this systematic uncertainty is added to the TMU over and above the standard uncertainty obtained for the AAS calibration (2-11% over the range of application), it provides a conservative bound for the various measurement scenarios that may be encountered.

In addition to better bounding the AAS calibration (and consequently reducing the uncertainty contribution to the TMU), the new approach also offers a method to investigate a priori the effect of other possible variations in the measurement geometry such as partially filled containers, heterogeneous

matrices, and Pu distributions. The new approach can also be readily applied as an investigative tool to aid in expert analysis following a measurement.

REFERENCES

- S. Philips, A. Bosko, S. Croft, R.D. McElroy, D. Moscato, R. Mowry, W. Mueller, D. Nakazawa, D. Petroka, R. Venkataraman, M. Villani, B. M. Young, H. Zhu, "An Overview of the Integrated Crate Interrogation System (ICIS) for use at the Savannah River Site", Proceedings of the 34th Annual Waste Management Conference, Phoenix, Arizona (2008).
- 2. D. Davidson, R. McElroy, D. Brochu and M. Villani, "A New 208 Liter Drum Neutron Coincidence Counter with Add-A-Source Matrix Correction", Proceedings of the 36th Annual Meeting on Nuclear Material Management, July 1995, Palm Desert, California, p.960.
- H.O. Menlove, J. Baca, J.M. Pecos, D.R. Davidson, R.D. McElroy, and D.B. Brochu, "HENC Performance Evaluation and Plutonium Calibration", Los Alamos National Laboratory Report, LA-13362-MS, 1997.
- 4. A. Bosko, S. Croft, S. Philips, R.D. McElroy, "The Add-A-Source Matrix Calibration of a Large Neutron Box Counter", Proceedings of the 34th Annual Waste Management Conference, Phoenix, Arizona (2008).
- 5. A. Bosko, S. Philips, S. Croft, R.D. McElroy and M.F. Villani, "Total Measurement Uncertainty Analysis for the Box Neutron Assay System", Proceedings of the 49th Annual Meeting of the Institute of Nuclear Materials Management, Nashville, Tennessee (2008).
- 6. J.F. Breismeister (Ed.), "MCNP-A general Monte Carlo N particle Transport Code Version 4B", Los Alamos National Laboratory Report LA-12625-M, Los Alamos, 1997.