**Model-Based Approaches to Calibration and Certification of Nondestructive Assay Systems
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ABSTRACT

Increasingly, model-based methods founded in physics are employed to calibrate, or extend the calibration of, nondestructive assay (NDA) systems used to characterize materials contaminated with plutonium, uranium, and other radioactive isotopes.   In addition, modeling and mathematical methods are used to establish system parameters, correction factors, and measurement uncertainty components in NDA measurements. The methods combine the characteristics of the measurement configuration (detector structure, object shape and dimensions, source-detector distance and orientation, and collimation), generator knowledge of the waste composition, and known physical properties of the system in order to establish these parameters, factors, and components. These methods offer several important benefits over traditional methods used to test, calibrate, operate, and certify radiation measurement systems. First, instruments can be set up and put into service more quickly and correctly using a model-based approach. Second, calibrations can be performed, tested, and modified with reduced use of plutonium and uranium standards when modeling and mathematical methods are employed. Also, modeled approaches avoid calibration error resulting from heterogeneous source distribution. Moreover, the parameters can be installed quickly and simply in an office setting without the need for the safety, security, and safeguards considerations that are present when standards are used.   Calibrations can also be tailored to fit unique or nonstandard container configurations (for example, overpacked drums), new container types, or specialized matrices. Finally, when modeling methods are applied, correction factors and measurement uncertainty elements can be developed, tested, and validated relatively quickly and without the need for surrogate matrix materials and a large number of test measurements.

A combined gamma and neutron NDA system has recently been moved from Idaho National Laboratory to the TA 55 facility at Los Alamos National Laboratory (LANL). The system will be used to assay transuranic waste packaged in drums. After the move, the system was modified to replace degraded components and improve system performance. Afterwards, it was set up to assay different container types and to extend the calibration range. Model-based approaches were used to adapt the system to its new purpose at LANL.

This report will describe the setup, calibration, and certification of the combined system at LANL Included in this discussion will be the measurement applications and details of the measurement methods. The steps in these processes that involved the use of modeling and mathematical calculations to replace or supplement more traditional methods will be identified and discussed. Finally, an overarching discussion

of the methods and advantages of model-based methods to establish the properties of NDA systems will be presented. We conclude that validated mathematical methods offer a robust, flexible, and mature approach which is technically defensible from first principle physics.

INTRODUCTION

Nondestructive assays of gamma-emitting materials based on modeled calibrations such as the In Situ Object Counting System (ISOCS) method have been in use for more than 15 years. The methods generally provide a user interface that enables users to describe the measurement geometry, physical parameters of the measured object, and shielding considerations; and apply these values to the measured gamma signature. These are then combined with computational algorithms to quantify radioactivity in samples. A combination of Monte Carlo, numerical integration, and ray-tracing methods have been used to compute the mathematical efficiency of the measurement geometry[1-2]. For the ISOCS method, each characterized detector is validated with a series of measurements performed by the manufacturer (Canberra Industries, Inc.) to determine the efficiency. The validation process compares the measurements with detailed MCNP[1] models of the detector, and a spatial set of vacuum efficiencies are calculated and stored within the software. The efficiencies are calculated as a function of gamma-ray energy. Next the algorithms construct the source, geometry, absorber, matrix, absorber, collimator, shielding, and detector model using pre-defined container templates. The technique has been used extensively for nuclear materials accountability, inventory verification, waste sentencing, and other purposes where the bias is well understood. NDA systems calibrated by this method have successfully met the Waste Acceptance Criteria for the WIPP transuranic waste facility and for several low level waste disposal sites.

LANL HENC 3 SYSTEM

The HENC 3 (Figure 1) is a combined neutron and gamma assay system that is used to assay TRU waste generated at the TA 55 facility at LANL. It had been located at the Idaho National Laboratory for several years but was not heavily used there. Prior to delivery at the TA 55 facility, it was refurbished with a new gamma detector Canberra Model 3825), amplifier (Canberra Model DSA 1000), pulser (Canberra Model 1654), and new software (Canberra Genie2000, NDA2000, and Multi Group Analysis). It uses a low energy photon filter that is placed in front of the detector face which is 51 cm from the axial center line of the turntable.

Although ISOCS can be adapted to almost any measurement geometry, it has only been used as part of fixed geometry NDA system for the Los Alamos HENC 3 system. That is, container templates, filters, collimators, and detector-to-container distances specific to the HENC 3 are used as part of the ISOCS model. These geometry parameters are combined with the bulk density range and matrix type of the waste to complete the model and allow the system to quantify the radioactivity in a variety of waste containers. This approach has been used successfully for NDA systems at Savannah River Site (SRS)[3] and at Los Alamos National Laboratory (LANL)[4,5]. The ISOCS model for the HENC 3 was determined for a range of densities from 0.01 g/cc to 2.5 g/cc. Separate calibrations were performed for three approved TRU waste container types (55-gal drum, POC (Pipe Overpack Container), and CCO (Criticality Control Overpack)) for both debris and homogeneous matrix types. The calibration models were based on container designs and site knowledge of expected waste matrix materials and were determined without the use of radioactive sources. The modeling was performed at the Canberra facility

in Meriden, CT. Afterwards, the models were loaded into the system's software at the TA 55 facility and confirmation and testing of the calibration parameters were undertaken. These were performed with NIST-traceable sources over the anticipated activity range. The system will be used to assay TRU waste containing both Weapons Grade (WG) and Heat Source (HS) plutonium that may be mixed with relatively large amounts of ^{241}Am, ^{237}Np, and other processing contaminants. The system's energy calibration was empirically established so that it could be used between 60 keV and 1500 keV. Pileup, rate loss and other experimental factors are well understood in this application and standard techniques are used to correct them.

Figure 1. LANL HENC 3 system.

Confirmation of the calibration was completed by performing six measurements each of several source loadings in a non interferring matrix to confirm that the modeling had been correctly performed and installed. The source loadings ranged from less than 1 g of Pu to 315 g and were performed for the three container types using both WG and HS sources. Next calibration validation measurements were performed with Pu sources in surrogate matrix drums containing combustibles ($\rho = 0.16$ g/cm^3), metals ($\rho = 0.26$ g/cm^3), glass (0.54 g/cc), and cement ($\rho = 1.85$ g/cm^3) matrices. These measurements tested the validity of the modeled efficiency parameters over a range of densities. The confirmation and validation measurements verified that the calibrations were appropriate for the container types, matrices, and plutonium mass range.

Prior to acceptance, the total measurement uncertainty budget was estimated. Determination of the budget was performed using measurements, modeled estimates of the variability due to source and matrix heterogeneity, and estimates based on experience. It includes uncertainty factors such as background, statistical uncertainty in the quantitative and Pu isotopic measurements, uncertainty associated with calibration, source and matrix heterogeneity effects, uncertainty due to detector positioning, and other potential sources of measurement uncertainty. Matrix and source heterogeneity variability are two of the chief uncertainty components and are especially difficult to determine. For this purpose, the ISOCS Uncertainty Estimator[6] (IUE) software was used to aid in the determination of these components in TA 55 TRU waste assays. IUE enables users to combine measurement properties such as container wall thickness, container dimensions, detector-to-source distance, sample matrix composition and uniformity, shielding, source heterogeneity, and other factors to estimate the overall uncertainty of a nondestructive assay (NDA). To determine source heterogeneity uncertainty for the HENC 3, all of the system's parameters and geometry factors were held constant in the ISOCS model but the source position was allowed to vary over the container volume. This established a range of possible values for the measurement results due to source position variability. From this, the uncertainty value was calculated. A similar calculation was used to determine the matrix heterogeneity uncertainty component. IUE was

employed to estimate these TMU components for the three container types and two matrices that are assayed by the HENC 3.

DISCUSSION

A number of benefits result from the use of model-based calibrations. First and most obviously, fewer standards are needed overall. That is, if source-based calibrations are used, primary or secondary sources are needed for calibration and a second set are needed to confirm the calibration. Primary and secondary plutonium sources are very expensive to produce and very few sites have a set for both calibration and confirmation. Using model-based calibrations, only one set, the confirmation sources, is needed to complete the calibration process. An adjunct of this benefit is that reduced security and safeguards measures need to be deployed if fewer sources are used. A second benefit is that the calibrations can be performed relatively quickly. This is because mathematical models of calibrations can be developed in a matter of one, or a few, days whereas calibration with standards generally requires preparation for receipt and movement of standards and several days to perform measurements with high precision. Moreover, if adjustments to the calibration are identified during the confirmation process, they can usually be completed within a few hours when model-based calibrations are used. This can be contrasted to the source-based approach which may require several days to remeasure a standard after a botched calibration measurement. Next, model-based calibrations do not suffer from calibration error resulting from the use of discrete sources to map diffuse plutonium loadings in measured items. During source-based calibrations, discrete plutonium sources are placed in a variety of locations which are typically not representative of where sources may be located in items to be assayed. This leads to an error due to differences between the response of the gamma detector to the discrete calibration sources versus its response to source location(s) inside an item to be measured. With model-based calibration however, the model can be based on homogeneous source distribution or, if source positions are known for specialized waste forms, those positions can be factored into the model. Finally, after the initial modeled calibration of an NDA system has been completed, new matrix types, detectors, filters, and new container calibrations, such as overpacked containers, can be performed quickly because the properties of the system are known in advance, whereas source-based calibrations require a new deployment of source materials and a new set of measurements for every new matrix or container type.

Model-based methods for TMU determination also offer several benefits over other methods. Use of models enables uncertainty estimates on source and matrix heterogeneity to be estimated relatively quickly, typically within two days. If these estimates must be determined from measurements alone, a great number of measurements must be performed so that all of the locations inside a container can be mapped. Also, the uncertainty for new containers and matrix types or for unusual source distributions can be quickly added for each new calibration configuration that is used by the system. This is possible because all of the system parameters and geometry factors have been previously modeled and stored. Finally, comparisons of different uncertainty models can be devised and compared without resorting to a large number of measurements. This can be especially helpful when process or other acceptable knowledge is incomplete and expert judgment must be used to create a variety of plausible variations.

SUMMARY AND CONCLUSIONS

With these benefits in mind, several conclusions on the continued use of model-based approaches to calibration and uncertainty estimations can be offered.   First, modeled calibrations and TMU component estimations should be considered whenever new calibrations of NDA systems are needed. The approach offers important advantages over source based methods as discussed above.   Second, model-based calibration methods should be deliberated in greater detail in national calibration guides and relevant ANSI/ASTM standards. They are founded on first principles physics, and so, provide a robust approach for navigating the complex and expensive landscape of NDA challenges. Although we have focused on gamma systems in this report, similar experience exists for modeling of neutron system calibrations and uncertainties using Monte Carlo codes. Third, modeling and simulation may be the only viable approach to building a comprehensive TMU for NDA systems.

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