ISOCS Waste Measurement Applications at the Oak Ridge National Laboratory–14138

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ABSTRACT

Increasingly, model-based methods have been used to calibrate nondestructive (NDA) systems used to characterize waste contaminated with plutonium, uranium, and other radioactive isotopes. Model-based measurement methods, such as the In Situ Object Counting System (ISOCS), make use of knowledge of the measurement configuration (detector structure; object shape, dimensions, and material; source-detector distance and orientation; and collimation) to establish calibration parameters, interpret data, and quantify measurement uncertainty of measurements of radioactive materials. These methods offer several important benefits over traditional methods used to determine calibration parameters and measurement uncertainty values. First, calibrations can be performed without the use of plutonium and uranium standards when modeling methods are used. In addition, 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, measurement uncertainty can be established relatively quickly and without the need for surrogate matrix materials when modeling methods are used. To date, NDA systems using modeled calibrations to assay transuranic (TRU) waste have employed fixed geometries; that is, fixed physical relationships between the waste container and the detectors. Examples of certified systems with model-based calibrations with fixed calibration geometries for assaying TRU waste include the large box counter at the Savannah River Site and the mobile box/drum counter at Los Alamos National Laboratory.

Model-based measurement approaches have been used for a variety of purposes at Oak Ridge National Laboratory (ORNL) including LLW/TRU sorting of a wide variety of radionuclides and waste streams in 55 gallon and overpack drums, disposition of LLW drums to the Nevada National Security Site (NNSS), near-real-time analysis of high-activity soil samples for mapping of underground radioactive contaminants, and characterization of TRU and LLW constituents in the presence of high levels of Cs-137 in boxed soil wastes for shipment to NNSS. A model-based calibration approach with variable geometry is now being developed for ORNL TRU waste.

This report will discuss several of the model-based measurement approaches used at ORNL. Included in this discussion will be the measurement application, accuracy and precision data, and details of the measurement methods. Then the variable geometry system under development for TRU waste measurements will be described along with information on its intended applications and the ways in which modeling will be used to establish the system's capabilities.

INTRODUCTION

Non destructive assays of gamma-emitting materials based on the In Situ Object Counting System (ISOCS) method have been in use since 1997. The method provides a user interface with a method to describe the measurement geometry, physical parameters, and computational algorithms to quantify radioactivity in samples. A combination of Monte Carlo, numerical integration and ray-tracing methods are used to compute the mathematical efficiency of the measurement geometry[1-2]. To determine the efficiency, each ISOCS-characterized detector is validated with a series of measurements performed by Canberra Industries, Inc. The validation process compares the measurements with detailed MCNP[3] models of the detector, and a spatial set of vacuum efficiencies are calculated and stored within the software. Using pre-defined container templates, the ISOCS algorithms then interpret any source, geometry, absorber, matrix, absorber, collimator, shielding, and detector combination, appropriately modeling the actual response of the particular detector used. The efficiency is calculated as a function of gamma-ray energy. The technique has been used extensively for nuclear materials accountability, inventory verification, waste sentencing, and other purposes.

Although ISOCS can be adapted to almost any measurement geometry, it has also been used as part of fixed geometry NDA systems for TRU waste assays. That is, container-specific templates, filters, collimators, and fixed detector-to-container distances are used as part of the ISOCS model. Several calibrations based on bulk density, matrix type, and container size are then constructed to allow the NDA system to assay a variety of waste forms. This approach has been used successfully for NDA systems at Savannah River Site (SRS)[3] and at Los Alamos National Laboratory (LANL)[4,5]. At SRS, ISOCS models for a variety of densities from 0.05 g/cc to 2.1 g/cc were used to calibrate the gamma component of a Large Box Counter[3]. Separate calibrations were performed for a number of approved TRU waste containers including Standard Waste Boxes (SWB), Standard Large Boxes – 2 (SLB-2), Ten Drum Overpacks (TDOP), and 55-gal drums. All of the calibrations were initially modeled based on container designs and site knowledge of expected waste matrix materials without the use of radioactive sources. After the arrival of the system at SRS, the calibrations were verified with NIST-traceable sources and containers over the anticipated activity range. Two LANL NDA systems have used a similar calibration approach. The LANL SuperHENC[4] combines passive neutron with quantitative gamma ray analyses to determine the total TRU content in Standard Waste Boxes (SWB). The gamma component of the system uses two Broad Energy Germanium (BEGe) detectors located on one side of a mobile platform. SWBs are rotated 180 degrees during a measurement cycle to ameliorate source and matrix heterogeneity effects. The detectors have been characterized at several bulk densities ranging from 0.0 g/cc to 2.5 g/cc in order to assay debris and homogeneous solids waste. Detector positions are fixed with respect to SWB positioning for all of the SuperHENC measurements. The Mobile ISOCS Large Container Counter (MILCC)[5], also at Los Alamos, uses two ISOCS-characterized BEGe detectors to assay TRU waste in 55-gal drums, 85-gal drums, SWBs, and Standard Large Box-2 (SLB-2) containers. ISOCS characterization of the two matched detectors has been performed for several fixed locations to accommodate different activity levels in the containers. Containers are positioned between the two detectors and measured.

Along with ISOCS characterization, all three systems have also employed ISOCS Uncertainty Estimator[6] (IUE) software to aid in the determination of Total Measurement Uncertainty (TMU) in TRU waste assays. IUE enables users to combine uncertainties due to 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). For the three NDA systems described above, IUE has been employed to estimate, in part, the TMU for the various containers and measurement geometries measured by the systems.

In this report, an extension of the use of ISOCS for TRU waste measurements at Oak Ridge National Laboratory (ORNL) will be described. The new system will be very similar to the LANL MILCC system but will be specific to ORNL waste streams and containers. Prior to describing the new system, some existing uses of ISOCS at ORNL will be discussed. Then a detailed discussion of the components and setup of the new system will be presented.

EXISTING ISOCS APPLICATIONS AT ORNL

In the past, ISOCS has been used at ORNL to quantify radioactivity for a variety of projects. ISOCS modeling was used to analyze gamma spectroscopic measurements of 269 drums of ^{232}U/^{233}U-contaminated waste generated in the ORNL Building 3019 Hot Analytical Facility. The drums were first scanned with a handheld NaI detector to check for gamma hotspots, and a hotspot model was determined, as necessary. Variable standoff distances from 17 to 65" were used to keep deadtimes below acceptable limits. U-232 was measured directly. U-233, other uranium isotopes, and Pu-239 were scaled from the measured ^{232}U using scaling factors provided by the customer. Only low concentrations of TRU isotopes were expected. Drums with directly measured or scaled TRU concentrations greater than 100nCi/g were rejected from the waste stream.

ISOCS measurements and modeling were used to provide same-day analyses of TRU and LLW radiological constituents in soil cores taken from the vicinity of the Corehole 8 Tank W-1A at ORNL. The purpose of this characterization project was to generate 3-D maps of the extent of TRU and LLW contamination around the underground tank. Geoprobe dual-tube soil sampling was used to collect 1.125"-diameter soil cores up to two feet long. These cores were immediately brought to two adjacent ISOCS gamma spectroscopy stations for NDA measurements. A typical NDA measurement measured a 12"-long core segment for 15 minutes, and the results were reported within 30 minutes. A range of standoff distances were used. Fast turnaround times were necessary to support the dynamic sampling plan. The primary detected radioactive contaminants were Am-241, Cs-137, Eu-152, Eu-154, and daughters of U-232, U-233, and Th-232. The quantification of Am-241 in the presence of high concentrations of Cs-137 presented some special challenges. Using customized counting geometries and lead shielding it was possible to quantify 45nCi/g of Am-241 in the presence of 21,000nCi/g of Cs-137. Detection limits for Am-241 were typically less than 10 nCi/g. Approximately 300 NDA measurements of soil cores from 64 probe locations were performed.

During the remediation phase of Tank W-1A, approximately 1000 cubic yards of contaminated soils and associated wastes were excavated and placed in lead-shielded B-25 boxes for disposition as LLW to the Nevada National Security Site (NNSS). ISOCS measurements and analyses were chosen in order to achieve direct quantification of the radioactivity in each B-25 box, accelerate completion of the project, minimize the safety risks of direct personnel contact with the contaminated soil, and cut project costs by eliminating sample preparation and off-site shipping charges. Two specialized Low Energy Germanium (LEGe) detectors were mounted in a fixed geometry for high-resolution spectroscopy measurements above each box. Gamma spectra were collected from the LEGe's using LYNX high-throughput digital multichannel analyzers. Gamma spectra from the LEGe detectors were typically acquired for 20 minutes.

TRU and DOT concentrations were calculated to determine whether all shipping and disposal requirements were met. 316 B25 boxes were measured over six months. The measurement throughput was up to eight boxes per day. 309 B25 boxes were shipped to NTSS for disposal. Analysis turnaround times were typically one hour. The 4,000 gallon tank was excavated, cut up, packed into boxes, and measured with NDA. Other wastes generated by the project, including Dry Active Waste, concrete, and metals, were also characterized using ISOCS modeling.

NEW ISOCS TRU WASTE SYSTEM AT ORNL

At ORNL, there are many high activity CH TRU waste drums that must also be assayed. Typically, the high activity results from the decay of Cs-137 to Ba-137m with the emission of a gamma ray at 661.7 keV. Because the activity range is great, an NDA system with sufficient flexibility to both meet WIPP Minimum Detectable Concentration (MDC) requirements for TRU isotopes while keeping dead time within an acceptable limit is needed. Based on experience for similar waste drums at Oak Ridge, three specific combinations of standoff distance and collimation will be used:

- NEAR: 17 in. standoff with 180 degree collimator
- MID: 76 in. standoff with 180 degree collimator
- FAR: 136 in. standoff with 90 degree collimator

In order to certify the new Oak Ridge NDA system to assay TRU waste, several steps must be completed. These include development of a calibration plan; establishment of daily background and performance criteria; completion of, and introduction of, modeling parameters into the ISOCS software; performance of calibration confirmation measurement; completion of calibration verification measurements; determination of Minimum Detectable Activity (MDA); initiation of weekly matrix check baseline program; determination of Total Measurement Uncertainty (TMU) budget; inclusion of the system in an approved measurement comparison program; and an audit of the system by DOE and EPA. Modeling of the calibration parameters will take into account the three detector-to-drum distances, generator knowledge of waste constituents and packaging considerations, generator knowledge of TRU isotope loadings and interferences, and measurement experience with similar waste streams. Verification and confirmation of the calibration will be performed via multiple measurements of weapons grade plutonium standards with certificate values traceable to nationally recognized standards. The system will use two Canberra BE5030 Germanium detectors with Cu/Sn filters over the face of the detectors. The gamma signals will be processed by a LYNX™[1] Digital Signal Analyzer. Signal processing and analysis will be performed with Canberra’s Genie 2000, NDA 2000, and ISOCS software modules whereas plutonium isotopic composition will be determined with both MGA and FRAM software modules. The gamma ray spectrometers are appropriate for direct quantification of the TRU isotopes Pu-238, Pu-239, Pu-240, Am-241, U-233, U-235, -U-238, Cs-137, and Np-237; but will also be used to assay other isotopes in order to quantify the OR Waste. The spectrometers will use a multi-curve efficiency calibration to compensate for variations in matrix density. TRU waste from several Oak Ridge waste streams will be assayed by the system. Initially, the system will only be used to assay 208-liter drums. The density range of the waste will vary from 0.001 g/cc to 2.5 g/cc.

[1] LYNX *is a trademark of Canberra Industries Inc in the United States and/or other countries.*

Calibration modeling was performed at the Canberra facility in Meriden, CT. The models were then loaded into the system's software at the Transuranic Waste Processing Center (TWPC) in Oak Ridge. All confirmation and testing of the calibration parameters were performed at the Oak Ridge facility.

Daily background and quality parameter baseline measurements will be performed first after setup is complete. Then confirmation of the calibration, consisting of six measurements of a non interferring matrix with several source loadings, will be made to confirm that the modeling has been correctly performed Next calibration validation measurements will be performed with Pu sources in matrix drums consisting of a surrogate combustibles matrix ($\rho = 0.29$ g/cm^3), metals matrix ($\rho = 0.42$ g/cm^3), and a sludge matrix ($\rho = 1.13$ g/cm^3). These will test the validity of the modeled efficiency parameters over a range of densities. Minimum detectable activity determinations will next be made for the same surrogate matrix types. These determinations will be made using multiple measurements of the matrix drums and using the Curie[7] approach to MDA determination. Finally, reference measurements for the weekly matrix check measurements will be performed. These measurements check the stability of the matrix correction factors. A list of the certification measurements to be performed is presented in TABLE I.

Prior to final review by EPA and DOE certification officials, the total measurement uncertainty budget will be estimated. Determination of the budget will be performed using measurements, modeled estimates of the variability due to source and matrix inhomogeneity, and estimates based on experience. It will include uncertainty factors such as background, statistical uncertainty in the quantitative and Pu isotopic measurements, uncertainty associated with calibration, source and matrix heterogeneity, uncertainty due to detector positioning, and other potential sources of measurement uncertainty.

Finally, audits will be performed by DOE and EPA representatives to verify compliance with WIPP Waste Acceptance Criteria and EPA regulations.

TABLE I . OR MILCC2 certification measurements.

Requirement	Pu Mass Loading (g)	Drum/Matrix	Detector Configuration	# Replicates
Calibration Confirmation	1	PDP-001 (empty)	NEAR, MID, FAR	6
Calibration Confirmation	30	PDP-001 (empty)	NEAR, MID, FAR	6
Calibration Confirmation	175	PDP-001 (empty)	NEAR, MID, FAR	6
Calibration Verification	10	PDP-003 (combustibles)	NEAR, MID, FAR	3
Calibration Verification	10	PDP-006 (metals)	NEAR, MID, FAR	3
Calibration Verification	10	PDP-005 (sludge)	NEAR, MID, FAR	3
LLD	none	PDP-003 (combustibles)	NEAR, MID, FAR	3
LLD	none	PDP-006 (metals)	NEAR, MID, FAR	3
LLD	none	PDP-005 (sludge)	NEAR, MID, FAR	3

Completion of the certification measurements and issuance of a calibration report are expected to be completed by April 1, 2014. Afterwards, the calibration and TMU reports will be reviewed by CBFO and EPA. In addition, the system will undergo a series of Performance Demonstration Program’s (PDP) blind comparison tests and audits by the COE – Carlsbad field office and the EPA before final certification.

SUMMARY

OR ISOCS will enhance OR TRU waste assay capabilities by making it possible to assay new waste forms and different containers, and it will speed up drum measurements. To date, the equipment has been procured and assembled at the designated measurement station, QC baseline parameters have been established, and confirmation and validation measurements are underway.

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