Non Destructive Assay Box Counter

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ABSTRACT

Many long established US DOE facilities are in possession of poorly documented wastes in a variety of forms spanning a broad range of waste containers, waste matrices and isotopic mixtures. Existing assay technologies such as the SuperHENC can address many of the expected waste configurations. However, there are also expected waste items that fall outside the normal operating range of these exceptional but "standard" systems. Given the inherent difficulties and high costs associated with repackaging even a small number of these containers, an assay solution combining multiple techniques has been studied with the objective of maximizing the likelihood of successfully assaying waste for eventual shipment to WIPP or low level waste disposal.

The Integrated Crate Interrogation System (ICIS) is comprised of a Box Segmented Gamma Scanner (BSGS) and a passive/active neutron counting system called the Super IWAS. These are two physically independent assay systems mounted within separate ISO Containers but connected via Ethernet to allow automated integration of the assay results from the two systems. In operation, the waste containers will first be assayed within the BSGS system to obtain both quantitative gamma-ray assay results and relative isotopic data using well-known algorithms such as the MGA or FRAM. In addition to scanning in front of an array of HRGSs the item is also stepped past a ^{60}Co transmission station on the same line. The operator will then move the container to the neutron assay system. The Super-IWAS concept considered, based on the successful IWAS installations at the AMWTP, provides both high-efficiency passive neutron coincidence analysis and active neutron interrogation using the Differential Die-Away technique (DDA). The prospect of combining all three complementary assay modes to provide a reliable assay result is discussed for realistic waste forms along with the extensive modeling results. Our objective was to devise an NDA solution to this pressing problem that stretched the current state of the practice but which could be implemented with low technical risk using a reasonably sized resource allocation in a predictable and timely fashion. At the conclusion of the study, a design without the DDA capability was selected for construction.

INTRODUCTION

A feasibility study for an integrated waste assay system for the characterization of suspect transuranic (TRU) waste was undertaken by Canberra Industries as part of the U.S. D.O.E. Program Research and Development Announcement (PRDA) No. DE-RA09-03SR22278[1]. The ICIS represents one of several system types examined in that study. The following sections of this paper discuss the performance of the Super-IWAS system for the characterization of wastes within large containers such as the Solid Waste Liner Box (SLB-2) and the Ten Drum Over Pack (TDOP).

The assay of large waste containers is not a new application[2,3,4,5,6,7,8]. The currently deployed box counter technologies have been used with varying degrees of success for a variety of matrix types and measurement conditions. However, they all share important limitations. First, all existing box counter measurements, whether gamma or neutron based, are dependent on waste matrix composition and source distribution. Matrix composition parameters that affect measurements include elemental form, bulk density, presence and distribution of multiple matrix materials, and the concentration of interfering materials (such as neutron moderators and absorbers) in the matrix. Although matrix effects are generally smaller for passive neutron measurements than for gamma-ray or active neutron instruments, all of the existing techniques are susceptible to some extent. Self shielding is another effect that has (provided the moderator content is not excessive) the potential for increasing the bias in active neutron and gamma-ray measurements. Although passive neutron analysis is also less vulnerable to this effect, it suffers from poorer sensitivity and significantly higher background effects. All existing systems are affected by non-uniform distributions of radioactive sources. Non-uniform source distribution, including non-uniform plutonium composition, can bias NDA measurements either high or low.

Several attempts have been made in the recent past to correct biases due to matrix composition uncertainty, self shielding, source distribution effects, and background irregularities in nondestructive measurements. Some of these attempts include random triggering of the accidentals gate to reduce backgrounds in passive neutron measurements; matrix non-uniformity correction algorithms for gamma-ray analysis; neutron imaging and tomography for neutron and gamma-ray analysis, respectively; the 'add-a-source' technique to compensate for matrix effects, etc. In addition, there have been attempts to provide matrix-specific calibrations for each different matrix form that the system is intended to measure. To some extent, all of these approaches have led to improvement in measurement accuracy. However, the improvements have generally been over a limited range of matrix types, under a limited set of conditions, or have required great effort to implement. Most of the correction approaches have also been applied to drum-sized, or smaller, containers. Their usefulness for analysis of boxed waste has not yet been validated.

The ICIS concept attempts to improve the likelihood of successfully characterizing radioactive wastes by integrating the assay results from three distinct assay modes, high efficiency passive neutron counting, active neutron interrogation, and box segmented gamma scanning. The ICIS would consist of two physically separate counters, the Super-IWAS passive active neutron system and the Box Gamma Segmented Gamma Scanner (BSGS). These systems would be based on modifications to existing counter designs and techniques with an automated data integration technique tailored to these large systems.

The Multi-Modal Approach

The ICIS provides three distinct assay modes. The different analysis methods are complementary in that the weaknesses of one assay mode are often times the strengths of another. The ICIS approach provides all the data that can be gathered on the waste container to minimize misclassification of wastes.

Passive neutron counting provides the following benefits:

- The passive neutron assay does not suffer from self shielding or self absorption effects, providing generally more accurate assay results than the HRGS or active neutron assays for higher plutonium and uranium loadings.
- More accurate than active neutron interrogation because it is less sensitive to matrix effects including thermal absorbers and less sensitive to source distributions. Matrix effects in passive neutron are smaller and more predictable, therefore, the errors from matrix effects can be bounded. This feature is necessary for approval of the Total Measurement Uncertainty (TMU) method for WIPP certification.
- Significantly less sensitive to the presence of high-Z material in the waste matrix than HRGS.
- Interference from uranium is small. For passive assay ^{238}U is a weak interference but its effects can be corrected for in the neutron analysis when combined with the gamma-ray assay.
- The total neutron rate is proportional to the total alpha activity that must be reported as part of the characterization requirements for WIPP. While this measurement is subject to the chemical form of the alpha emitter it can be used to bound the sample's alpha activity.
- The ^{240}Pu-effective from the passive measurement can be combined with a gamma isotopic measurement to provide reliable reporting of plutonium and other isotopes.

High resolution gamma spectroscopy provides:

- Measurement of the plutonium, americium and/or uranium isotopic ratios. Standard isotopic analysis codes such as Multi-Group Analysis (MGA) and FRAM code are available and have been approved for isotopic measurements by DOE-CAO during multiple site audits.
- Quantitative measurement of plutonium and uranium for low density matrices. By summing the spectra from all eight detectors, the proposed system can achieve detection levels well below the 60 nCi/g TRU detection level for plutonium wastes.
- Detection levels for U-235 < 0.5 grams for uniform source/matrix distributions for matrix density <1.2 g/cc.
- Direct measurement of other gamma emitters in the waste that are not identified in the isotopic measurement (e.g., ^{244}Cm).
- Basic positional information to improve the accuracy of both the gamma and the neutron measurement.

Active Neutron Interrogation provides:

- Measurement of plutonium in the presence of interfering neutron emitters such as curium and californium.
- Measurement of plutonium in the presence of (alpha, n) neutron interferences
- Confirmation that the container is below fissile limits when the uranium content can not be definitively tied to the passive neutron measurement.
- "Arbitration" when passive neutron and quantitative gamma results disagree.

- Provides rapid screening for LLW wastes that are difficult to assay by means of the passive neutron or gamma modes. .
- Measurement of uranium in the absence of plutonium.

SYSTEM DESIGN

Box Segmented Gamma Scanner

The ICIS gamma station will perform four functions:

[1] Gamma Assay of the crate to detect and quantify gamma emitting nuclides
[2] Plutonium and uranium isotopics measurements, both localized and averaged
[3] Matrix heterogeneity measurement
[4] Source non uniformity measurement

In order to address the four measurement requirements, the gamma station would ideally perform High Resolution Gamma-Ray (HRGS) measurements along the sides, top, and bottom of the container. A system such as this was discussed in the IBIS project report[2]. However, the IBIS design would be costly and not easily accommodated by a mobile housing such as a trailer or standard ISO container.

The BSGS is based on the standard Canberra Gamma-Ray Box Counter shown in Figure 1. The gamma-ray box counter has been designed to assay waste crates and ISO containers but is highly adaptable to other container configurations. The system may also be configured for a variety of other large containers such as 55 gal drums, B-12 boxes, large pipes or tanks, etc. The design of the system is based on a modular ‘building block’ approach so that it can be configured in various ways, depending upon the size of the sample container, the desired detection level, and preferred sample throughput. The system utilizes four collimated High Purity Germanium (HPGe) BEGE detectors arranged on two opposing pillars. The waste container is scanned through the detector array and the gamma-ray spectra recorded as a function of container position and detector number. The BSGS would provide both quantitative values for the activity within the container and plutonium isotopic abundances using the MGA or FRAM codes. Unlike the typical gamma-ray box counter, the BSGS will also include a ^{60}Co transmission assembly to provide data on material fill height and matrix density. The basic design parameters for the BSGS are given in Table 1.

Figure 1. Photograph of a standard Canberra Modular Gamma Box Counter installed at a customer site. In this case this system used two pillars with 2 shielded HPGe detectors on each side of the crate. The crate is scanned through the counter as it moves from one process area to another. The distance between detector and crate is adjustable to allow the system to be used with higher exposure rate containers.

Table 1. BSGS basic design parameters.	
Detector Towers	2
Number HPGe Detectors	4
Detector Type	Broad Energy Germanium (BEGE 2820)
Transmission Assemblies	2
Transmission Source	250 mCi ^{60}Co
Detector Assemblies	2
Detector Type	3x5 NaI
Intensity Ranges	2

ICIS Passive Active Neutron Counting System (Super-IWAS)

The Super-IWAS neutron station will perform the following functions:

[1] Passive Neutron Coincidence Assay of the crate to detect and quantify spontaneously fissioning nuclides (e.g. ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{244}Cm)

[2] Active Neutron Interrogation using the 2nd Generation Differential Die-Away (DDA) technique[11], to detect and quantify the fissile mass (e.g. ^{235}U, ^{239}Pu)

[3] Matrix heterogeneity measurement

[4] Source non uniformity measurement

The Super-IWAS builds on the IWAS[12] approach of integrating active neutron interrogation into an assay cavity optimized for high efficiency passive neutron counting and integrates elements of the Super-HENC[7] and CWAM[3] systems. The neutron detection cavity was designed making extensive use of the Monte Carlo N-Particle Transport code[6] (MCNP™) to evaluate numerous variants of existing counter designs modified for the large containers of interest. Several basic design concepts were studied and modeled so that the final assay system design would provide performance comparable to that of the best performing system currently in use. For example Figure 2 shows the modeled relative passive coincidence performance of several design concepts modified for Passive/Active operation to a metallic waste matrix.

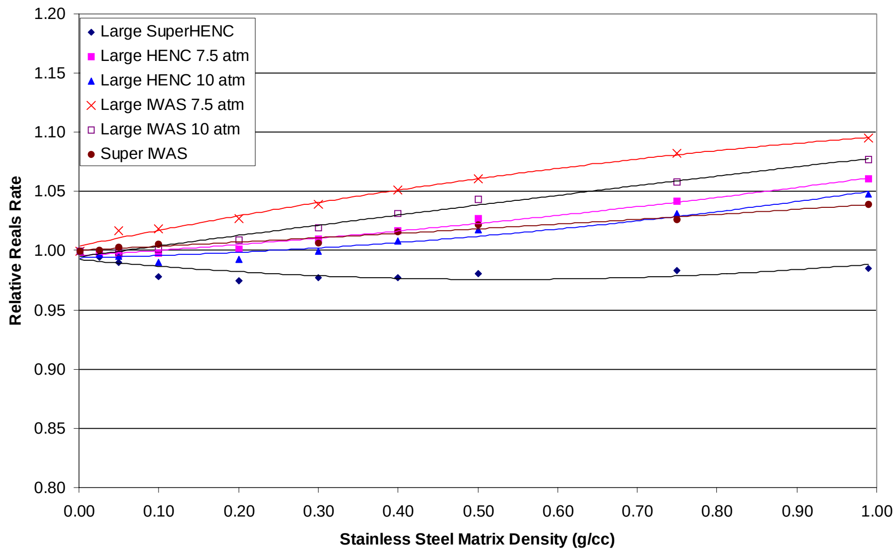

Figure 2. Numerous variants of existing neutron counter designs were examined to provide a neutron detector tube arrangement that optimizes detection efficiency, die-away time and insensitivity to reflective matrices.

The active neutron interrogation is based on the modified 2nd Generation Differential Die-Away technique[12] but to provide more uniform interrogation of the containers, the Super-IWAS incorporates a scanning neutron generator. The general neutron detector arrangement is a cross between the IWAS and Super-HENC systems (Figure 3). As in the IWAS the DDA measurement utilizes a small subset of the over all neutron detectors in order to minimize the amount of cadmium needed for the Fast Neutron Detector Packages (FNDP). However, the ^{3}He proportional tubes are staggered as in the Super-HENC to minimize the neutron energy dependence resulting in relative insensitivity in the coincidence response to the presence of steel within the waste matrix. The basic performance parameters for the Super-IWAS are given in Table 2.

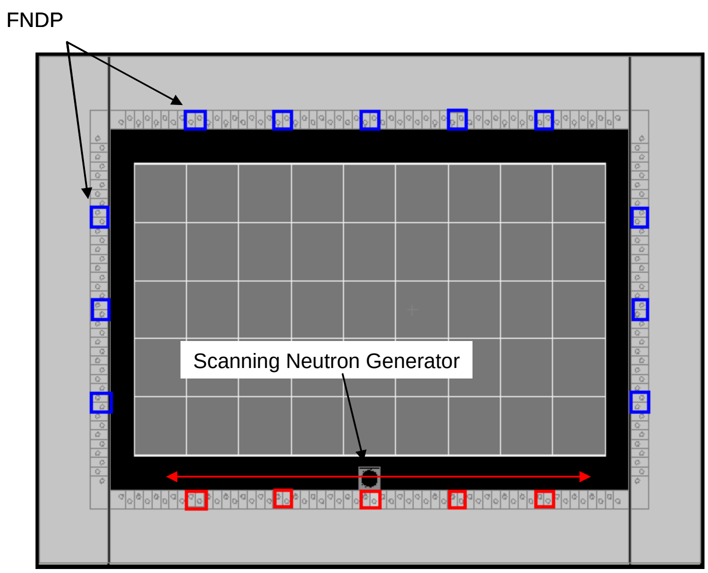

Figure 3. Illustration of the arrangement of the FNDPs about the assay cavity. The FNDPs within the wall nearest the generator (outlined in red) would not be used for quantitative assay as the system was to be operated in a transmission analysis mode. The neutron generator is automatically moved to the predefined interrogation positions.

Table 2. Super IWAS expected performance based on Monte Carlo modeling campaign	
Number He-3 Tubes	320
Passive Detection Efficiency	28.2%
Coincidence Gate Width	128 μs
Pre-Delay	2.5 μs
Doubles Gate Fraction	0.772
Triples Gate Fraction	0.596
Calculated Calibration Coefficient	55.5 R/s/g ^{240}Pu
Local Coincidence Veto correction	0.965
Expected Calibration coefficient	53.6 R/s/g ^{240}Pu
Fast Neutron Detection Efficiency	3.7%
Fast Detector Die-Away Time	39.5 us
Zetatron Pulse FNDP Detection Efficiency:	2.0%
Zetatron Pulse FNDP Die-Away Time:	39.6 μsec
Early Gate Start	1000 μsec
Early Gate Width	2000 μsec
Total Pulses per assay	15000
MDA (^{239}Pu) empty crate	23 mg
Inner Cavity Dimensions	3050 x 1980 x 2010 mm (LxWxH)
HDPE Moderator/Shield Thickness	350 mm

PERFORMANCE MODELING

The largest uncertainty components in the TMU analysis in the assay of large waste containers are typically from non-uniformity in source and matrix distributions. Presently systems utilize multiple measurement or interrogation positions to minimize the effects of these inhomogeneities. The evaluation of the proposed neutron and gamma-ray assay systems was carried out using a variety of mathematical tools including the MCNP[9] (neutron and gamma-ray) and ISOCS[14] (gamma-ray). The models were used to examine the effects of source distribution, matrix composition and fill height on each assay mode.

In passive mode the Super-IWAS utilizes a 4π detection geometry, the Add-A-Source matrix correction, and makes use of a modification of the Hydrogen Correction[10] to minimize the source distribution effects. The Hydrogen correction is essentially an empirical efficiency correction algorithm developed for the Super-HENC. Based on the modeling performed in this study, the modified algorithm will provide a 40% reduction in the standard deviation of the error for a single point source within a moderating waste matrix. A brief summary of the passive measurement performance is presented in Table 3. As seen in the table the fill height has a significant effect on the accuracy of these corrections. It is anticipated that the use of either Radiography results or the gamma-ray transmission measurement could be used to estimate the fill height and allow an improved AAS correction.

Table 3. Results of passive coincidence counting models for the Super-IWAS indicating the relative performance of the system for moderating matrices. The test matrices were various fill heights and densities of High Density polyethylene.

			Average Recovery			Standard Deviation	
Matrix Density	Matrix Description	Source Distribution	Without AAS	With AAS	AAS & Hydrogen Correction	With AAS	AAS & Hydrogen Correction
0.27 g/cc	100% full	Uniform	5%	97%	107%	NA	NA
0.13 g/cc	100% full	Uniform	17%	104%	131%	NA	NA
0.07 g/cc	100% full	Uniform	41%	99%	110%	NA	NA
0.27 g/cc	100% full	30 points	4%	110%	77%	131%	98%
0.13 g/cc	100% full	30 points	16%	104%	106%	56%	30%
0.07 g/cc	100% full	30 points	40%	96%	100%	23%	3%
0.29 g/cc	10% void space	30 points	5%	120%	84%	108%	85%
0.33 g/cc	20% void space	30 points	4%	100%	78%	133%	79%
0.44 g/cc	40% void space	30 points	6%	132%	90%	122%	82%
0.53 g/cc	50% full	30 points	2%	62%	44%	196%	157%
0.53 g/cc	50% full	Uniform	3%	45%	15%	NA	NA
0.95 g/cc	25% full	30 points	2%	38%	39%	192%	147%
0.95 g/cc	25% full	Uniform	2%	15%	31%	NA	NA

The Super-IWAS active interrogation mode implements a scanning neutron generator that interrogates the container at multiple locations along the long axis of the container. The scanning provides a smoothed response and reduction in errors from both the source and matrix non-uniformity. Given the large volume of these containers, a minimum of 5 interrogation positions along both sides of the container are required. Table 4 presents a short comparison of the performance of the scanning active system relative to the passive neutron assay. While the overall results are not as accurate as provided by the passive coincidence mode the technique provides an alternate assay result for those assays not conducive to passive analysis.

Table 4. Modeled response results for the passive and active analyses for 2000 kg iron matrix of varying fill height. Based on the results of 30 runs each for a single point source randomly positioned within the matrix.					
Matrix Density	Fill Height	Average Recovery	Standard Deviation	Minimum/ Average	Maximum / Average
Active Analysis					
1.2 g/cc	25%	55%	24%	0.68	1.69
0.6 g/cc	50%	95%	36%	0.31	1.77
0.3 g/cc	100%	108%	26%	0.59	1.64
Passive Neutron Coincidence Analysis					
1.2 g/cc	25%	102%	4%	0.8	1.08
0.6 g/cc	50%	102%	4%	0.9	1.1
0.3 g/cc	100%	98%	5%	0.9	1.13

BSGS Gamma-Ray System Performance

The standard gamma box counter assumes that the container is filled to a specified height and provides an efficiency correction assuming a uniform source and matrix distribution. The deviation of the real distributions from the uniform case introduces an error into the response. The impact of the container fill height on the standard gamma-ray box counter response for a uniform source distribution was examined by comparing the simulated response from many thousand point sources located at random within a waste matrix of various fill heights. The results of the calculations are presented in Table 5 assuming the system is calibrated for 50% fill height. The BSGS gamma system makes use of a horizontal scan and multiple detectors performing a minimum of 5 measurements for each of the 4 detectors to minimize the source positioning effects. The inhomogeneity in the matrix distribution is addressed in part by transmission measurements. In addition to estimating the density of the waste matrix, the transmission measurement will provide a very crude determination of the fill height for each of the horizontal measurement positions. The approximate graduations provided will be 25%, 50%, and 75%. We will also seek to integrate any available RTR data to provide a more refined fill height determination.

Table 5. Average Recovery for 10,000 randomly placed point sources within a container as a function of Fill Height				
Fill Height	Net Weight of 180 x 180 x260 cm container			
	1053 kg	2106 kg	4212 kg	8424 kg
25%	62%	59%	53%	52%
50%	100%	98%	97%	98%
75%	117%	126%	131%	132%
100%	119%	130%	144%	144%

<u>BSGS Point Source Effects</u>

To illustrate the effect on the reported activity when all the activity is concentrated in a single point source, a series of MCNP simulations were performed. These runs provided the estimated detection efficiency for a point source randomly positioned within waste containers of various shapes and sizes. For example, the detection efficiency was estimated for point source of ^{239}Pu randomly distributed within a waste container and again for a uniform source distribution. The ratio of the efficiencies for the point sources to the efficiency for the uniform source was used to estimate the point source effects. We consider that a single point source is an unlikely case and also examined the efficiency ratios assuming that the activity was distributed between 3 randomly distributed sources. Figure 4 below provides the ratio of the detection efficiency for gamma-ray emitted from a single and three randomly distributed point sources of ^{239}Pu to the detection efficiency for a container of uniformly distributed ^{239}Pu activity within a 1.8 x 1.8 x 3 m box of low density (0.1 g/cc).

These distributions also provide estimates for the worst case and two sigma error limits for point sources within a large container. The two sigma limits correspond to the activity ratio where 95% of the distribution lies within these bounds. Table 6 shows these bounds for point sources of ^{239}Pu within the container. For reference, the net matrix weight of the 0.3 g/cc container is approximately 2100 kg.

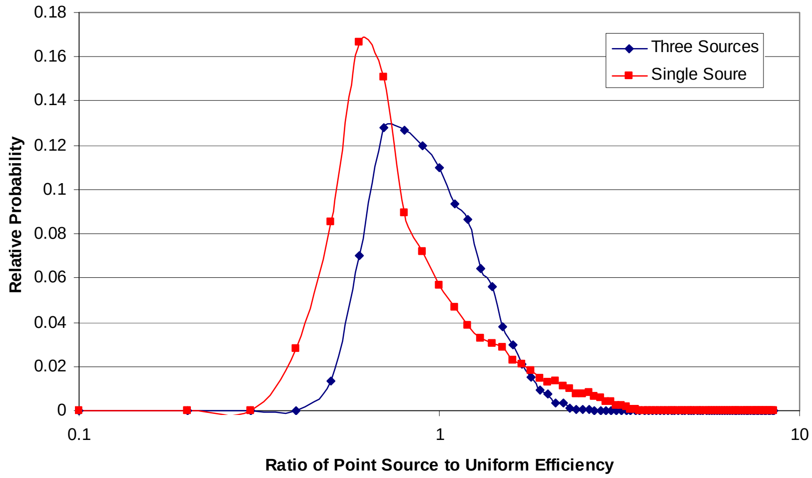

Figure 4. MCNP generated distribution of the ratio of the response from a single and 3 randomly distributed point sources of ^{60}Co activity to the response from a uniform distribution of ^{239}Pu within a 1.8 x 1.8 x 3 m box. Results estimated for a five segment (20 zone) scan. Note that with increasing number of sources the closer the multi-source distribution approaches a Gaussian centered about unity.

Table 6. Bounding limits for relative activity error for a single ^{239}Pu point source.

Density	minimum reported value	2 sigma minimum	2 sigma maximum	maximum reported value
Assuming a single ^{239}Pu point source.				
0.3	0.09	0.26	4.2	5.93
0.5	0.02	0.04	5.7	8.59
1.0	0.001	0.002	8.7	15.6
Assuming three ^{239}Pu point sources.				
0.3	0.16	0.23	2.55	3.98
0.5	0.04	0.09	3.10	6.95
1.0	0.002	0.01	4.55	10.2

DETECTION LEVELS

Detection levels for box counters are generally discussed for uniform source and matrix distributions. The detection level estimates are based on measured data for the BSGS and on modeled performance estimates for the neutron systems.

<u>BSGS Detection Levels</u>

For gamma-ray energies greater than a few hundred keV, the detection levels are dependent on the bulk density of the waste but are only slightly dependent on the elemental composition of the waste matrix. For the 1.8 x 1.8 x 3 meter containers the container will be assayed in 5 horizontal segments. The detection levels for a uniform matrix, uniform source distribution shown in Table 7 are based on measured data for a similar system.

Table 7. Observed MDAs for a 0.3 g/cc container (matrix weight =2100 kg) in 1 hour emission count time for a standard gamma box counter.		
Isotope	Total Act (Bq)	Concentration (Bq/g)
^{60}Co	8.06E+02	1.35E-03
^{137}Cs	1.09E+03	1.83E-03
^{238}U	2.82E+07	4.70E+01
^{239}Pu	3.36E+07	5.63E+01

Neutron System Detection Levels

This system was designed primarily for use within the context of a waste program consistent with the U.S. WIPP facility requirements so that the target detection levels for the system are modest, <100 nCi/g. The passive neutron detection system will make use of the Canberra's Cosmic-Ray Reduction (CRR)[12] algorithms and the Local Coincidence Veto[7,8] method developed by Los Alamos National Labs for use with the Super-HENC to provide low and reproducible detection levels. The following estimates are given for a uniform source and matrix distribution for a system installed near sea-level.

Table 8. Expected detection levels for the Super-IWAS passive neutron coincidence analysis assuming a 2400 second passive measurement.				
	Weapons Grade Pu		Heat Source Pu	
Matrix	mass (g Pu)	TRU Activity (nCi/g)	mass (g Pu)	TRU Activity (nCi/g)
454 kg Fe	0.039	6.6	0.001	33.2
909 kg Fe	0.043	3.6	0.001	18.1
1818 kg Fe	0.051	2.2	0.002	11.3
3636 kg Fe	0.065	1.4	0.002	6.8
454 kg HDPE	0.108	18.3	0.003	90.6
909 kg HDPE	0.212	17.9	0.006	90.5
1909 kg HDPE	0.463	18.6	0.013	94.8

Examination of the active neutron performance indicates that for weapons grade plutonium contained within a moderating matrix, the active interrogation mode can provide lower detection levels than the passive analysis. However, the active mode is not suitable for heat source materials since the dominant plutonium isotope is ^{238}Pu rather than ^{239}Pu. For heat source materials, TRU/LLW sorting must be performed using the gamma-ray or passive neutron analysis.

Table 9. Expected detection levels for the Super-IWAS active neutron interrogation analysis assuming a 600 second active measurement.				
	Weapons Grade Pu		Heat Source Pu	
Matrix	mass (g Pu)	TRU Activity (nCi/g)	mass (g Pu)	TRU Activity (nCi/g)
1818 kg Fe	0.045	1.9	0.140	1052.1
454 kg HDPE	0.015	2.5	0.047	1404.3
909 kg HDPE	0.024	2.0	0.074	1122.2
1818 kg HDPE	0.035	1.4	0.109	779.3

CONCLUSION

We have conducted an extensive review of the complex assay performance requirements, built on our existing systems and drawn on the published experiences of successful counting systems to configure an optimized solution merging proven technologies. An extensive modeling campaign was undertaken to estimate the performance of the ICIS system for both benign and difficult to assay waste containers. The results of the study indicate that in the absence of significant acceptable knowledge of the contents of the waste container, it will be necessary to rely on the expert data reviewer to determine the suitability of each assay mode. The resulting integrated system design provides a significantly greatest probability of success for the characterization of large waste containers.

Ultimately it was determined that the active neutron interrogation subsystem was not required due to cost of ownership, operation and reliability in addition to concerns over self-shielding bias. The assay system manufactured from this study included the BSGS system as described in this document but the Super-IWAS passive active system was replaced by a high efficiency passive coincidence counter.

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