

INTEGRATED WASTE ASSAY SYSTEM (IWAS) AND ANALYSIS ENHANCEMENTS

R.D. McElroy, Jr. and S. Croft
Canberra Industries, Inc.

ABSTRACT

The Integrated Waste Assay System (IWAS) is a hybrid waste assay system developed to provide high throughput and improved data quality for characterization of transuranic wastes contained within 200 liter drums and 320 liter overpacks. The IWAS combines high efficiency passive neutron coincidence counting, active neutron interrogation and high resolution gamma-ray spectroscopy into a single assay cavity. The complementary nature of the multiple assay modes yields a higher success rate than any single assay mode alone allowing the system to accommodate a wide range of source and matrix types. Performance of the individual assay modes and performance of the combined analyses is discussed.

Modifications and enhancements of the traditional analysis algorithms for the passive and active neutron analyses were implemented in the IWAS. Reductions of the passive neutron detection levels of up to 50% have been achieved without the need to increase the neutron detection efficiency. Utilization of the Add-A-Source matrix correction measurement for determination of the active interrogation moderator index provided a means to implement a moderator correction for low Pu mass samples and uranium only samples. The modified algorithms and performance improvements are discussed.

The IWAS analysis makes use of data obtained from the three assay modalities to identify and flag problem waste drums. The analysis makes use of the Total Measurement Uncertainty (TMU) analysis for each assay modality to identify the mode providing the result that most likely represents the actual drum contents. The utility of this approach is discussed.

INTRODUCTION

The Integrated Waste Assay System [1] was designed to quantify plutonium and uranium in 220 liter (55 gallon drums) and 320 liter (85 gallon) over-packs. The IWAS provides passive and active neutron interrogation and quantitative gamma analysis allowing rapid characterization of TRU wastes for proper shipment and disposal. The system offers performance, speed and ease of use while using a fraction of the floor space of three independent systems.

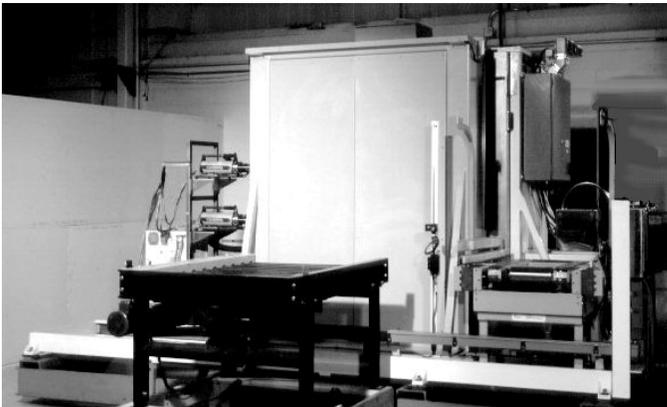


Figure 1. Photograph of the Integrated Waste Assay System.

The IWAS is based on the Canberra High Efficiency Neutron Counter (HENC) [2] design with integrated active neutron interrogation mode using the Differential Die-Away (DDA) [3] analysis and Q2 style High Resolution Gamma-Ray Systems [4]. Results from individual assay modes are combined automatically by the system software to provide the best result for the item. Because all assays are performed by the same system in a single sequence there is no confusion over item identification or modification of drum contents between assays.

The IWAS is designed to be operated as an automated counting system which can process batches of drums, or can be incorporated in a facility process line. The following sections describe the various assay sub-systems, hardware, software and indicate typical performance characteristics.

SYSTEM DESCRIPTION

The primary design goal was to provide the accuracy of passive neutron coincidence counting for high plutonium mass samples with the sensitivity of a multi-detector gamma-ray spectroscopy system. The gamma-ray spectroscopy system provides quantitative analysis for plutonium, uranium, and various fission and activation products, but also provides Multi-Group Analysis (MGA) based plutonium and uranium isotopic abundances. An active neutron interrogation capability based on the DDA technique was added to ensure low detection levels for problem matrices and to provide diagnostic information for difficult to assay wastes (e.g. sludge drums containing with high (α , n) emission rates). The sensitivity target was to achieve a detection level of 10 mg weapons grade plutonium (equivalent to 30 nCi/g for a 0.12 g/cc drum) while achieving a through-put of 22 minutes per drum including sample loading and unloading and source checks. In addition, the system needed to accurately assay samples containing 200 grams or more of plutonium.

The system assay cavity is based on the High Efficiency Neutron Counter (HENC). The HENC body seemed an ideal starting point since its 40 cm thick High Density Polyethylene (HDPE) moderating/shield walls would also serve as a personnel shield against the 14 MeV neutrons emitted by the Zetatron pulsed neutron generator needed for the active neutron analysis. The HENC assay cavity was modeled using MCNP/4B [5] and the basic parameters were compared against the measured data for the three HENC systems already installed to serve as a baseline for the IWAS design. A number of changes were required to the HENC design to add the gamma-ray and DDA capabilities. A summary of the modifications to the HENC is as follows:

- Enlargement of the Assay Cavity to accommodate 83 US gallon over-packs.
- Optimization of the outer HPDE shield thickness to limit the personnel exposure rates from the active neutron interrogation source.
- Addition of HPGe detectors for gamma-ray measurement
- Placement and optimization of the fast-neutron detector and flux-monitor assemblies for DDA analysis.
- Addition of an automated detector shield mechanism to extend useful life of HPGe detectors in presence of Zetatron neutron generator.
- Replacement of the HENC's multi-position Add-A-Source (AAS) matrix correction assembly with a single position AAS assembly to maintain a close coupling with multiple drum sizes.
- Incorporation of a thick stainless steel cavity liner to meet a facility specific contamination barrier requirement.

The final cavity design contains no graphite and uses the HDPE neutron moderator/shield assembly as the external gamma-ray shield for the system. Figure 2 shows a cross section through the assay cavity and illustrates the relative placement of the HPGE detectors and Add-A-Source interrogation position.

Passive Neutron Counting System

The IWAS is a 4π passive neutron counter using ^3He proportional tubes embedded in an HDPE moderator/shield assembly. A Canberra JSR-14 multiplicity shift register is used for the passive neutron coincidence data acquisition. The neutron detection design and performance is very similar to that obtained

from the HENC. The number of tubes and tube lengths increased somewhat to accommodate the larger sample sizes but the same fill gas mixture is used. The overall neutron detection efficiency was lower for this design due in large part to the large mass of steel forming the glove box liner and the inclusion of the cadmium required for the active neutron analysis. However, the loss is offset somewhat by a decrease in the neutron die-away time which allows for greater coincidence gate fractions. The measured performance exceeded the target values and the overall sensitivity of the IWAS was comparable to the HENC.

Table 1. IWAS passive neutron performance parameters.		
Chamber Characteristic	HENC	IWAS
³ He Proportional Tubes:	113	122
³ He partial pressure:	7.5 atm.	7.5 atm.
Efficiency, ²⁴⁰ Pu Spontaneous Fission Neutrons:	31%	27%
Die-Away Time:	52 μs	45 μs
Characteristic Dead time:	111 ns	29.6 ns
Doubles Gate Utilization Fraction:	0.62	0.71
Pre-delay:	4.5 μs	4.5 μs
Gate Width:	128 μs	128 μs
Sensitivity (Reals Rate in cps/g ²⁴⁰ Pu _{eff}):	53.7	46.8
Detection Level (mg ²⁴⁰ Pu _{eff})*:	3.2	4.6

* Nominal 600 second count time, at sea level for a benign waste matrix. Stated for 5% Type I, 5% Type II errors.

Add-A-Source (AAS) Matrix Correction

Neutron Waste assay systems suffer from moderator effects generally leading to a reduction in the neutron detection efficiency. For highly moderating drums such as a 55 gallon (220 liter) sludge drum, the moderator content can reduce the neutron coincidence detection rates by a factor of 5 for a uniform source distribution. The IWAS includes the single position AAS matrix correction technique [6, 7], to correct the measured coincidence rates. The AAS correction provides a means of measuring the impact of the waste matrix on the neutrons emitted within the drum. In practice a small ²⁵²Cf source (about 100,000 n/s) is introduced into the assay cavity with no sample in the counter. The measurement is repeated after the sample is loaded and the results compared. The difference in the measured count rates can be used to correct the measured sample rate. The correction for a uniformly distributed source is accurate to within a few percent. Results from testing with surrogate matrix drums are shown in Figure 3.

The AAS measurement is also used in the active neutron analysis and in correction of the passive neutron background levels. Additional discussion is presented in the following sections.

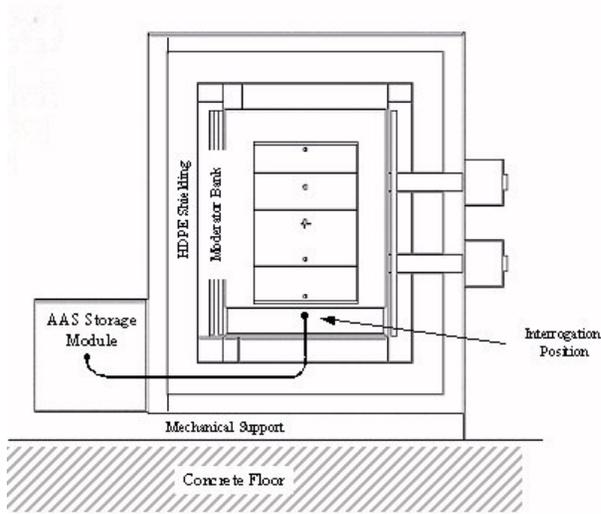


Figure 2. Sketch of the layout of the IWAS assay cavity. The figure shows the AAS source interrogation position in the center of the rotator assembly below the center of the drum.

The AAS mechanism is also used for state of health quality checking of the system. The system uses a special ^{252}Cf source manufactured seeded with ^{137}Cs . Prior to each assay the AAS source is counted in the empty assay cavity as a means of confirming the correct operation of the neutron and gamma-ray sub-systems.

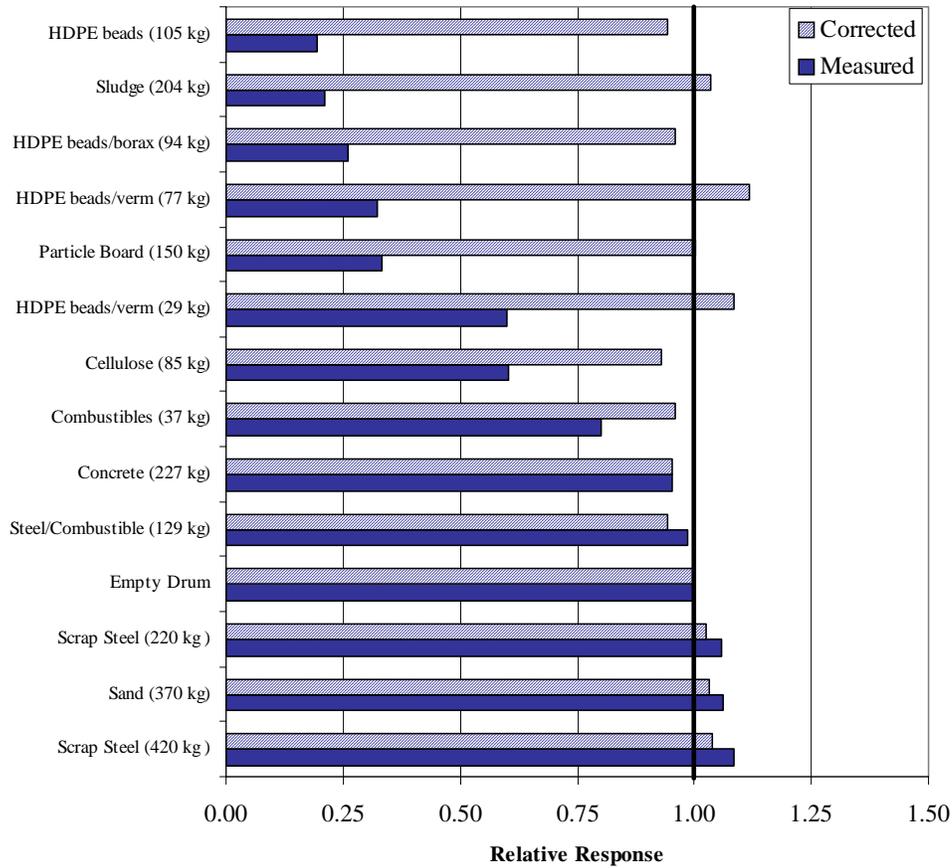


Figure 3. Measured performance of the Add-A-Source correction technique for uniform source and matrix distributions. The solid bars show the relative performance prior to application of the AAS correction.

Passive Neutron Coincidence Background Corrections

The coincidence neutron background has two primary sources, the presence of nearby fission sources and cosmic-ray induced neutron events. The IWAS shielding is sufficient to remove most of the ambient (random, room) neutron background but can not eliminate the neutrons generated by cosmic-rays interacting with the counter's body or with the contents of the item. Drums containing high-Z materials such as lead or steel have an associated cosmic-ray induced coincidence background (or interference) that results in a positive bias in the reported mass if not corrected. Conversely, highly moderating drums (e.g. organic sludge) reduce the cosmic-ray induced coincident neutron detection rate potentially resulting in a negative bias if no correction is applied as shown in Figure 4. The IWAS corrects for these effects using the AAS measurement and neutron multiplicity analysis.

The moderator content of the drum affects the coincident neutron background rate in a similar manner as it affects the fission neutron emission rate. Coincidence rates were measured for a series of surrogate matrix drums including metals, sand, soils, plastics and combustibles. Figure 4 shows the ratio of the measured coincidence rate for these drums relative to an empty drum measurement as a function of the measured AAS correction factor. Failure to correct for this effect results in a negative bias in the reported plutonium mass. The magnitude of this effect depends on the system and overall neutron background rates, but for

typical sludge matrix drum (Add-a-Source correction factor equal to 5) the bias can be as large as 2 g weapons grade plutonium. The IWAS system incorporates a patented correction algorithm to eliminate this bias [8].

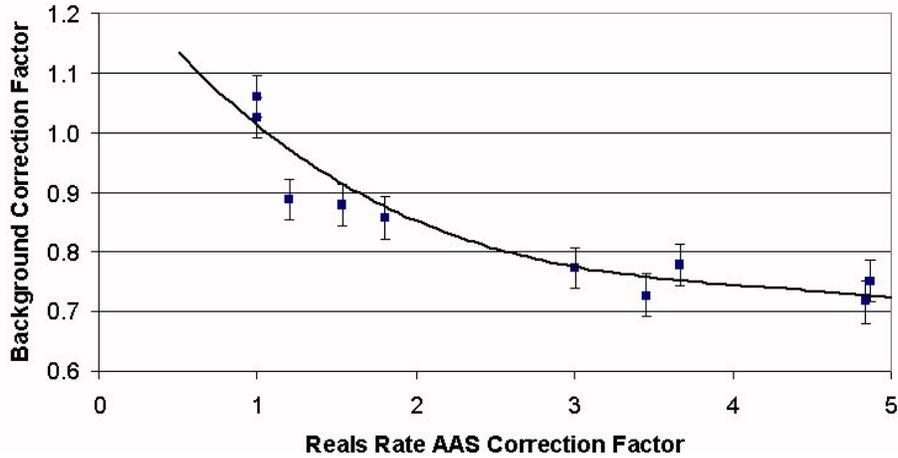


Figure 4. Plot of the neutron coincidence background as a function of AAS correction factor for the drum. The data points show the observed reduction in the empty chamber coincidence rate for a series of drums with increasing moderator content.

High-Z materials such as steel and lead, are known to create a background of coincident neutrons due to the interaction of cosmic-rays and with the waste matrix. At high enough elevations, such as at Los Alamos or Rocky Flats, a 100 kg of steel can result in a 0.5 gram positive bias in the reported plutonium mass (total WG). Methods such as statistical filtering and truncation of the multiplicity histogram have been employed to minimize the effect of these high-Z materials but these methods do not completely eliminate the bias. A correction for the high-Z content of the waste item has been developed based on multiplicity analysis. In a manner similar to the separation between plutonium and curium [9], the analysis takes advantage of the difference in the moments of the multiplicity distribution between plutonium fission and cosmic-ray induced events. The analysis effectively measures the high-Z content of the waste matrix and corrects the reported plutonium mass for its contribution to the coincidence rate.

The application of these background reduction algorithms reduces both the magnitude of the cosmic-ray induced neutron coincidence rates and the fluctuations of these events with time.

Active Neutron Interrogation Mode

The active neutron interrogation mode allows the IWAS to provide lower detection levels than would normally be achievable by a passive neutron assay system in a short counting time. The active neutron analysis uses an intense pulsed neutron source to induce fission in the plutonium and uranium contained within the drum. The neutrons from the induced fission events are detected in a sub-set of the IWAS ³He tubes mounted in cadmium wrapped HDPE packages. The detection efficiency for induced fission neutrons within these Fast Neutron Detector Packages (FNDP) is 2.8%. The arrangement of the FNDP is illustrated in Figure 5. Figure 6 illustrates the relative time response for an empty drum with and without a small uranium standard. The basic performance values for the active mode are provided in Table 2. The active interrogation mode met the design detection limit target of 30 nCi/g. It is interesting to note that unlike most DDA systems, the IWAS active mode uses high pressure, 25.4 mm diameter tubes and does not use a graphite moderating assembly yet the performance provided is equivalent to that of more traditional DDA systems with equivalent fast detector efficiency.

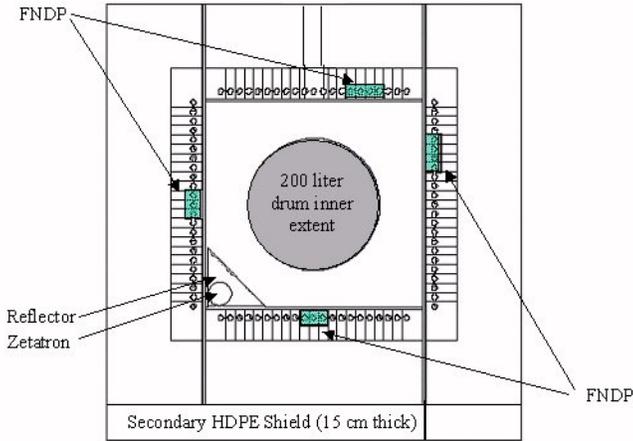


Figure 5. Sketch of the layout of the IWAS active neutron subsystem. The figure shows the location of the four fast Cd covered neutron detection packages (FNDP).

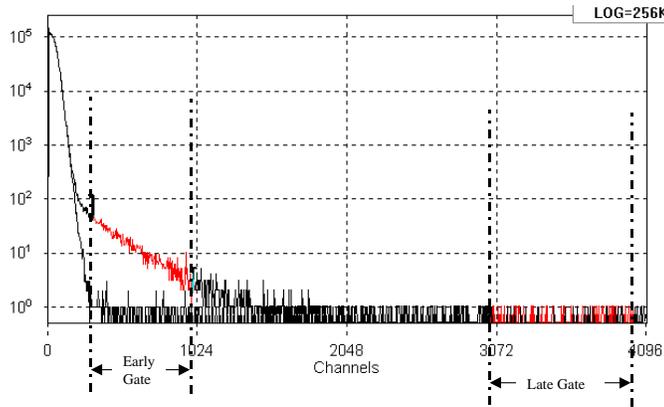


Figure 6. Time response of the Fast Neutron Detector Packages for a zero matrix drum. The upper curve shows the response for a 1 gram ^{235}U source and the lower for a drum with no fissile material. (The time scale is 2 μsec per channel).

Table 2. Active Neutron Interrogation Performance Values	
Induced Fission Neutron Efficiency:	2.8%
Fission Neutron Effective Die-Away Time:	28 μs
Zetatron Pulse FNDP Detection Efficiency:	0.7%
Zetatron Pulse FNDP Die-Away Time:	38.2 μs
Early Gate Start	625 μs
Early Gate Width	1374 μs
Zetatron Repetition Rate	100 Hz
Zetatron Pulses per Assay	12,000
Sensitivity (counts/g $^{239}\text{Pu}/10^8$ neutrons)	38
Background Rate (includes ambient)	0.08 counts/pulse in early gate
MDA (^{239}Pu) empty drum	10 mg ^{239}Pu
MDA (^{239}Pu) 60 kg combustibles matrix	8 mg ^{239}Pu
MDA (^{239}Pu) 220 kg scrap steel matrix	21 mg ^{239}Pu
MDA (^{239}Pu) 150 kg sludge matrix	30 mg ^{239}Pu

Active Neutron Detection – Moderator Index

The IWAS active neutron interrogation method follows the basic approach of the 2nd generation Differential Die-Away method [3] but the treatment of the matrix correction factors has been extended to more accurately correct for U and low Pu mass samples. The traditional approach has been to use the passive neutron count rates to calculate a moderator index and moderator correction factor. Samples containing uranium only or small levels of plutonium do not provide a useful passive neutron count rate. Also, biases can occur due to the shift in average neutron energy for samples with high (α, n) emission rates. However, the IWAS system calculates the moderator index from the measured AAS correction factor allowing the calculation of the moderator correction for these problem sample types. Figure 7 illustrates the effectiveness of the matrix corrections for a High Enriched Uranium (HEU) sample.

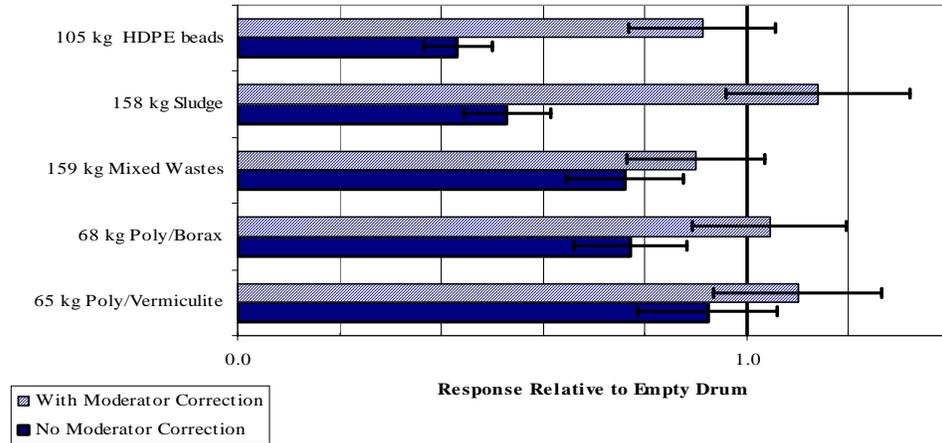


Figure 7. Relative performance of the active neutron matrix correction using the AAS based moderator index for an HEU point source. The results displayed relative to the effective uranium mass (0.28 grams ^{235}U).

High Resolution Gamma-Ray Spectroscopy (HRGS) Measurement

The system's HRGS assembly incorporates the High Purity Germanium (HPGe) detectors to allow quantitative measurement of the gamma-ray emitting isotopes within the drum as well as plutonium isotopic abundance determination. Each HRGS detector is a Canberra BE2820 Broad Energy Germanium (BEGe) detector. The BEGe detector offers the high resolution expected of a planar detector but with the high-energy efficiency of a large coaxial detector. The BEGe gives sufficient efficiency over the full energy range used in this application (60 keV to 1500 keV) while providing suitable resolution for the operation of the MGA plutonium isotopics algorithm in both its low and high-energy modes. Two detectors were chosen for this system rather than the three typically used in the Q2 type system [4] due to the size of the assay cavity and the relatively modest detection level requirements.

The HRGS measurement provides the plutonium isotopic data required to convert the ^{240}Pu effective result from the passive analysis and the fissile mass result from the active neutron measurement into total plutonium mass. However, the HRGS measurement provides the only direct measurement of other radioisotopes that might be present in the waste container (e.g. ^{60}Co or ^{137}Cs). The detection levels observed for the IWAS system with a 660 second acquisition time are shown in Table 4. The ^{239}Pu detection level is less than 10 mg for typical low density drums (< 0.3 g/cc).

The plutonium isotopics measurement capability of the system was limited by the thickness of the thick stainless steel liner needed to meet operational requirements of the facility and by the relatively short assay times. Testing of the system with surrogate waste matrices indicates that MGA analysis can be obtained for plutonium masses of 100 to 200 mg low burn-up plutonium even with these short assay times of 660 seconds.

Table 4. Detection levels for the IWAS HRGS system for a uniform source/matrix distribution and 660 second acquisition time.					
Nuclide	Energy (keV)	Typical LLD (pCi/g)			
		0.1 (g/cc)	0.3 (g/cc)	0.8 (g/cc)	1.8 (g/cc)
¹³⁷ Cs	662	0.72	0.32	0.20	0.16
¹³⁴ Cs	800	0.75	0.35	0.20	0.15
⁶⁰ Co	1173	0.65	0.26	0.17	0.13
²³⁵ U	186	1.60	0.75	0.55	0.50
²³⁸ U	1001	93	39	24	20
²³⁸ U	609	1.30	0.60	0.40	0.30
²³⁹ Pu	414	2.0E+04	9000	8500	5600

Multi-Modal Analysis

The Multi-Modal Analysis refers to the integration of the multiple assay modes to provide a single defensible assay result. This technique takes advantage of the insensitivity in the response of one assay technique to a characteristic of the waste item that may interfere with the response of another assay technique. Table 4 provides a brief listing of the many potential measurement interferences associated with the various assay techniques available [10]. The multi-modal approach was originally developed by following the steps typically taken by a data review expert in the analysis of a waste container. It must be emphasized that this is not an artificial intelligence method. The approach uses well defined logic steps with adjustable threshold parameters. This flow-chart style approach was chosen to facilitate data quality audits by external regulatory agencies.

Table 4. Examples of Interferences by Analysis Mode.	
Gamma-Ray Interferences	<ul style="list-style-type: none"> Other gamma-ray emitting nuclides High bulk density of the matrix Compact High-Z, High mass shield objects Self Shielding of the emitted gamma-rays
Passive Neutron Interferences	<ul style="list-style-type: none"> Other neutron emitters: (e.g. (α,n) reactions , curium, californium) High Moderator Content Extremely non-uniform moderating matrices Multiplication effects from high fissile mass loadings Multiplication effects from low mass loading due to high (α, n) rates (n,2n) neutron reactions (e.g. surface contaminated Beryllium) Neutron poisons in the presence of highly moderating matrix
Active Neutron Interferences	<ul style="list-style-type: none"> High Moderator Content Neutron poisons Extremely non-uniform moderating matrices Other neutron emitters: (but to a lesser extent than passive neutron analysis) Self Shielding from the interrogating flux

The Multi-Modal Analysis first determines the validity of the individual assay mode result based on examination of measured and known values against pre-determined action levels. For example, if ²⁵²Cf is indicated by the gamma-ray measurement, the passive neutron result will be considered invalid (assuming that ²⁵²Cf was not expected). Alternately, should any of the assay results indicate the presence of multiple grams of plutonium, the active neutron measurement would generally be considered invalid due to the

possibility of significant self-shielding of the plutonium from the interrogating neutron flux. Many checks such as these, serve to improve the data quality and increase the likelihood of successfully characterizing the waste container. A partial listing of the data quality checks is provided in Table 5.

If only one of the three assay modes is available and valid then that result is reported. If two or more assay modes are considered valid the mode providing the lowest Total Measurement Uncertainty [12] is selected. At this time there is no attempt to perform a weighted averaging, we attempt only to select the best result when multiple results are available.

For the more complex waste streams such as those containing plutonium and fission/activation products or uranium, the assay result may select results from more than one mode. For example, the plutonium mass value may be determined by passive neutron coincidence counting while the ⁶⁰Co activity would almost always be reported from the quantitative gamma-ray assay.

As a consequence of the analysis the Multi-Modal Analysis software indicates what test conditions caused a given assay mode to be rejected or analysis technique to be selected. These flags are intended to alert the operator of problematic conditions and suspect assay results simplifying the data review process increasing the reliability of the analysis.

Final status of each assay mode	Isotopic Interferences
Fissile Mass	Unexpected Isotopes or Isotopic Mixture
²³⁹ Pu and total U mass per mode	Self Absorption
²³⁵ U and total U mass per mode	Radiography data if available
Passive Neutron Matrix Correction Factor	Count rates and dead-times
Active Neutron Correction Factors	Gamma-ray Peak Widths
Drum weight	Matrix type if available
Acceptable Calibration Range	Total Measurement Uncertainty for each mode

Several IWAS systems have been in use in conjunction with a waste assay program in compliance with the transuranic waste acceptance criteria of the U.S. Waste Isolation Pilot Plant (WIPP) [10]. The systems have been used to assay more than 10,000 drums. The drums assayed consisted of a wide range of matrix types and isotopic distributions. Isotopic mixtures included weapons and heat source plutonium, depleted uranium, and wastes dominated by ²⁴¹Am. Matrix types included but were not limited to: debris, combustibles, metals, sludge, and graphite. Fissile mass loadings in excess of 100 grams were possible. From the analyses of these drums the following general performance data are estimated [12]:

Final Plutonium Assay Results derived from the	
Quantitative Gamma-Ray Analysis:	58%
Passive Neutron Coincidence Analysis:	39%
Active Neutron Interrogation:	3%
Assays Flagged by the software for Expert Review:	50%
Assay Results Modified during Expert Review Activities:	15%
Drums successfully characterized and dispositioned:	>99.9%

For this processing facility no matrix type or isotopic mixture data was available during the initial data analysis due to facility operating restrictions. This resulted in the relatively high rate of assay results flagged for review by a non-destructive assay subject matter specialist. Due to the extraordinarily wide range of waste types, the threshold values in the data quality checks were required to be set much more tightly than optimal. For example, it is known that uranium contained within the sludge matrices tends to be diffuse and that dense concentrations of uranium are rare resulting in a much reduced TMU estimate - particularly so for the gamma analysis. However, the data review specialist knows that the drum is a sludge matrix allowing reanalysis with a different parameter set. The data review software can also accommodate this type of information. If available at the time of assay, matrix specific test parameters could be used significantly reducing the fraction of drums flagged for expert review drums.

CONCLUSION

The Integrated Waste Assay Systems and the Multi-Modal Analysis have been utilized to successfully characterize and disposition TRU waste drums in compliance with the requirements of the WIPP Waste Acceptance Criteria. The combination of multiple analysis techniques allows a greater fraction of drums to be successfully characterized than by the use of a single assay technique alone. As a consequence of the larger volume of data obtained from this more complex assay approach, the Multi-Modal Analysis software was developed to simplify data review activities and improve the data quality. The approach adopted maintains traceability of the data flow and usage as required by regulatory audit.

Enhancements to the analysis software such as the cosmic-ray induced coincidence neutron corrections and AAS based moderator corrections to the active neutron interrogation mode have expanded the useful range of the neutron counting systems.

The Multi-Modal Analysis was devised originally for the accurate and reliable quantification of plutonium bearing wastes but can be applied to other waste streams as well. Preliminary implementations of this expanded analysis are presently under test. The Multi-Modal Analysis is also being extended to other combinations of assay systems such as the HENC plus Segmented Gamma Scanner.

With respect to the active neutron analysis, of the 3% of drums characterized by the active neutron interrogation mode, approximately 1/6th of these drums could have been successfully characterized by the gamma-ray analysis. This implies that of the 10,000 drums assayed only 250 required the use of the active neutron interrogation system. At first glance this small number would seem to argue against the inclusion of the active neutron subsystem, however, this cost must be weighed against the cost of local long term storage of these drums or repackaging the contents.

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