DESIGN AND PERFORMANCE OF THE INTEGRATED WASTE ASSAY SYSTEM (IWAS)

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ABSTRACT

The Integrated Waste Assay System (IWAS) is a hybrid non-destructive assay (NDA) system combining passive neutron coincidence counting, active neutron interrogation, and quantitative high-resolution gamma-ray spectroscopy. The IWAS assay chamber is based on the High Efficiency Neutron Counter (HENC) but modified to incorporate the gamma-ray spectroscopy and active neutron interrogation sub-systems. The system has been optimized to take advantage of the strength of each of the three measurement techniques over the mass range of interest. Design constraints encountered and overcome to combining these three normally incompatible systems into a single instrument are discussed. Results of the numerical modeling used in the design are compared with the measured values.

The integrated approach provides a reliable assay result for a wide range of waste matrix types over the mass range from 10 mg to more than 200 grams of weapons grade plutonium in less than 20 minutes assay time. The combination of the individual assay results is performed automatically by the system software using well defined selection criteria and performance based parameters. Because the analysis is not “intelligent” there is no learned behavior and the analysis is simple to reconstruct from raw data. Calibration results and detection levels are presented for each of the analysis modes illustrating the preferred operating regions for each.

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INTRODUCTION

The radio-isotopic characterization of legacy wastes intended for disposal at the Waste Isolation Pilot Plant (WIPP) must be performed in compliance with the requirements of Waste Acceptance Criteria (WAC)[1]. The radio-isotopic characterization is often complicated by the wide range of waste characteristics and poor quality of the Acceptable Knowledge (AK) available for the waste items. The IWAS system was designed to meet the radio-isotopic characterization requirements of the WAC for a broad range of wastes types (refer to Ref. 2 as an example) that are contained in 55 US gallon drums and 83 US gallon over-packs without prior knowledge of the drum contents. The IWAS integrates passive neutron coincidence counting, quantitative gamma-ray spectroscopy, and active neutron interrogation techniques to provide suitable non-destructive assay capability for as wide a range of waste types as possible. To improve data quality, the system uses a system of warning algorithms based on measured parameters to identify items with characteristics outside of the expected range.

COUNTER DESIGN

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 MGA based plutonium and uranium isotopic abundances. An active neutron interrogation capability based on the Differential Die-Away (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 (α, n) emission rates). The sensitivity target was to achieve a detection level of 10 mg weapons grade plutonium while achieving a throughput of less than 20 minutes including sample loading and unloading. 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)[3]. The HENC body seemed an ideal starting point since its 40 cm thick 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[4] 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:

- Enlarge Assay Cavity to accommodate 83 US gallon over-packs.
- Optimize outer HPDE shield thickness to limit the personnel exposure rates from the active neutron interrogation source.
- Add HPGe detectors for gamma-ray measurement.
- Place and optimize Fast Neutron Detector and flux monitor assemblies for DDA analysis.
- Detector shield mechanism to extend useful of HPGe detectors in presence of Zetatron neutron generator.

- Replace 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.
- Incorporate a thick stainless steel cavity liner to meet a facility specific radiation barrier requirement.

The final cavity design contained no graphite and used the HDPE neutron moderator/shield assembly as the gamma-ray shield for the system. Figure 1 shows a cross section through the assay cavity and illustrates the relative placement of the HPGE detectors and Zetatron neutron source.

Passive Neutron Counting System

The IWAS is a 4π passive neutron counter using ^{3}He 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 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. The measured performance exceeded the target values and the overall sensitivity of the IWAS was comparable to the HENC.

Add-A-Source (AAS) Matrix Correction

Neutron Waste assay systems suffer from moderator effects, the presence of moderating material within the drum can lead to reduction in the neutron detection efficiency. For highly moderating drums such as a 55 gallon sludge drum, the effect can reduce the neutron coincidence detection rates by a factor of five for a uniform source distribution. The IWAS includes the single position AAS matrix correction technique[5,6], 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 ^{252}Cf 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 at the factory are shown in Figure 2.

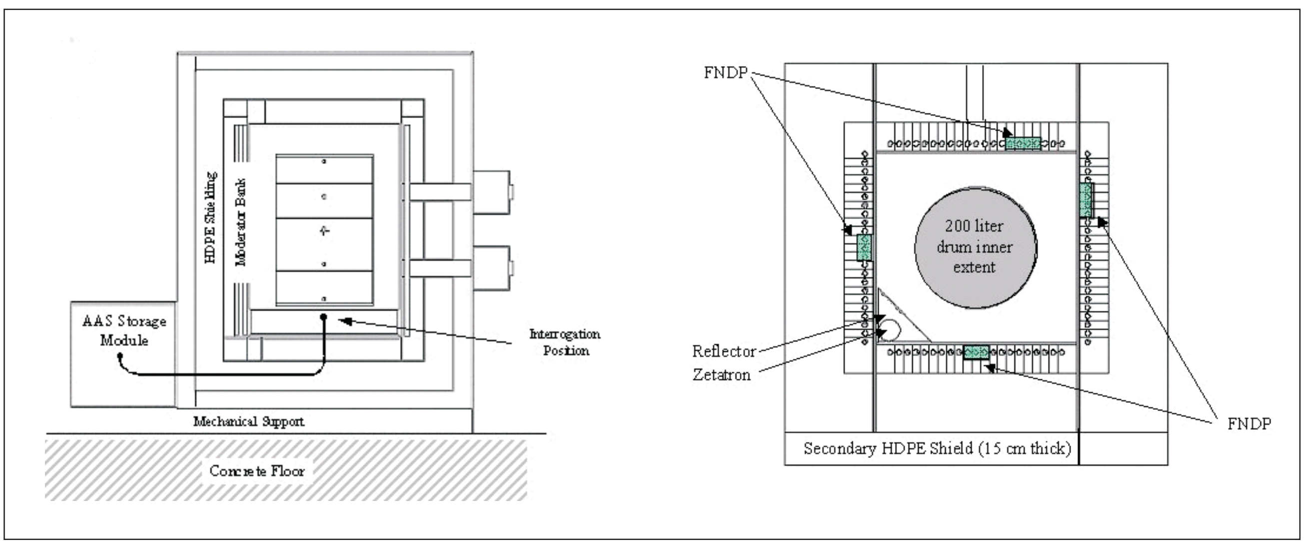

Figure 1.
Sketch of the layout of the IWAS assay cavity. The figure at left shows the AAS source interrogation position in the center of the rotator assembly. The figure at right shows the location of the four fast Cd covered neutron detection packages (FNDP).

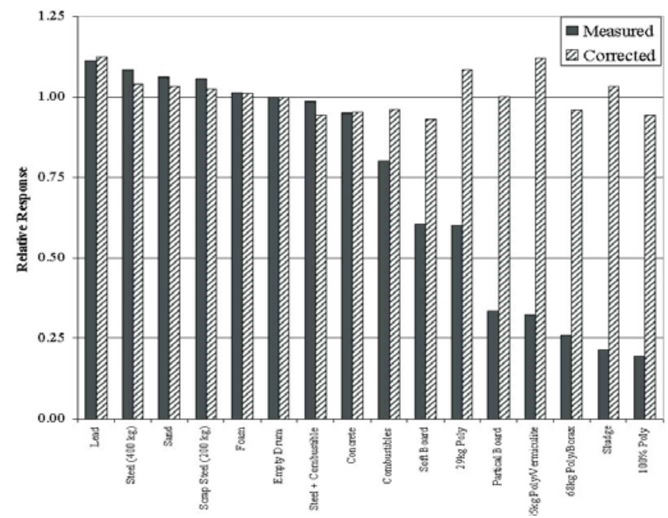

Figure 2.
Measured performance of the Add-A-Source correction technique for uniform source and matrix distributions. The solid bars show the relative performance prior to application of the AAS correction.

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.

Passive Neutron Detection Levels

The passive neutron detection levels for the IWAS system are similar to those provided by the HENC. The detection levels are highly sensitive to the location of the assay facility (note that the cosmic-ray flux increases with altitude) and the construction of the assay building itself (heavy concrete structures reduce the cosmic-ray flux reaching the assay system. Table 2 provides the approximate detection levels for the IWAS systems for weapons grade plutonium with 10 minutes passive acquisition time in a non-interfering matrix. Passive neutron multiplicity counting allows cosmic-ray fluctuations to be suppressed.

Active Neutron Interrogation Sub-System

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 IWAS follows the basic approach of the second generation Differential Die-Away (DDA) method[7] with some detailed changes to specific correction factors. The active neutron analysis uses an intense pulsed neutron source to

Table 1.
Comparison of HENC and IWAS passive neutron performance.

Passive Assay Parameters	IWAS	HENC
^{3}He Proportional Tubes	122	113
^{3}He partial pressure	7.5 atm	7.5 atm
Efficiency, ^{240}Pu, Spontaneous Fission Neutrons	27%	31%
Die-Away Time	45 µs	50 µs
Characteristic Dead time	29.6 ns	111 ns
Doubles Gate Utilization Fraction	0.71	0.59
Pre-delay	4.5 µs	4.5 µs
Gate Width	128 µs	128 µs
Sensitivity (Reals Rate in cps/g $^{240}Pu_{eff}$)	46.8	52

Table 2.
Measured Passive Neutron Coincidence counting detection levels for the IWAS.

Altitude	Building Type	Coincidence Background	Detection Level ($^{240}Pu_{eff}$)
30 meters	Light structure	1.1 Reals/second	4.6 mg
1600 meters	Light structure	3.7 Reals/second	8.3 mg
1600 meters	Heavy concrete structure	0.24 Reals/second	2.2 mg

induce fission in the plutonium and uranium contained within the drum. Neutrons from the induced fission events are detected in a sub-set of the ^{3}He tubes that are mounted in cadmium wrapped HDPE packages. The difference in the characteristic decay time for the source neutrons to thermalize and induce fission and the characteristic decay time for neutrons to be detected gives the technique its sensitivity.

The design of the IWAS system is a balance between the active neutron sensitivity and the passive neutron detection efficiency. The large number of high pressure ^{3}He tubes required for the passive neutron counting tends to poison the interrogating neutron flux from the generator. Optimizing the counter for active interrogation tends to decrease the passive counting efficiency and increase the characteristic die-away time for the counter.

The active neutron detection system consists of four separate Fast Neutron Detector Packages (FNDP) arranged as in Figure 1 above. The FNDP contain either three or four ^{3}He tubes each embedded in cadmium wrapped HDPE moderator blocks. To minimize dead-time losses, each of the 12 ^{3}He tube has its own ^{111}A pre-amp/amplifier discriminator board. A single FNDP is located in each of the vertical walls of the assay cavity as shown in Figure 1. In addition to the FNDP detection assemblies, the active measurement requires two flux monitors. These are low pressure ^{3}He tubes of 15 cm active length used to monitor the neutron output of the generator (cavity flux monitor) and to estimate the neutron flux within the drum (barrel flux monitor). Data is acquired using four multichannel scalers modules recording the FNDP, flux monitor and total neutron count rates as a function of time following the Zetatron pulse. A typical assay is comprised of 12,000 pulses from the generator operating with a repetition rate of 100 Hz. The measured performance parameters for the active mode are given in Table 3.

The active mode detection levels were examined by performing multiple assays on several type of matrix drum. The resulting detection levels are given in Table 4. These are based on 5% Type I and Type II errors.

Table 3.
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
Sensitivity (counts/g ^{239}Pu/10^8 neutrons)	38
Background Rate (includes ambient)	0.08 counts/pulse in early gate
MDA (^{239}Pu) empty drum	9 mg

Table 4.
Measured Limits of Detection for representative matrix types.

Matrix Description	Detection Level
Empty Drum	12 mg ^{239}Pu
50 kg combustibles	8 mg ^{239}Pu
65 kg HDPE beads/vermiculite	11 mg ^{239}Pu
150 kg surrogate sludge (PDP standard)	30 mg ^{239}Pu
220 kg scrap steel	21 mg ^{239}Pu

Gamma-Ray Sub-System

The gamma-ray analysis sub system is based on the Canberra Q^2 system approach8. The Q^2 system uses multiple HPGE detectors mounted in a 4π low background steel shield and a close coupling between the HPGE detectors and waste container to provide very low detection levels. The IWAS design target was 30 nCi/gram of TRU activity in 700 seconds count time. The system also needed to provide plutonium and uranium isotopic measurements. To achieve this goal, two Canberra BE2820 Broad Energy Germanium (BEGe) detectors were selected (refer to Figure 3). The 40 cm plus thickness of HDPE provides an effective gamma-ray shield to minimize background effects and the inadvertent movement of a drum past the system.

The IWAS systems are fully automated and intended to perform many thousand assays per year. Exposure to the active neutron interrogating flux would render the detectors useless after only a few assays if not protected. To extend the life of the detectors, the detectors are automatically retracted from the assay cavity and shielded by thick plugs made from HDPE and cadmium. With this provision, the detector life time is estimated to be from 15,000 to 20,000 thousand active assays.

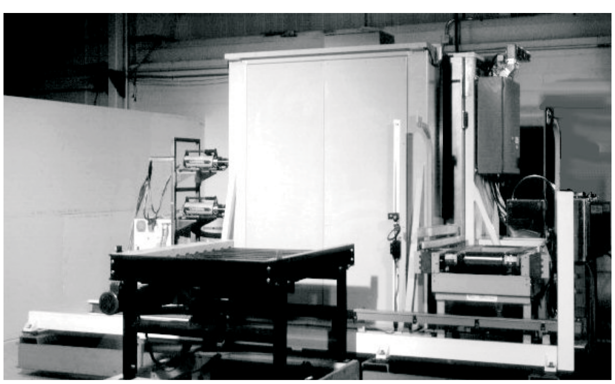

Figure 3.
Photograph of the IWAS system. The retractable HPGe detector assemblies are seen at the left of the counter.

Waste drums containing several Ci of ^{241}Am are expected so the detectors were purposefully shielded to eliminate almost all of the 60 keV gamma-rays. A 0.1 cm of cadmium layer was added to the front face of the BEGe detectors in addition to the 0.8 cm of stainless steel required for the assay cavity liner. Test measurements indicate source loadings of 10 Ci ^{241}Am will result in dead times of less than 30%. Note that the quantification measurements for this arrangement do not use the characteristic 60 keV line from ^{241}Am. Instead a weighted average based on the 125, 662 and 722 keV lines provides an assay result less dependent on the matrix effects than if the 60 keV line were used. Interference corrections are applied if significant quantities of ^{137}Cs are present.

Gamma-Ray Detection Levels

The detection limits for the gamma-ray system are given in Table 5. Detection limits are based on the measured efficiency profiles for the system and measured spectra for surrogate waste matrices. The ^{239}Pu detection level for the 0.1 g/cc waste drum is approximately 10 mg for a source located at the point of least sensitivity.

ANALYSIS ENHANCEMENTS

The IWAS system analysis software includes several enhancements to the traditional analysis techniques. The more important enhancements are listed in the following paragraphs.

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 neutron background but can not eliminate the neutrons generated by cosmic-rays interacting with the counter's body or the contents of the sample. Drums containing lead or steel have an associated cosmic-ray induced

Table 5.
IWAS Gamma System Detection Levels (uniform matrix, 660 second count times).

Nuclide	Energy (keV)	Typical LLD (pCi/g)[a] Density (g/cc)				Worst Case LLD (pCi/g)[b] Density (g/cc)			
		0.1	0.3	0.8	1.8	0.1	0.3	0.8	1.8
^{137}Cs	662	0.72	0.32	0.20	0.16	1.04	0.52	0.60	1.76
^{60}Co	1173	0.65	0.26	0.17	0.13	0.86	0.39	0.39	0.69
^{232}Th	908	2.2	1.0	0.6	0.5	2.9	1.6	1.5	3.2
^{238}U	1001	93	39	24	20	126	60	61	135
^{235}U	185	1.6	0.75	0.55	0.5	2.3	1.4	3.2	21.5
Pu detection levels in nCi/g									
^{239}Pu	414	19.9	9.0	8.5	5.6	30	16	25	124

a. Detection levels for a nominal 600 second passive assay.
b. Estimated detection levels for a non-self attenuating point source at the position of lowest sensitivity in the drum.

coincidence background (or interference) that results in a positive bias in the reported mass if not corrected. Conversely, highly moderating drums (e.g., sludges) tend to attenuate the cosmic-ray induced coincident neutrons potentially resulting in a negative bias. The IWAS can correct for these effects using the AAS correction measurement and multiplicity analysis[9].

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 five) the bias can be as large as 2 g weapons grade plutonium. The IWAS system incorporates a patented correction algorithm to eliminate this bias[9].

High-Z materials such as steel and lead, 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. Methods such as

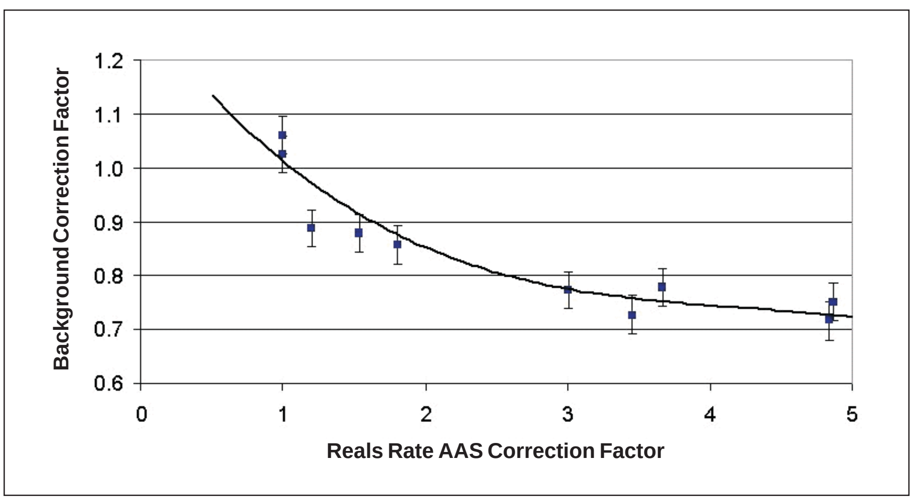

Figure 4.
AAS based background correction factor eliminates the negative bias that occurs for highly moderating waste matrices.

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[10], the analysis takes advantage of the difference in the moments of the multiplicity distribution between plutonium and cosmic-ray induced events. The analysis effectively measures the high-Z content of the waste matrix and corrects the reported plutonium mass.

Active Neutron Detection – Moderator Index

The IWAS active neutron interrogation method follows the basic approach of the second generation Differential Die-Away method[8] but the treatment of the matrix correction factors has been extended to more accurately correct for uranium 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, this system calculates the moderator index from the measured Add-A-Source correction factor allowing the calculation of the moderator correction for these problem sample types. Figure 5 shows the effectiveness of the matrix corrections for a High Enriched Uranium (HEU) sample.

Total Measurement Uncertainty

While it is still quite common for the Total Measurement Uncertainty (TMU) to be calculated in a post assay analysis, or simply assigned on a waste stream basis, the IWAS approach calculates the TMU for each of the analysis modes for each drum. The TMU calculation is based on measured parameters, such as matrix correction values, drum weight, count times, etc. and the result is then used in determination of the suitability of the assay result for disposition of the drum. All of the information required for processing is handled by the NDA 2000 software.

INTEGRATED APPROACH

The physical integration approach of the IWAS system was driven primarily by a desire to limit floor space requirements and improve throughput. Without the need to load/unload/transport the drum between instruments, material handling is minimized and there is greater time available for assay. Because all measurements are made in a single chamber there is little likelihood that the gamma-ray results from one drum will be integrated with the neutron results of another, so data quality is improved.

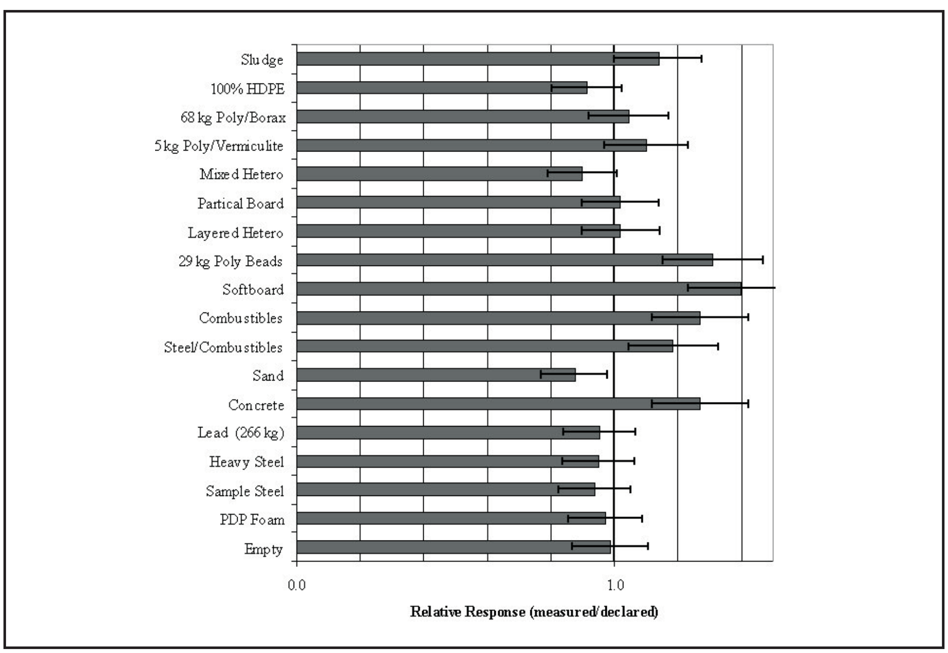

Figure 5.
Relative performance of the active neutron matrix correction using the AAS based moderator index for an HEU point source. The results displayed relative to the effective uranium mass (0.28 grams ^{235}U).

The IWAS integrates not only the multiple assay techniques into a single measurement chamber it also provides a fully integrated data analysis. The process is not based an Artificial Intelligence approach, but instead the software attempts to emulate the data review steps that would be followed by an NDA specialist. The analysis software integrates the results from the passive neutron, active neutron and gamma-ray analysis measurements and AK using a well defined set of logic steps to choose the "best" assay result. The observed count rates, dead times, matrix correction factors, plutonium isotopic abundances, etc., are compared against a set of action levels to determine first the acceptability of each analysis method, and then to select the best result from the valid methods based on a well defined hierarchy. For example, should the drum weight of the drum exceed the alarm point, the quantitative gamma-ray result is considered invalid and the assay report is flagged for review by an NDA expert. Should multiple techniques, say quantitative gamma analysis and active neutron both return acceptable results, the technique that results in the lowest TMU is selected (there is no attempt to average the results from two or more techniques). Additionally, if two or more results are considered valid, the results are compared and if they do not agree within the TMU values, the results are considered suspect. The layered consistency checks help to ensure that problem drums are identified prior to review by the NDA personnel and minimizing the chances for mischaracterization.

SUMMARY

The IWAS system provides NDA results using the three most commonly used analysis techniques integrated both physically and analytically. New analytical techniques have been developed to improve the quality of data available from each of the assay techniques and to lower the sensitivity and TMU available as individual measurements. The automated combination of results provides improved accuracy and reliability of the final assay values. Four systems are in or are about to enter service. The first two have successfully passed round nine of the PDP trials.

REFERENCES

1. "Contact-Handled Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant", DOE/WIPP-02-3122, July 25, 2002.
2. "Waste Description Information for Transuranic-Contaminated Wastes Stored at the Idaho National Engineering Laboratory" INEL-95/0412 Appendix A and Appendix B, December 1995.
3. H. Menlove, et. al. "HENC Performance Evaluation and Plutonium Calibration", LA-13362-MS, October, 1997.
4. Judith F. Briesmesiter, LA-12625-M, Version 4B, "MCNPTM – A General Monte Carlo N-Particle Transport Code Version 4B", March 1997.
5. H. Menlove, et. al., "WDAS Operation Manual, Including the Add-A-Source Function", LA-12292-M, April, 1992.
6. H. Menlove, et. al., "NBC Operation Manual, Including the Multi-position Add-A-Source Function". LA-12737-M.
7. J. T. Caldwell, "Calibration, Matrix Corrections, and Verification Activities with the Los Alamos Second Generation Combined Passive and Active Neutron Assay Systems", Document Q2-TN-85-102, May 1985.
8. F. Bronson, "Q2 – A Very Low Level Quantitative and Qualitative Waste Assay and Release Certification", Proceedings of Waste Management '90, Tucson, Arizona, February, 1990.
9. R.D. McElroy, et. al., "Method for reduction in the interference of cosmic ray-induced neutron events in passive neutron coincidence and multiplicity counting", US Patent #6,509,563.
10. P. Rinard et. al. "Applications of Curium Measurements for safeguarding at Large-scale Reprocessing Plants", ESARDA 19th Annual Symposium on Safeguards and Nuclear Material Management Proceedings (ESARDA 28; EUR 17665 EN), 1997 PP. 221-5.