IPAN™/GEA ACTIVE NEUTRON ASSAY: FINDING THE PLUTONIUM LUMP

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ABSTRACT

Self-shielding effects from large particles present difficulties for active neutron and gamma systems. If Pu "lumps" are present, these systems can only "see" a fraction of the true mass, causing an underestimate of the Pu. BNFL Instruments has developed a new algorithm that allows the large particle presence to be recognized and quantified. This algorithm has been incorporated into BNFL Instruments' Mobile Assay System (MAS) and utilizes the active portion of the imaging passive/active neutron (IPAN™) measurement and gamma energy analysis (GEA). In October 1998, BNFL Instruments' IPAN™/GEA MAS completed Cycle 5A of the Performance Demonstration Program (PDP) for the Waste Isolation Pilot Plant. As part of Cycle 5, large particle PDP standards were created to test system capabilities to properly quantify Pu that may exist in this form throughout the DOE complex. The BNFL Instruments lump correction algorithm was tested for the first time during this cycle. The method produced results consistent with the activity loading of the drum of 0.321 Ci passing both the precision and accuracy requirements. The correction factor for the PDP drum, produced by this new method, indicated that the true mass was four times greater than the active neutron measurement alone indicated.

INTRODUCTION

In October of 1998, BNFL Instruments' Mobile Assay System (MAS) participated in Cycle 5A of the Performance Demonstration Program (PDP) for the Waste Isolation Pilot Plant (WIPP). As part of this cycle, PDP standards were constructed containing large particles or "lumps" of Pu. These lumps were expected to be greater than 100 microns in diameter with significant self-shielding properties.

The MAS is a comprehensive system that utilizes several measurement techniques. ^{3}He tubes collect neutron data for imaging passive/active neutron (IPAN™) analysis while a High Purity Germanium (HPGe) detector gathers information for gamma-ray energy analysis (GEA). Separately, both techniques have become widely used and are proven methods.

However, large particles cause well-known difficulties in the assay of waste. For gamma measurement techniques, self-absorption effects cause differentials in emissions at varying energies

for a single isotope. These effects manifest themselves in much the same way as attenuation effects from the surrounding matrix material. This makes it difficult to distinguish between the attenuating properties of a waste drum and the self-absorption properties, and therefore it is difficult to correct for the underestimate that results. For active neutron techniques, interrogation neutrons are unable to penetrate to the interior of the fissile material resulting in self-shielding. The result is that only the outside surface of the particle is interrogated and an underestimate of the quantity of Pu present occurs. Although attenuation effects on gamma measurements can be quantified, reducing the difficulty in identifying and quantifying self-absorption properties, there is no known way of identifying the presence of large particles using active neutron techniques. Passive neutron techniques do not suffer from these same difficulties. However, these techniques are limited by their sensitivity, and often cannot assay quantities of material that may exist in large particle form.

BNFL Instruments has developed a new technique that allows the presence of large Pu particles to be identified and self-shielding effects to be quantified. The proprietary Lump Correction Algorithm (LCA) utilizes information from acquired gamma spectra as well as imaging information from the active neutron measurement to produce several correction factors. These correction factors are applied to the isotopics analysis and the active neutron measurement in order to remove inaccuracies in isotopic determination and reduce the potential for low bias in the active neutron measurement.

THE MAS IPAN^{TM}/GEA SYSTEM

The MAS developed by BNFL Instruments, fig. 1, performs three separate measurements that are integrated into a single, comprehensive, characterization of drummed waste. Two of these are based on neutron measurement techniques, while the third is based on gamma measurement techniques. Matrix corrections are made based on the absorbing and moderating characteristics of the waste. Absorption effects are measured using a barrel flux monitor (BFM). As the amount of absorbing material increases, the interrogating flux measured by the BFM decreases. The ratio of the BFM to the flux measured by the moderator assembly flux monitor provides information on the absorbing characteristics of the matrix. Epithermal neutron measurements provide information about the moderating effects of a matrix. The count rate of epithermal neutrons exiting the side of the drum opposite the neutron generator decreases with increasing moderator effects.

Once a drum has been loaded onto the conveyor system, it is automatically moved into the cavity. During the passive neutron measurements, ^{3}He detectors, in conjunction with a programmable multi-channel coincidence module (PMCCM), are used to perform neutron coincidence analysis to provide a quantification of the $^{240}Pu_{eff}$ present in the waste. In addition, imaging data is obtained from each of the detectors as the drum is rotated in sixteen 22.5 degree increments. The imaging allows source position corrections that increase the accuracy of the assay.

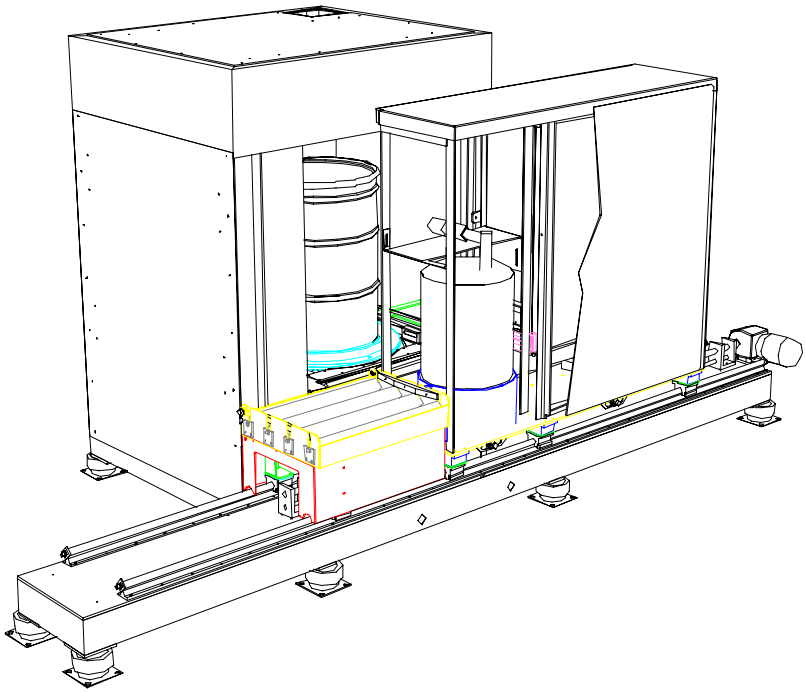

Figure 1. BI Mobile Assay IPAN^{TM}/GEA Drum System

The active neutron measurement, also performed using the imaging ^{3}He detectors, is based on the differential die-away technique (DDT). Interrogating neutrons from the neutron generator, housed in the moderating assembly (MA), are thermalized in the MA and induce fission reactions in isotopes such as ^{239}Pu and ^{235}U contained in the waste drum. The distinct die-away times of the interrogating flux and the induced fission signal allows for a quantification of the fission isotopes.[2] Because the drum is rotated through 360 degrees, the container is evenly interrogated with the thermal flux from the MA.

In addition to the ^{3}He detectors, the MAS also houses an HPGe detector located in the door as seen in Fig. 1. The HPGe is moved into an optimum position for the passive gamma measurement. It remains fixed as the drum is rotated through 360 degrees over a typical time period of 200 seconds. The HPGe analysis software is currently configured to identify and quantify 33 gamma emitting isotopes including ^{239}Pu, ^{235}U, ^{60}Co, and ^{137}Cs using count rates and the BI patented efficiency-times-attenuation/isotopic correlation (ETA/IC) method carried out during expert analysis.

THE LUMP CORRECTION ALGORITHM

Although large particles of Pu are not expected to exist in waste, often the reality is that some such materials go unidentified before packaging. Because self-shielding and self-absorption properties always result in an underestimate, a method was needed to first, identify the presence of large particles, and second, correct assay values for their effects. Previously, these effects were included in total measurement uncertainty (TMU) analysis. Exact quantification of the effect on the TMU is

difficult, however due to lack of access to Pu sources and self-shielding Pu sources in the private sector. The advent of PDP cycle 5A and the creation of large particle Pu standards therefore offered a unique opportunity.

In an effort to reduce or remove the self-shielding contributions from the TMU and provide a more accurate assay, BNFL Instruments developed the LCA. Because the development had to be completed before cycle 5A, BI employed Monte Carlo (MCNP) modeling techniques in conjunction with real waste data taken for approximately 600 drums at Lawrence Livermore National Labs (LLNL). This data was searched for possible large particles that would allow for initial testing of the LCA prior to cycle 5A. Three drums were identified in the LLNL data as likely candidates for large particles.

Identification of the possibility of a Pu lump is based on the gamma spectra acquired during the gamma energy analysis (GEA). In general, materials will selectively absorb lower energy gamma rays more than higher energy gamma rays. This effect occurs whether the absorbing material is Pu or the surrounding matrix. Since Pu239 emits gamma rays at energies ranging from low energies to high energies, it is possible to compare these emissions. A threshold value for the ratio of the higher energies to the lower energies can therefore be used to indicate that large particles of Pu may be present in the waste drum.

Once the possibility of Pu lumps has been identified, the LCA can be employed. The active neutron measurement can be affected by a variety of conditions including particle size, particle geometry, particle location and isotopes present. MCNP modeling can be used to quantify these effects and relate these effects to the self-absorption effects on the gamma measurement. Due to the unique nature of BI's patented ETA/IC gamma energy analysis and BI's proprietary active neutron imaging analysis, it is then possible to correct both the gamma isotopic analysis and the active neutron measurement.

The ETA method developed by BI has been designed to "precorrect" all measured gamma line intensities for their in situ "efficiency-times-attenuation" factors prior to calculating actual gamma line ratios. This BNFL Instruments proprietary method is an iterative process which "fits" the spectral data to produce a relative quantification of the isotopes detected. Additional corrections are added when a Pu lump has been identified.

The active neutron imaging method developed by BNFL Instruments has been designed to correct measured neutron emissions for positional effects prior to calculating the fissile content of the waste drum. Additional corrections are added when a Pu lump has been identified.

PERFORMANCE RESULTS

The LCA has been applied to the three LLNL real waste drums as well as the PDP cycle 5A test drum. The LLNL waste drum results are listed in Table 1. Although it is unknown whether these real waste drums contain large particle sources, comparison of the passive neutron, active neutron and gamma analysis provides strong evidence that they do. Because the Pu quantities present in

two of the drums are within the passive neutron measurement range, we can compare the corrected results with the passive results. In both cases, the corrected active measurement agreed within error with the passive measurement while the uncorrected active result was well outside error considerations.

Table 1 LLNL Results for Self-Shielding Pu Particles

DRUM ID	APPROX. DRUM GRADE	MATRIX	SELF-SHIELDING FACTOR	UNCORRECT TOTAL PU (ACTIVE)	CORRECTED TOTAL PU (ACTIVE)	TOTAL PU (PASSIVE)
980211-8	WGPu	Dry Comb.	4.28	0.51 g	2.2 g	Below Detection Limit
980331-5	WGPu	Dry Comb.	5.95	9.65 g	56.2 g	45.33 g
980521-7	WGPu	Dry Comb.	3.54	10.4 g	36.84 g	38.81 g

The results from the LLNL drums provided supporting evidence for the functionality of the LCA, but it was necessary to obtain well documented, traceable sources in order to complete final verification of the new algorithm. Because self-shielding Pu sources were not available commercially, this final verification was completed when the MAS completed the blind PDP test with large particle Pu standards at LLNL. The results are presented in Table 2.

Table 2 PDP Cycle 5A Comparative Measurement Results

RUN ID-REPLICATES	DRUM GRADE	MATRIX	SELF-SHIELD. FACTOR	UNCORRECT TOTAL PU (ACTIVE)	CORRECTED TOTAL PU (ACTIVE)	TOTAL PU (PASSIVE)
981029A-5	WGPu	PDP Dry Comb.	4.06	1.3 g	5.27 g	4.47 g
981029A-6	WGPu	PDP Dry Comb.	3.75	1.28 g	4.79 g	4.69 g
981029A-7	WGPu	PDP Dry Comb.	3.94	1.30 g	5.14 g	5.02 g
981029A-8	WGPu	PDP Dry Comb.	4.18	1.31 g	5.47 g	4.12 g
981029A-9	WGPu	PDP Dry Comb.	4.56	1.28 g	5.84 g	5.36 g
981029A-10	WGPu	PDP Dry Comb.	3.69	1.29 g	4.76 g	4.63 g

In the case of the PDP large particle Pu standards, the Pu mass is within the passive range, so a direct comparison is possible. All corrected active results and passive results are well within errors for final quantification. Adding in the gamma analysis data, the final PDP cycle 5a results are listed in Table 3.

Table 3 PDP Cycle 5A Final Results

RUN ID-REPLICATES	TOTAL α-ACTIVITY (MEASURED)	TOTAL α-ACTIVITY (ACTUAL)	ACCURACY (% TAG)	ACCURACY PASS CRITERIA
981029A-5	0.427 Ci	0.321 Ci	133%	40-190 %
981029A-6	0.41 Ci	0.321 Ci	128%	40-190 %
981029A-7	0.401 Ci	0.321 Ci	125%	40-190 %
981029A-8	0.447 Ci	0.321 Ci	139%	40-190 %
981029A-9	0.484 Ci	0.321 Ci	151%	40-190 %
981029A-10	0.451 Ci	0.321 Ci	140%	40-190 %

Although the LCA factors for the GEA portion of the measurement were not implemented at the time of PDP cycle 5A, each of the six replicate measurements of the PDP Dry Combustibles drum passed the accuracy requirements for this activity loading. In all cases, the corrected active neutron combined with the GEA results were reported. The source of the apparent high bias has not yet been determined, however most of this high bias is likely due to the unimplemented GEA portion of the LCA. Reported results also easily passed precision tests with a measured 7% RSD compared to a 12% pass/fail criterion. Finally, the measured position located the standards at the center of the drum, consistent with the actual positioning.

CONCLUSION

Without the LCA in place, the MAS would have selected the active neutron measurement, combined it with the GEA results, and reported the value 0.108 Ci. This result would produce an accuracy of 33% and a clear failure to pass the accuracy acceptance criteria. With the LCA, the MAS selected the active neutron measurement, corrected the measurement, combined it with the GEA results, and confidently reported a value within the acceptance criteria. It is expected that these results will improve even further with the implementation of the GEA portion of the LCA.

With the implementation of the LCA, the IPAN^{TM}/GEA systems are capable of not only identifying the presence of large particles, but correcting the sensitive active neutron and gamma ray emission measurements for the effects of self-shielding. This leads to a more accurate assay and effectively removes the tendency to underestimate the Pu present in waste drums. With the removal of this bias, contributions to the TMU from self-shielding effects are reduced significantly.

REFERENCES

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