PERFORMANCE OF WIPP CERTIFIABLE MOBILE WASTE CHARACTERIZATION NDA MEASUREMENTS ON TRU WASTE DRUMS AT LLNL

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ABSTRACT

BNFL Instruments Inc., formerly Pajarito Scientific Corp., has successfully performed WIPP certifiable mobile non-destructive assay (NDA) measurements at Lawrence Livermore National Laboratories, LLNL. The NDA system used to perform these measurements is a combined Imaging Passive/Active Neutron and Gamma Energy Analysis (IPAN/GEA) system. The measurements meet the requirements of WIPP's Transuranic Waste Characterization Quality Assurance Program Plan (QAPP), CAO-94-1010. This work is one of only two ongoing privately contracted mobile waste characterization projects within the DOE complex. This paper describes the WIPP certification efforts, the CAO accepted measurement technique, and summarizes the measurement results.

INTRODUCTION

The University of California, Lawrence Livermore National Laboratory's (LLNL) Hazardous Waste Management Division (HWM) subcontracted BNFL Instruments Inc. to provide mobile waste characterization services for the non-destructive assay of 602, 208 liter drums, containing Transuranic (TRU) and Low-level (LLW) waste. The contract required that all TRU measurements satisfy the requirements of WIPP's Transuranic Waste Characterization Quality Assurance Program Plan (QAPP) CAO-94-1010, Interim change, November 15, 1996. The TRU assays were required to demonstrate that the waste meets the definition of TRU waste and that all isotopes contributing greater than 1% of the total activity are quantitatively identified.

In August of 1997 BNFL Instruments Inc. moved their mobile IPAN/GEA assay system, see Figure 1, to LLNL to perform the required nondestructive assay measurements. The mobile IPAN/GEA system incorporates three different measurements, active neutron interrogation and monitoring, passive neutron monitoring and passive Gamma Energy Analysis (GEA). The passive and active neutron measurements use the IPAN analysis technique that has been developed over the past several years by BNFL Instruments Inc. The GEA incorporates a Planar HPGe detector collimated to scan the full height of the drum for one full rotation. A robust gamma analysis algorithm has been developed to automatically correct the measured energy response for the effects of both the detector efficiency and the matrix attenuation.

LLNL requested a surveillance be performed by the CAO to ensure that all facets of the NDA process comply with WIPP certification requirements. In September of 1997 the CAO sent a team of experts to LLNL to perform an initial surveillance. Several concerns with BNFL Instruments Inc.'s documentation and QA procedures were found and documented in Corrective Action Reports (CARs). BNFL Instruments Inc. and LLNL addressed these concerns and completed all the documentation by March of 1998. The CAO surveillance team returned and after extensive review of procedures, methods, and documentation the CAO team concluded that "the BNFL Instruments Inc. Mobile IPAN/GEA NDA process employed at LLNL is technically satisfactory for characterization of TRU waste as a subcontractor for LLNL and as a member of a CAO-qualified mobile vendor team."

Figure 1. BNFL Instruments Inc.'s Mobile IPAN/GEA System.

IPAN/GEA SYSTEM

The BNFL Instruments Inc. IPAN/GEA system performs three different measurements and integrates the results to yield one final CAO accepted WIPP certified assay result.

The first measurement performed is an active measurement based on the differential die-away technique [1]. During this portion of the measurement, the matrix is interrogated with neutrons of a wide energy spectrum ranging from 14MeV to thermal energy levels. This interrogation is used to measure the fissile content of the drum and determine both the neutron absorbing properties and the neutron moderating properties of the waste matrix.

A typical neutron time history recorded by the imaging detectors is shown in Figure 2, Signal Detectors. The "B" (background) window does not start until the interrogation flux levels have decayed to negligible levels. The large initial peak in the curve is due to the large pulse of neutrons from the neutron generator. As these neutrons become thermalized in the waste package, fissionable isotopes that are present undergo an (n,f) reaction. These fission events produce the neutron counts shown in the time history in area A. Area A gets larger as the amount of fissile signal in the waste package increases. Area B is the constant background portion that is the result of cosmic rays, (α, n) sources and spontaneous fission neutron emitters.

A small ^{3}He tube, flux monitor, housed in close proximity to the neutron generator in a moderating assembly measures the interrogating neutron flux produced by the generator (see Figure 2, Flux Monitor). Area C represents the time average interrogating flux produced during the time when induced fissions are occurring. It has the same start and stop times as the signal detectors area A.

The amount of fissile material in the waste drum is proportional to (A-B)/C. The measurement is complicated by the fact that neutron moderators and absorbers within the waste box produce effects requiring corrections to the simplified formula given above. A flux monitor mounted close to the drum, BFM, is used to determine the effective interrogation flux. As the amount of absorber in the drum increases, the amount of flux measured by the BFM decreases. The ratio of the BFM to the internal MA flux monitor yields a measure of absorber effects. The moderator effects are determined with an epithermal neutron transmission technique. This involves measuring the epithermal neutrons exiting the waste drum on the side opposite the neutron generator. The epithermal count rate decreases rapidly as the amount of moderator in the waste drum increases.

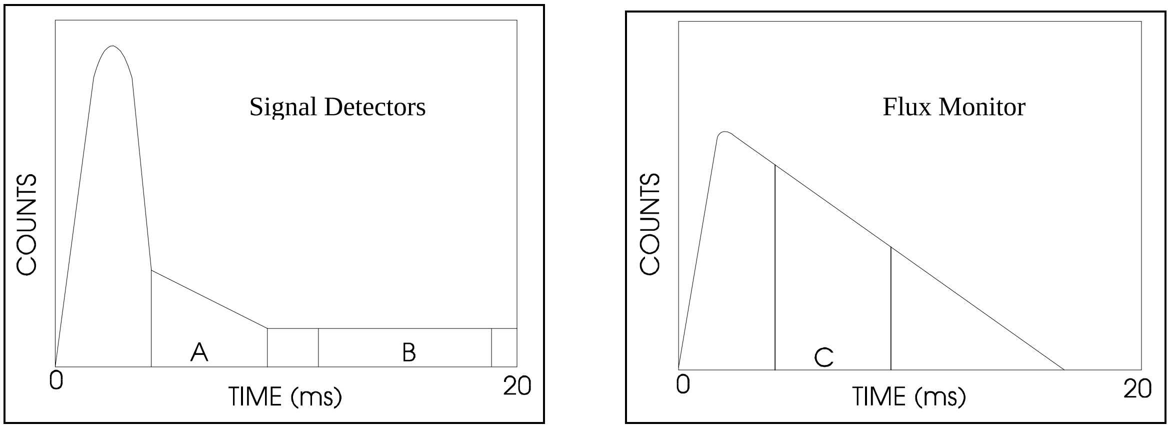

Figure 2. Typical active neutron time histories.

The BFM data (neutron absorber index) and the Epithermal transmission data (moderator index) are measured with a set of "calibration matrix drums" that span the moderator and absorber range expected in the waste drums to be assayed. This "matrix identification" data is used with BNFL Instruments Inc.'s proprietary imaging algorithm [3] to determine basic imaging matrix corrections for each waste drum. The imaging source data then is analyzed (source position imaging analysis) with these proper matrix corrections to produce an accurate measure of the fissile content of the waste matrix.

The passive neutron measurement portion of the IPAN/GEA technique measures the neutron coincidence produced by spontaneous neutron emitters, such as, ^{240}Pu, ^{238}Pu and ^{242}Pu. During this measurement the neutron generator is turned off and the system acts as a neutron coincidence counter. Although the IPAN/GEA system is capable of measuring neutron multiplicity rates, the detector efficiency of approximately 12% is too low to obtain statistically sound multiplicity data. Multiplicity data is recorded using an electronics module developed at LANL, Programmable Multi-Channel Coincidence Module (PMCCM) [2]. This multiplicity data is then reduced to obtain a coincidence measurement that yields a measure of the ^{240}Pu effective. The waste matrix neutron moderator properties determined in the active measurement and the passive imaging [3] results are combined to apply the proper matrix corrections to yield an accurate measurement of the ^{240}Pu effective content of the drum.

Knowledge of the plutonium isotopics is required to convert either the ^{240}Pu effective mass or the fissile mass to a total plutonium mass. It is also important to determine the presence of other interfering isotopes before converting either mass to a total plutonium mass. The Gamma Energy Analysis (GEA) section of the IPAN/GEA system is used to determine the Pu isotopics and to measure all other gamma emitting isotopes.

The BNFL Instruments Inc. Mobile IPAN/GEA system has a Planar HPGe detector built into the system, see Figure 3. This detector is a 20 cm^2 Low Energy Planar germanium detector collimated to scan the full height of the drum. During a drum assay the first measurement performed is the gamma measurement. During this measurement the system door is closed to a position where the HPGe detector is centered on the drum. The acquisition software starts acquiring the gamma spectrum and the drum begins one full rotation. The rotation rate is defined by the preprogrammed gamma measurement time. The default gamma measurement time is 200s.

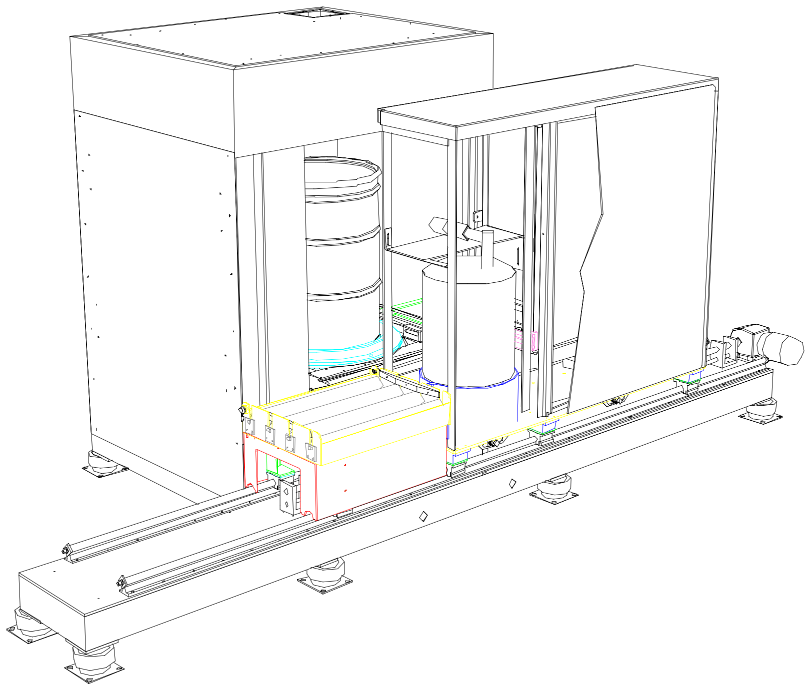

Figure 3. BNFL Instruments Inc. Mobile IPAN/GEA system.

The gamma spectrum analysis implements a recently developed technique to determine the Pu isotopics from a relatively weak gamma signal measured through a highly variable matrix density as typically seen in waste measurements. This technique uses multiple ^{239}Pu-emission lines spaced periodically throughout an extensive region of spectral interest, 95-500KeV, to determine a relative gamma attenuation and efficiency function. Once this function is determined all gamma lines in the measured spectrum can be corrected. This analysis technique will be described in more detail in a separate paper presented by

BNFL Instruments Inc. at this conference. The outputs of the GEA analysis are relative mass quantities of all gamma-emitting isotopes. Converting these relative mass values to quantified isotope masses is accomplished by associating the relative ^{239}Pu-gamma quantity with the quantified ^{239}Pu-neutron measurement.

Integration of the three IPAN/GEA measurements is accomplished after thorough expert review of the gamma spectra to ensure all possible perturbations of the spectra have been considered. This integration consists of determining the ratio of the ^{239}Pu-gamma mass to the ^{239}Pu-neutron mass calculated using either the active neutron measurement or a combination of the gamma isotopic ratios and the ^{240}Pu effective measurement. This ratio is then used to correct the relative gamma mass of all measured isotopes. The ^{239}Pu ratio and quantification of all radioisotopes are performed automatically in the IPAN/GEA application software.

WIPP QUALIFICATION PROCESS

The first step in the BNFL Instruments Inc./LLNL WIPP NDA qualification process was to prepare all the documentation required by the QAPP. The QAPP was reviewed and documents were found or written to meet the requirements. These documents were then organized into an easily accessible set of binders for use during the surveillance.

A surveillance was then scheduled and on September 18th and 19th of 1997 a CAO surveillance team evaluated the LLNL NDA TRU waste characterization activities implemented using the BNFL Instruments Inc. Mobile IPAN/GEA system. They concluded that the NDA process was not satisfactory due to deficiencies in the implementation of the QA program in the areas of calibration, software testing and supplier QA program evaluation. The surveillance team issued corrective Action Reports (CARs) and observations.

BNFL Instruments Inc. realized that a small set of system/project specific documents could satisfy a large number of the deficiencies and observations. Therefore, the following documents were assembled:

Calibration Test Report	7141-CTR-001
Software Validation Verification	W050-DVVD-001
Algorithm Description Document	W050-ADD-001
Project Organization Description	7141-POD-001
QAO Compliance Document	7141-QAO-001

When the CAO surveillance team had reviewed these documents and was satisfied that all the required corrective actions had been completed the follow up surveillance was scheduled, March 17th through the 19th. The follow up surveillance verified the satisfactory completion of all corrective actions and satisfactory responses to all observations from the initial surveillance. The surveillance team identified no deficiencies during the surveillance.

The surveillance team performed an initial review of BNFL Instruments Inc.'s calculation of the IPAN/GEA Total Measurement Uncertainty (TMU). They determined that the calculated and reported TMU appears acceptable but additional reviews were needed prior to acceptance of the TMU. The surveillance team commented that the TMU calculations appeared highly conservative and the outcome of the final review of the TMU was not anticipated to negatively affect the usability of the NDA data by LLNL. The TMU approach was later accepted by the CAO after implementing changes to remove some of the conservancy in the calculation.

MEASUREMENT RESULTS

BNFL Instruments Inc. measured a total of 602 waste drums at two different locations on the LLNL site between February 2nd and May 21st. During this period there was one month of downtime while the LLNL safety operating documentation was prepared for operations at their plutonium facility. An average of 13 drums per day (one eight-hour shift) was assayed to generate a total of 48 batches of WIPP certified data.

Drum weights ranged from a maximum net weight of 148.1 kg to a minimum of 4.3 kg and averaged 40.8kg, see Table 1 for a summary of measurement results. The plutonium content in the drums ranged from no Pu to over 65 grams of total Pu. The isotopic grades of Pu varied from standard weapons grade, ~6% ^{240}Pu, to LLNL reactor grade, ~25% ^{240}Pu. The Am content of the drums also varied greatly. Some drums contained Am levels consistent with in-growth from relatively young ^{241}Pu while others contained levels that could only be due to ^{241}Am enrichment. All measurements and analysis were performed assuming no acceptable knowledge. As the analysis progressed, acceptable knowledge was found to be sufficient for a handful of low-level waste drums where no gamma signal could be measured. Many different radioisotopes were found to be present in the LLNL waste. Here is a complete list of the measured isotopes:

^{238}Pu	^{235}U	^{137}Cs	^{249}Cf
^{239}Pu	^{238}U	^{152}Eu	^{252}Cf
^{240}Pu	^{241}Am	^{154}Eu	^{85}Kr
^{241}Pu	^{243}Am	^{228}Th	^{227}Ac
^{242}Pu	^{237}Np	^{243}Cm	
^{233}U	^{60}Co	^{244}Cm	

As can be seen in Table 1, the waste matrix properties varied considerably. Neutron moderator and absorber indices varied from levels close to an empty drum to indices at the upper limit of the current accepted range of the IPAN/GEA system. The maximum measured moderator index, 18.8, is in the range of the IPAN/GEA concrete matrix calibration. The maximum measured absorber index, 231.9, is in the range of the IPAN/GEA metals matrix calibration.

Out of the 602 expected TRU waste drums measured, 60 were determined to be low-level. That is, the measurement result demonstrated at a 95% probability that the TRU activity density was less than 100nCi/g.

Table 1

Summary of Measurement Results

	Net Weight (kg)	Moderator Index	Absorber Index	Total Pu (g)	Total α-activity (Ci)	TRU α-activity Density (nCi/g)
Maximum	148.1	18.8	231.9	65.8	20.5	493,941.0
Average	40.6	5.3	132.4	6.2	0.97	25,021.7
Minimum	5.4	1.9*	22.1**	DL(<0.001)	DL(<0.001)	DL (<20)
* Moderator index for an empty drum is 1.44						
** Absorber index for an empty drum is 14.7						

CONCLUSIONS

BNFL Instruments Inc. began its effort to obtain CAO WIPP certification in August of 1997. After seven months of effort from LLNL and BNFL Instruments Inc., the BNFL Instruments Inc. NDA process implemented at LLNL was certified by the CAO for characterization of TRU waste. The process of obtaining this certification was intensive. The use of an NDA system that is technically capable of measuring the entire range of waste does not guarantee a WIPP certified measurement. The greatest amount of effort must be applied to meeting every requirement (every sentence) in WIPP's Transuranic Waste Characterization Quality Assurance Program Plan (QAPP), CAO-94-1010. The use of the QAPP compliance matrix in determining how each requirement is satisfied is the most thorough method. BNFL Instruments Inc.'s approach was to not overwhelm the auditors with documents and data but to be as concise as possible. A set of binders was prepared that contained the documents necessary to show compliance with the QAPP. This method worked well with the auditors. It led to a good working relationship between the CAO audit team and the BNFL/LLNL NDA team.

The assortment and combinations of isotopes encountered at LLNL has prompted the BNFL Instruments Inc. NDA team to refer to the drums as "gourmet waste". The drums at LLNL emphasize the need for the expert analysis factor as an essential part of the NDA process. There were very few drums that actually proved to be standard weapons grade drums with no "surprises". In addition, very few of the drums were "alike", so that programming for automatic analysis of possible combinations would not only be time intensive, but would also fail to account for every situation that will occur when assaying real waste drums. In the LLNL waste, the biggest surprise was probably the number of drums that contained added ^{241}Am, ~20%. The number of drums with measurable amounts of ^{237}Np, ~70%, was also surprising. The other commonly seen isotopes were ^{243}Am, ^{243}Cm, and ^{235}U. We also encountered WGPu drums that, in addition, contained ^{244}Cm. In all of these cases, and in all of the "standard" drums, each component of the IPAN/GEA provided useful and necessary information. For instance, additional confirmation of a ^{244}Cm presence was seen in the imaging data as a strong passive source not co-located with the active source. We also saw at least one case where the source was very clearly a lump at the center of the drum. This was a situation where the active system did not provide the best assay, but for which the passive system did extremely well in conjunction with the imaging. In general, all analysis was carried out successfully under the WIPP certified procedures while still allowing for the flexibility apparently necessary for real waste assay. The need for all of the systems, passive neutron, active neutron, passive gamma, and expert analysis, seems to be a prerequisite for any useful waste characterization effort.

REFERENCES

[1] J. Caldwell, R. Hastings, G. Herrera, W. Kunz, E. Shunk, "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers," Los Alamos National Laboratory, LA10774, September 1986.

[2] G. Brunson, G. Arnone, A New System for Analyzing Neutron Multiplicities: Characterization and Some Specific Applications," Los Alamos National Laboratory, LA-11701-MS, November 1989.

[3] J. Caldwell, J. M. Bieri, T. Kuckertz, M. Newell, K. Plettenberg, T. Peterson, R. West, W. Embry, "High Sensitivity Imaging Passive and Active Neutron (IPAN) Transuranic Assay System," 92PSC135, May 1992.