# IPAN/GEA EFFICIENCY-TIMES-ATTENUATION METHOD FOR WASTE CHARACTERIZATION

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## ABSTRACT

In June 1998, BNFL Instruments Inc (BII) completed measurements utilizing the IPAN/GEA Mobile Assay System on approximately 600 waste drums at Lawrence Livermore National Labs (LLNL). Incorporated in the system is an innovative approach to obtaining isotopic information from the high-purity germanium (HPGe) detector and merging it with the imaging passive/active neutron (IPAN) measurement to produce a final quantification of all isotopes present in the waste. The technique, known as the Efficiency-Times-Attenuation (ETA) method, is a component of the gamma-ray energy analysis (GEA) portion of the measurement. This BII proprietary method is an iterative process which, accounting for the detector efficiency and attenuation effects of the matrix, produces a relative quantification of the isotopes detected. These quantities, along with an isotopic correlation method, are reviewed during offline analysis performed by BII to arrive at final isotopic quantities for all isotopes present. The results are then combined with either the imaged active neutron measurement or the imaged passive neutron measurement to quantify the total Pu content of the drum as well as the activity produced by all isotopes detected. The performance of this method on the LLNL waste and the expert analysis process is presented. A large range of matrix types and Pu loadings, as well as an assortment of isotopes, which provided an excellent forum for the system's capabilities, were analyzed for the site.

## INTRODUCTION

From February 1998 to May 1998, approximately 600 waste drums were assayed for Lawrence Livermore National Lab (LLNL) using the BNFL Instruments Inc (BII) Mobile Assay System (MAS). The waste drums were expected to contain materials running the gambit of absorber and moderator effects, Pu loadings, and Pu grade isotopic concentrations with an assortment of alpha emitting isotopes. Although WGPu (5.9% $^{240}$Pu for LLNL) was expected to be the primary source of neutrons, from both spontaneous and induced fission, additional contributors, such as U235, Cm244, and Cf252, were expected in a small percentage of the drums.

In order to accommodate the assortment of waste expected throughout the Department of Energy (DOE) complex, and demonstrated through the assay of the LLNL waste, the MAS was designed to include 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 considered proven methods.

Throughout the history of these techniques, however, a gap has existed in reconciling data from one technique with data from another. To that end, BII has developed a proprietary technique known as the Efficiency-Times-Attenuation Method. This method, along with isotopic correlation (IC) and off-line expert analysis, allows data from IPAN measurements and HPGe measurements to be utilized to the fullest extent possible. The basic premise is that all nuclear signatures observed—active neutron, passive neutron and passive gamma—can be reconciled and co-processed in a coherent fashion to obtain a final result that is more complete and accurate than would be possible in a separate analysis of the three signatures.[1]

The LLNL waste drums assayed provided a rigorous test of this blending of IPAN and HPGE data. It also reinforced the best aspect of the MAS: It is a stand alone system, fully capable of discerning low-level waste (LLW) from tranuranic (TRU) waste and providing complete assay documentation for WIPP-bound DOE waste drums.

## THE MAS IPAN/GEA SYSTEM

The MAS developed by BII, 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. Absorbing 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 determines 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.

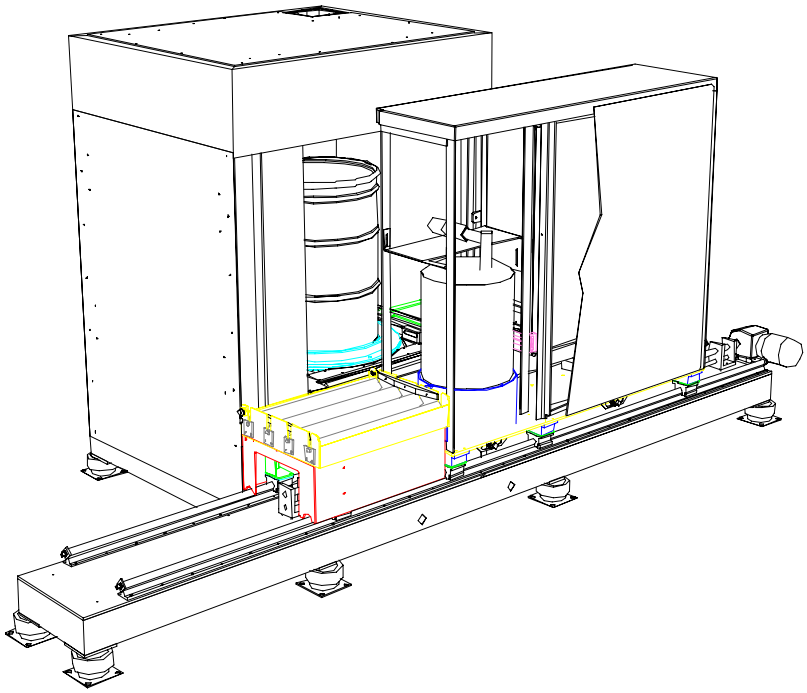

**Figure 1** BII Mobile Assay IPAN/GEA Drum System

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 ETA/IC method carried out during expert analysis.

## THE EFFICIENCY-TIMES-ATTENUATION METHOD

The PSC planar HPGe detector is utilized to obtain a comprehensive gamma spectrum of the contents of the entire waste drum. Since it is expected that the gamma ray emissions of Pu239 usually will dominate such spectra, the analysis is formulated in a fashion to facilitate the calculation of ratios of all other gamma emitting constituents to Pu239, as is possible for a given spectrum. The fact that Pu239 has strong emission lines spaced periodically throughout an extensive region of spectral interest—95-500 keV--is a key factor in this analysis.[1]

In a given waste drum the amounts and types of waste present as well as the geometric disposition of the gamma emitters relative to the waste constituents—produce a complicating situation to the easy determination of gamma line ratios. The detector intrinsic efficiency curve is considerably modified by the presence of absorbing waste drum constituents as well as by source material self-absorption effects. The basic premise of the standard isotopic gamma line ratio method is to use only pairs of lines that differ so little in energy that such complicating effects are negligible.[3] However, in reality, a sufficient number of conveniently adjacent and measurable gamma line ratios is not possible for real waste spectra. Even worse it is often the case that the underlying assumptions that are valid for isotopic line ratio analysis of "laboratory spectra" of small source samples are simply not valid for the far more complex spectra emanating from large, heterogeneous waste packages. Thus, even though the usual isotopic line pairs can be measured—use of them in the usual simplistic fashion can result in a considerable systematic bias in final isotopic ratios.[1]

The ETA method developed by BII 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 BII proprietary method is an iterative process which "fits" the spectral data to produce a relative quantification of the isotopes detected. Before a final fit is completed, however, the data is reviewed by BII "experts" for such complicating factors as uncorrected interference and low statistics. For instance, if a large amount of $^{243}$Am appears to be present, the "expert" may choose to exclude the 100 keV region from the analysis.

## THE ISOTOPIC CORRELATION METHOD

Once the data has been reviewed, the fit is completed and mass quantities of the isotopes identified in the spectrum are reported, corrected for the matrix effects. However, in order to produce an accurate and complete determination of the Pu isotopics, an isotopic correlation method is necessary. Even the most comprehensive gamma spectral measurement determination of Pu isotopics in an unknown sample or waste drum must be supplemented by use of isotopic correlations—always to determine the proper amount of $^{242}$Pu present since this isotope does not emit a detectable gamma ray.[1] In the case of low activity waste drums, some additional Pu isotopes may also be "silent". In particular, $^{238}$Pu and $^{240}$Pu lines may be missing.

The basic nuclear physics underlying isotopic correlation methodologies is that all the Pu isotopes from a single batch are produced in a common nuclear reactor environment. The ratios among the various Pu isotopes in different batches will vary primarily according to the product of flux and exposure time. Pu239 is produced by a single neutron capture in the original U238 target material. Pu240 results from the capture of a second neutron, Pu241 from the capture of a third neutron and Pu242 from the capture of a fourth. Pu238 results from (n, 2n) reactions (high-energy neutrons in the reactor core flux) in the Pu239. The relative amounts of each isotope produced in different batches generally vary in a smooth fashion with the underlying primary flux-exposure time parameter. It is this functional dependence that makes isotopic correlations feasible.

| PU ISOTOPIC DATA FOR A VARIETY OF PU MATERIALS,BY WT % | | | | | | |
|---|---|---|---|---|---|---|
| DESCRIPTION | 238 | 239 | 240 | 241 | 242 | 240eff |
| LLNL,WG | 0.015 | 93.77 | 5.95 | 0.31 | 0.06 | 6.08 |
| LANL,WG | 0.015 | 92.97 | 6.72 | 0.55 | 0.07 | 6.87 |
| MARCOULE | 0.020 | 91.50 | 7.85 | 0.60 | 0.08 | 8.04 |
| NFS-DUPONT | 0.080 | 85.63 | 11.53 | 1.16 | 0.25 | 12.17 |
| NFS-HALDEN | 0.290 | 79.53 | 14.51 | 2.24 | 0.83 | 16.70 |
| NFS-CONS PWR | 0.360 | 75.81 | 16.34 | 2.91 | 1.18 | 19.32 |
| BELGO--PU/MOX | 0.870 | 64.53 | 23.77 | 7.40 | 3.43 | 31.96 |
| BELGO--PU/MOX | 0.880 | 64.87 | 23.58 | 7.12 | 3.55 | 32.00 |

**Table 1** Isotopic Correlation fit data.

We have determined appropriate functional fits to the various database quantities utilized for our isotopic correlations based on the data in Table I. From this data, the correlated % $^{238}$Pu, $^{239}$Pu, $^{240}$Pu, original $^{241}$Pu, and $^{242}$Pu (relative to total Pu mass) as a function of the parameter $^{240}Pu_{eff}$ (also expressed as a % of

total Pu mass) can be determined. An example is shown in Fig 2. This plot shows the functional dependence of $^{238}$Pu (Col 2, Table 1) with increasing % $^{240}Pu_{eff}$ (Col 4, Table 1)--clearly indicating a smoothly varying systematic dependence.[1] This is true for all of the Pu isotopes from which additional functional relationships can be determined.

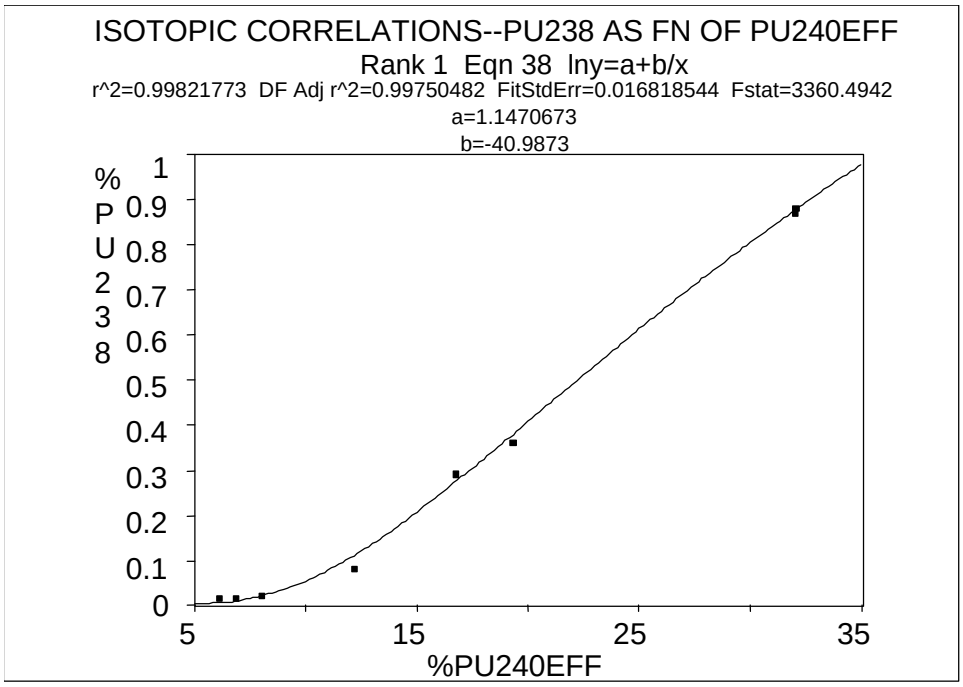


Figure 2. Isotopic Correlation of %$^{238}$Pu as a Function of % $^{240}Pu_{eff}$

Once the ETA method has been applied and the IC method has produced isotopics for the "silent" Pu isotopes present in a particular drum, the final isotopics are produced. All isotopic quantities are determined relative to the total Pu mass determined from the HPGe data. These ratios are then applied to the $^{239}$Pu quantity from the active neutron measurement or the $^{240}Pu_{eff}$ from the passive neutron measurement. The final isotopic mass quantities and total Pu reported for a drum are thus a combination of the more accurate and sensitive neutron measurements and the isotopics produced by the gamma measurement.

## PERFORMANCE RESULTS

The ETA/IC method was applied to the analysis of approximately 600 drums at LLNL. Of these, 49 were found to be LLW. The remaining drums were certified for final disposition at the WIPP site in Carlsbad, New Mexico. The drums were expected to contain the Pu grades indicated in Table 2. Combinations of these grades were also expected as several packages with different types of contamination could make up the contents of a given drum. The BII isotopic results for several LLNL drums are listed in Table 3. The LLNL column is the average grade expected for the indicated drums..

| Pu GRADE | %238Pu | %239Pu | %240Pu | %241Pu | %242Pu | %241Am |
|---|---|---|---|---|---|---|
| Weapons | 0.016 | 93.464 | 5.900 | 0.381 | 0.040 | 0.202 |
| Fuel | 0.066 | 78.964 | 17.427 | 1.180 | 0.432 | 1.942 |
| Reactor | 0.011 | 73.657 | 24.896 | 0.424 | 0.018 | 0.994 |
| Americium Enriched | 0.055 | 63.557 | 14.027 | 0.950 | 0.347 | 21.073 |
| Mixed | 0.042 | 86.149 | 11.714 | 0.784 | 0.237 | 1.079 |

**Table 2** LLNL average isotopics for several grades of Pu.

| BII Drum ID | LLNL Grade | BII %239Pu | BII %240Pu |
|---|---|---|---|
| 980209-15 | Weapons | 93.75 | 6.04 |
| 980210-4 | Weapons | 93.12 | 6.64 |
| 980210-9 | Weapons | 94.00 | 5.69 |
| 980210-13 | Mixed | 89.21 | 10.20 |
| 980211-6 | Weapons, Fuel | 87.20 | 11.27 |
| 980211-15 | Weapons, Fuel, Mixed | 88.83 | 10.76 |
| 980212-5 | Weapons, Fuel | 90.17 | 9.29 |
| 980212-6 | Weapons, Mixed | 91.43 | 8.24 |
| 980212-7 | Weapons | 92.42 | 7.30 |
| 980212-11 | Fuel | 85.31 | 13.34 |
| 980213-10 | Fuel | 83.87 | 14.15 |
| 980213-11 | Weapons | 93.53 | 6.22 |

**Table 3** Isotopic results for LLNL waste drums

The drums presented in Table 3 were randomly selected as a good representation of Pu grade drums assayed by the MAS. The results from the BII ETA/IC method were consistent with previous measurements done on the drums using other assay instrumentation, and in several cases, BII's analysis indicated errors made in the original non-WIPP certified quantifications.

Of the Pu contaminated drums assayed, most contained any number of isotopes not associated with Pu decay, like $^{241}$Am or $^{237}$Np. Some of the isotopes identified and quantified by BII include $^{243}$Am, $^{243}$Cm, $^{235}$U, $^{152}$Eu, $^{60}$Co, and $^{85}$Kr. In all cases, the isotopes expected to be present by LLNL were either detected or at levels below the detection limit of the system. In these cases, the ETA method was used for detection limit quantification or, in the case of the Pu isotopes, the IC method was used for quantification. Table 4 shows examples of drums containing isotopes other in addition to Pu.

| **BII Drum ID** | **LLNL Additional Isotopes** | **BII Additional Isotopes** |
|---|---|---|
| 980402-8 | Added Am241, Cm244, Cs137, Kr85, U235, and U238 | Added Am241, Cs137, Kr85<br>Detection Limits for Cm244, U235, and U238<br>Also detected: Th228 |
| 980515-6 | Added Am241 | Added Am241, Np237<br>Also detected Cf249 |
| 980515-8 | Added Pu238 | Added Pu238, Np237 |
| 980402-12 | No additional isotopes | No additional isotopes |

**Table 4** Isotopes quantified using the ETA method (Pu isotopes were also present and quantified using the ETA/IC method)

Quantities determined using the ETA/IC method and the neutron measurements generally agreed with the LLNL records for the waste drums. In most cases, additional isotopes were identified and quantified.

## CONCLUSION

The BII MAS provides a complete and accurate assay of all types of real waste drums. Individually, the three measurement methods provide a great deal of information about the drum and its constituents. Without generator acceptable isotopic knowledge or known matrix effects, however, the picture from each measurement is incomplete. The ETA/IC method eliminates the need for either of these inputs and

produces, using data gathered from all three, a detailed and "focused" quantification of the drum contents. As a result, the drum activity is fully determined and documented for storage at WIPP.

The LLNL drums provided an excellent testing ground for the MAS and the BII ETA/IC proprietary method. In the 600 drums, the Pu content, matrix effects, and additional constituents varied widely. The ETA/IC method proved capable and flexible enough to handle most situations that arise in waste drums.

## REFERENCES


(1) BII Document BII-7141-EAD-001 Mobile Assay System Expert Analysis Description.
(2) Stephanie A. Jones, Randy F. Lucero, and Mike A. Purcell, "Measurement Results for the IPAN/GEA Boxed Waste Assay System" INMM Annual Meeting, Naples, Florida, 27-30 July 1998.
(3) Reilly, D., Ensslin, N., Smith, H., *Passive Nondestructive Assay of Nuclear Materials*, pp.221-271, U.S. Nuclear Regulatory Commission, Washington, DC 20555, March 1991.