

GREATLY IMPROVED TRANSURANIC WASTE ASSAY ACCURACY USING NEUTRON SIGNAL IMAGING

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HISTORICAL BACKGROUND

A group of nuclear scientists and engineers at the Los Alamos National Laboratory developed and patented the original Passive Active Neutron (PAN) assay technology in the 1978-85 time period. (Several of these original inventors, led by John Caldwell and Walter Kunz, later formed Pajarito Scientific Corporation (PSC) in 1986 as a technology transfer company specializing in implementation and further development of the PAN assay technology.) Since the early 1980's Non-Destructive Assay (NDA) specialists in virtually all countries having a significant nuclear industry have utilized this technology for a variety of nuclear safeguards and waste management applications. As in all other NDA technologies the twin development goals of improving on existing assay sensitivity and assay accuracy have been pursued. This paper describes PSC's most recent developmental work in PAN assay technology — the use of neutron signal imaging to significantly improve assay accuracy.

IMPROVED ASSAY SENSITIVITY

Several 208 liter drum-size PAN units of the Los Alamos Group N-2 type were put into service at various DOE sites during the time period 1982-86. Much of the transuranic (TRU) wastes destined for ultimate disposal at the Waste Isolation Pilot Plant (WIPP) have been assayed in these units. The field performance record of these units has been highly satisfactory, with the lower limit of detectability generally being in the 2 - 10 mg ²³⁹Pu range.

One of the primary goals PSC identified for its PAN technology development program in 1987 was the significant improvement of minimum detectable fissile amounts. Our first generation improvements were incorporated in the "INTEGRATED FIVE STATION NDA SYSTEM" delivered to Nuclear Fuel Services of Erwin, TN in 1989-90, and described in the references. This system provides comprehensive NDA support for the Decontamination and Decommissioning (D&D) of an older Plutonium Mixed Oxide (MOX) fuel fabrication facility.

Incorporated in this system are two separate PAN units (one large glove box size and one drum and compressed waste bale size). These units incorporate PSC's proprietary "External Source"

neutron interrogation source — an improvement that produces a greater than 500% improvement in bottom line useful interrogation fluxes relative to the originally installed DOE site PAN systems. A second significant improvement was achieved with the development of lower active background detection packages — about a factor of two improvement relative to the original system detection packages. The combination of these two improvements (interrogation flux, detector backgrounds) results in a corresponding improvement in plutonium assay sensitivity. At Erwin, for example, the (5' X 6' X 10') glove box PAN unit achieves routine 5 nCi/g performance and the drum-bale unit achieves a 1 nCi/g sensitivity.

This same low detector background and external interrogating source technology has been incorporated into the design of the 208 liter size PAN system described in this paper.

IMPROVED ASSAY ACCURACY

The primary sources of assay error in PAN systems (as well as in all other NDA systems used to assay bulk wastes) are all directly related to non-uniform measurement responses encountered in real world wastes. These in turn amount to errors associated with the unknown spatial locations of the sources being measured — which differ greatly from one waste matrix type to the next. PSC has been working on a new type of PAN system which attacks this primary source of assay error. The approach is that of "**Imaging Passive-Active Neutron**" or **IPAN**. In this approach the PAN system detectors are arranged in a configuration and the PAN measurement is performed in a manner to facilitate neutron imaging of both signal and interrogating fluxes. The signal portion produces an image of all source materials within the waste package and the interrogating flux portion provides waste matrix imaging and identification. Together, this truly neutron imaging approach allows the measurement process to achieve a 300% or better improvement in assay accuracy relative to the original PAN systems.

We have built and tested two generations of 55/85 gallon size IPAN units, both at our D.P. Road facilities in Los Alamos and at a DOE site. The most recent demonstration unit accommodates standard US/international 55 gallon/220 liter waste/scrap drums, as well as standard 85 gallon overpacks.

DRUM SIZE IPAN UNIT DESCRIPTION AND TEST RESULTS

We have just completed an exhaustive series of waste matrix tests with this first production run 200 liter size IPAN unit. These tests consisted of a systematic set of distributed fissile mass measurements within each of 10 mockup waste matrices placed inside typical US style 210-220 liter, mild steel drums. The matrices were chosen to mock-up a large variety of actual waste forms commonly encountered in DOE, commercial and international nuclear facilities. Among these were: (1) Drums filled with light (<10kg) and heavy (50kg) cellulose-combustibles mockups. (2) Three drums designed to simulate the range of 5% to >75% PVC content in mixed combustibles-PVC waste. (3) Two drums simulating glassware having a range of borax content between 5 - 15%. (4) A drum filled with 250 kg of miscellaneous iron-steel scrap and pieces. (5) A drum filled with 250 kg of 5 - 10 cm diameter concrete pieces, simulating typical concrete wall and floor debris generated from decontamination and decommissioning activities. (6) A drum filled with 450 kg of solid concrete to simulate concreted, processed waste forms. These data were acquired with PSC's on-line imaging data acquisition hardware and software, and processed using our on-line imaging software algorithm. A standard personal computer

(IBM/PC/AT type, 33 MHz processor) was used with combined passive and active neutron data acquisition times of 10 minutes or less and imaging/other processing times of two minutes or less.

Each drum contained four, 2.5 cm diameter sample placement tubes (construction: thin wall aluminum) placed axially in each drum at (a) the geometric center-line (b) the mid-radius line (10 cm out from center) of the central 1/3 volume of the drum (c) the mid-radius line of the middle 1/3 volume of the drum (20 cm out from center) and (d) the mid-radius line of the outermost 1/3 of the drum volume. Fissile samples could thus be placed at precise, reproducible locations within each test matrix form, facilitating the determination of accurate systematic behaviors.

Each standard IPAN measurement consisted of 16 separate angular measurements for both active and passive portions. The drum is rotated under automatic, programmed stepping motor control through 16, 22.5 degree rotations for both active and passive portions. Angular reproducibility for all drums, including those weighing 250 and 450 kg, was better than one part in 10,000 for a full revolution — that is, the angular deviation between starting and final positions of the turn table following one revolution (16 stops/starts along the way) was <.04 degrees on the average. This level of rotational accuracy was found to be adequate for our purposes. The operational and program control protocol results in an automatic active-passive acquisition cycle upon initiation of a run — about 3 minutes total for the active portion and about 6 minutes for the passive portion. Thus, two full revolutions of the turn table occur for each total acquisition cycle of 10 minutes or less.

MATRIX EFFECTS

Several in depth discussions of the effects of matrix materials of various types — as they perturb both passive and active neutron measurements — may be found in the published literature. The solution to the complex and perplexing "matrix correction" problem is not satisfactorily completed by a simple "matrix identification" process. This is only the first step required. Once proper matrix identification is achieved, the next, and more difficult step is to take into account the several position sensitive matrix effects that occur at different locations within a drum — for example, variation of interrogation flux and detection efficiency with radius and height in a constant composition drum matrix.

For a specific case of interest that we have investigated (drum filled with 450 kg of solid concrete) we observed that a fissile sample placed at the drum's center produces only 1/100 the active neutron signal that the same fissile sample produces near the same drum's outermost radius. Thus, even if one uses a properly averaged single "solid concrete matrix" calibration factor for this drum, a very large measurement error will occur on average. Measured assay values will be dramatically low for fissile waste distributions located near the drum's center and dramatically high when the fissile waste occurs at an outer radius.

On the other hand, if the fissile waste distribution is "imaged" and the proper positional response is taken into account for all distribution volume elements — an accurate assay will be obtained for all waste distributions — independent of their locations within the drum.

In essence, that is exactly what PSC's IPAN system hardware, software and imaging algorithm accomplishes. The first step of "matrix identification" is accomplished using pulsed active neutron matrix identification and imaging measurements. Ten specially designed and height/radius/rotational angle sensitive flux monitors located both in the assay chamber open volume and within the chamber walls are utilized to "measure" the drum matrix. Note that both moderator and absorber effects are measured at each angular position of the drum as well as at

different heights spanning the complete drum volume. Note also that these pulsed neutron matrix measurements are made and recorded simultaneously with the active signal measurements. A drum volume weighted average matrix composition is determined at present using a methodology similar to that described in Los Alamos report LA-10774. Heterogeneous matrix distributions may be taken into account as required by a more complicated analysis.

NEUTRON SIGNAL IMAGING

Next, the neutron signal imaging data is processed using a least-squares matrix reconstruction analysis that compares observed signal data with processed calibration response sets derived from comprehensive experimental response data. This step determines in a least squares sense the most probable actual fissile (and passive source) signal distributions within the drum that are also compatible with the indicated matrix material properties. Note that this image reconstruction algorithm is an intrinsically three dimensional, volume element process that minimizes the squared differences between observed and predicted active and passive neutron sources — a process that is very sensitive to both the shape and magnitude of the entire set of measured quantities. The "symmetric distribution redundancy problems" sometimes encountered in image reconstruction algorithms do not occur here. For instance, a neutron source placed at the drum's center is easily distinguished from any "centrally symmetric" distribution of sources.

Finally, the same imaging algorithm is used to apply the appropriate spatial response matrix corrections required for the "least squares distribution", in a volume element by volume element sense. The corresponding fissile and passive masses are calculated from these "microscopically" matrix corrected data quantities — as are the corresponding assay errors.

FISSILE DISTRIBUTION MEASUREMENTS

PSC has available a set of some 20 calibrated fissile sources (^{235}U as the fissile isotope) ranging in fissile content from a few mg to several gm quantities. These are used to determine both ^{235}U and ^{239}Pu response characteristics (active neutron interrogation portion of the assay) through use of the constant $^{239}\text{Pu}/^{235}\text{U}$ response factor ratio of 1.50 +/- .03. That is, one gram of ^{239}Pu produces an active signal (total prompt fission neutron signal) that is a factor of 1.50 times the corresponding pulsed active signal produced by one gram of ^{235}U .

250 KG CONCRETE RUBBLE MATRIX

Figure 1 shows the results of the PSC imaging measurement and analysis as performed with a set of very small fissile samples, in the range 0 - 30 mg Pu equivalent. The matrix is the drum filled with 250 kg of small concrete pieces — approximately 10 cm average diameter. This matrix simulates typical concrete rubble associated with D&D projects. The average MCF for this matrix is 1.5, a significant matrix correction. This plot demonstrates the "robust" nature of both the data and the least squares matrix reconstruction analysis. Linearity of response with fissile mass is obtained with this very heavy matrix — and measurements of the same mass at different locations within the drum all fall within a small band of response.

Figure 1

Note that this same data processed in the usual non-imaging manner leads to a much larger scatter of response for each fissile mass. Note also that the minimum detectable fissile mass deduced from this data set is 0.3 mg ^{239}Pu — which corresponds to a specific activity for the concrete rubble matrix of about 0.1 nCi/g of total alpha activity, assuming nominal 6% Pu^{240} isotopics.

Figure 2 shows additional data taken with this same matrix using a single, very small fissile source. This source is approximately that associated with 3 nCi/g total alpha activity plutonium waste. A total of 21 data points were taken: bottom, middle and top height variations combined with center, middle and outer radius locations. The top plot shows this data analyzed with PSC's imaging algorithm and the bottom plot shows the same data analyzed with the standard single parameter PAN calibration. Note that the average assay result (set of 21 values) from each analysis is the same. It is readily apparent, however, that the PSC imaging analysis produces a considerably more accurate assay result (factor of two on average — 21% average error for PAN and 11% for IPAN) than the standard analysis — even at these very low fissile loadings with their attendant poor counting statistics. The imaging methodology is robust.

FIGURE 2

Figure 3 shows another distribution set measured for the concrete rubble matrix. (Details of the distribution are discussed below — a total fissile mass of 1.5 gm ²³⁹Pu equivalent in three 0.5 gm sources was used.) This distribution set (44 separate locations of the three sources) samples the entire drum volume in a systematic fashion as described below. Because the counting statistics are not a contributing factor at these signal levels — the overall assay accuracy differential between IPAN and PAN analyses for this set is even greater — **a factor of 6.4 improvement** is observed using the imaging.

FIGURE 3

SYSTEMATIC FISSILE DISTRIBUTION DETAILS

For the 10 matrix systematic fissile distribution response tests the 20 fissile sources were used both singly and in complex distributions of as few as two and as many as ten sources at a time placed at specific locations within the drum. For each of the ten matrix drums at least 40 individual, spatially different fissile mass distributions were measured. For most of these tests a "quasi-standard" distribution set was utilized to facilitate comparisons between matrices. This standardized set utilized three uranium sources, each producing a fissile signal equivalent to 0.5 gm ²³⁹Pu. The distribution set was obtained by first placing each of the three uranium sources in a separate radius sample tube and lowering all samples to the drum bottom. A bottom measurement was made followed by a sequence of 25 - 30 measurements made with the sources raised in unison by 2.5 cm for each new measurement until the entire drum height had been measured. Next the three sources were all placed in the same radial source tube for a sequence of 10 - 15 measurements of this "line source" at different average heights. At least two radial tubes — an inner one and an outer one — were so measured for each matrix drum.

The 40+ measurements (all data taken in imaging format) so made with the same total fissile mass distributed in 40 different ways within the drum were then analyzed using the PSC imaging algorithm as described above. At the same time each measurement was also analyzed in the traditional PAN fashion using the "most probable" single value calibration factor (i.e., an experimental volume weighted average calibration factor). The two mass values for each of the 40+ measurements were then tallied, entered into a spread sheet and statistically analyzed to determine the average standard deviation (relative to the 1.5 gm ²³⁹Pu equivalent tag value) for each method of data analysis. The data set for each matrix was then plotted for visual examination purposes.

Also, the "average measurement error" (i.e., the one sigma standard deviation for the set) for both analyses and for each of the 10 matrices was calculated. At the end of the paper we show a plot of the average assay errors as a function of the "Average Matrix Correction factor" for each drum. This is defined as the ratio of the measured volume weighted average response for an empty drum divided by the corresponding volume weighted average response measured for the given matrix drum.

For orientation purposes, the Average "MCF" is 7.8 for the worst case examined — the >75% PVC + combustibles matrix and 1.0 for both the light and heavy combustibles matrix. (As discussed above, the average MCF is 1.5 for the concrete rubble matrix.) Generally speaking, one expects the average measurement error to increase as the average MCF increases. This is indeed observed.

SOLID CONCRETE MATRIX

Figure 4 shows the "standard distribution data set" for the 450 kg (1000 lbs) solid concrete matrix. As can be seen the usual (PAN) analysis leads to a large scatter of response about the tag value number. The deviations are particularly large for the inner and outer radial "line source" measurements. On the other hand, the imaging (IPAN) analysis leads to a narrow response band centered about the expected assay value. (Note that the average matrix correction factor is 5.0 for solid concrete — large.) The average assay error determined for this data set with the PSC image reconstruction analysis is 14.1% relative to the tag value. Using the usual PAN single value calibration analysis, the average assay error is 62.3%. The value of the imaging analysis is quite

clear. In fact, we observe that the "nastier" the matrix (i.e., greater total matrix correction factor) the more dramatic is the difference between imaging and non-imaging analyses.

FIGURE 4

HIGH ABSORPTION MOCK GLASS MATRIX

Figure 5 shows the results for the high absorption mock glass matrix. As can be seen, this matrix displays a considerably different intrinsic response profile (i.e., PAN analysis) than either of the others. Here distributions near the drum bottom provide an enhanced response and those near the top a decreased response. This matrix also has a large MCF — an average factor of 3.8. The PAN analysis results in an average assay accuracy of 32% and the imaging analysis in an average error of 11% — an improvement of about 300%. In fact, the same qualitative results are obtained for all ten matrices — the assay improvement factor ranges from a factor of three times better to more than six times better for the imaging analysis relative to standard PAN analysis.

FIGURE 5

The final figure — Figure 6 — shows the composite data — the average assay error as a function of MCF, plotted for imaging analysis (crosses) and for standard analysis (solid squares). As can be seen — the imaging analysis greatly improves the assay accuracy for all matrices — by an improvement factor that averages more than 300% for the set of 10 different matrices. The set of 10 matrices we have tested span a very large range of types that are difficult to measure using any NDA method — and passive and active neutron methods in particular. The basic assay accuracy advantages of PSC's imaging type of assay system (IPAN) and accompanying image reconstruction analysis are apparent.

FIGURE 6

SYSTEM COSTS AND COMPUTATIONAL REQUIREMENTS

The IPAN system cost differential relative to a standard PAN unit of the same size is approximately 10% - 15%. This cost differential is produced by the greater number of He³ counters and associated counting electronics required as well as the more complex rotational assembly and matrix measuring instrumentation. The additional computational complexity required by the image reconstruction algorithm is readily achieved in present generation 25 - 33 MHz 386/486 personal computers. We have demonstrated that the required, typical "number crunching" associated with routine use of this technology can be accomplished in about two minutes per measurement using a 33 MHz 386 type personal computer.

SUMMARY AND CURRENT IPAN SYSTEM STATUS

We have demonstrated both improved assay sensitivity (0.2 mg ²³⁹Pu in low absorption waste drums) and improved assay accuracy (300% average for the complete spectrum of typical TRU wastes) with our prototype **IPAN** assay system. These improvements considerably extend the capability of previous state-of-the-art PAN assay technology.

In addition, our IPAN assay system's high active neutron detection efficiency (10 %) can be utilized to provide a high sensitivity "active neutron co-assay" of ²³⁹Pu and ²³⁵U for waste mixtures containing both fissile isotopes. This co-assay can be performed accurately for all ²³⁹Pu/²³⁵U ratios in the waste, with assay sensitivities in the mg-to-sub-mg range for both isotopes — singly or in mixtures. This co-assay capability does not exist in PAN systems based on the older technology.

We are also now implementing a recently developed, patented neutron coincidence and neutron multiplicity processor (Los Alamos National Laboratory) in the IPAN system. This processor is considerably advanced from the previous state-of-the-art and provides individual neutron multiplicity analysis as well as total rate and coincidence rate data. With this system, for example, we will be able to perform accurate passive neutron co-assays of waste mixtures containing ²⁴²Cm, ²⁴⁴Cm and ²⁴⁰Pu. This capability does not exist in older shift-register based passive neutron coincidence systems.

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