EXPERIENCE OPERATING LANL'S MOBILE PASSIVE/ACTIVE NEUTRON (PAN) ASSAY SYSTEM

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ABSTRACT

We present a summary of our operating experience with LANL's mobile PAN\textsuperscript{1} assay system. This system was acquired from the Carlsbad Area Office in 1994, refurbished, calibrated and fielded for the first time on LANL's TRU waste in the winter of 1996. It is functionally identical to other PAN systems throughout the DOE complex and its software operating system is the same as that used at INEL.

Since January 1996, it has passed the first round of the Performance Demonstration Program (PDP) and has been used to assay several hundred drums of LANL's TRU waste. We will report on the difficulties experienced assaying homogeneous wastes with high (\(\alpha,n\)) neutron fluxes. We will also report on our experience assaying debris waste in both the active and passive modes of PAN operation.

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INTRODUCTION

Since the last NDA/NDE conference, we have had an opportunity to use LANL's Mobile Passive/Active Neutron Assay System for the first time to assay a variety of LANL TRU wastes
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and to participate in the Performance Demonstration Program. We intend to use the PAN to begin certifying waste for shipment to the Waste Isolation Pilot plant along with our Tomographic Gamma Scanner (TGS) and newly acquired Canberra High Efficiency Neutron Counter (HENC). This past year's experience has been invaluable in pointing out the relative strengths and weaknesses of our various assay systems and has led to a clearer idea of how waste will be routed from storage to the correct instrument to perform the certifying assay.

WASTE ASSAY

National Transuranic Program Office Project

Our first experience using the mobile PAN on TRU waste at LANL was in support of a RCRA characterization project in the winter of 1995/1996. The goal of the project was to obtain RCRA characterization data of four LANL TRU waste streams to support WIPP's permitting process. The wastes selected for characterization were cemented sludges and pyrochemical salts.

The assay results were characterized by large uncertainties due to the very large \((\alpha, n)\) neutron signals produced in the low-Z waste matrix. An effect of these large \((\alpha, n)\) neutron signals is indicated in Figure 1. This plot of gross coincidence count rate (before correction for accidental coincidence events and dead time) versus totals count rate shows that the \((\alpha, n)\) contribution to the coincidence signal generally pushed the assay into a regime which most closely resembled a purely singles \((i.e.- (\alpha, n))\) neutron source. In fact, the totals count rate generally greatly exceeded what would be expected from a drum.
loaded with 200 grams WG-Pu (6% $^{240}$Pu) metal. The resulting effect of high singles neutron detection rates on assay precision and accuracy has been observed and simulated by others$^{2,3}$.

Figure 1 - Effect of High ($\alpha, n$) neutron signal on assay results. Labeled lines are expected results from pure $^{252}$Cf or $^{240}$Pu fission events or pure single (i.e.- ($\alpha, n$)) neutron events.

For these wastes, one of the basic limitations of our mobile PAN system was its electronics. It uses gated scalars (in the passive mode) which are limited in their ability to reject unwanted single neutron events when they are detected at high count rates. In fact, beyond event detection rates of about 1000 per second the gross passive coincidence count rate (uncorrected) begins to approach the totals count rate (the basic discrimination mechanism breaks down).

We have several options to circumvent this problem in the future. One is to use the Tomographic Gamma Scanner. We found
that we obtained quite reasonable results\textsuperscript{4} using the TGS on these waste drums and others which were assayed during the summer of 1996. The TGS also provides a direct assay of the $^{241}$Am which was present in non-equilibrium quantities (not due to ingrowth from the $^{241}$Pu content of the waste). Another option is to assay these wastes using the High Efficiency Neutron Counter. This system has superior electronics for discriminating against unwanted (α,n) neutrons. Finally, we are planning to install shift register electronics on our existing PAN systems (following the lead of our colleagues at INEL) which also are intrinsically better able to discriminate against unwanted neutron signals.

Summer 1996 Debris Waste Assay

During the summer of 1996, we had an opportunity to assay several hundred drums of mostly debris waste. From the point of view of (α,n) neutron production, these wastes were far more benign and amenable to assay using our PAN than the cemented sludges and pyrochemical salts assayed during the winter. These assays provided us with information regarding the neutronics properties of LANL's debris wastes as well as a cleaner comparison of PAN assay results to declared Pu content than we obtained earlier.

During a PAN assay, the instrument "assays" the waste matrix as part of the process of quantifying Pu content. A global average quantity representative of the waste matrix moderating and absorbing characteristics is determined and subsequently used to correct for the effect of these processes on the final assay result. Figures 2 and 3 present the values for the "moderator index" and "absorber index" determined for the debris wastes. For comparison, the corresponding values of the same parameters
determined for the PDP test drums used to date are plotted as well.

Figure 2 - Absorber indices of debris waste drums assayed using LANL's mobile PAN. Corresponding values for PDP drums used to date are indicated by heavy vertical lines.

Figure 3 - Moderator indices of debris waste drums assayed using LANL's mobile PAN. Corresponding values for PDP test drums used to date are indicated by heavy vertical lines.
Figure 4 compares the PAN assay results obtained in the passive mode with the declared values from the waste generator. Generally the agreement is good. In fact, the sum of the assay values for this set of drums exceeds the declared value by only 3.5%. This may be fortuitous though. Closer inspection of the PAN assay results shows evidence of a slight bias (10-15%) low relative to the declared values with the majority of PAN assay results coming within 25-30% of the centroid of the distribution. In full production, outlyers would be reassayed on the TGS to pin down the cause of the apparent difference between the PAN assay result and generator's declared value.

![Figure 4](image)

Figure 4 - Comparison of PAN assay results obtained in passive mode with generator supplied estimate (declared value) of Pu content of waste.

The active assay results (not shown) for the same waste drums do not exhibit the same behavior. Thermal (active) neutron assay results are more sensitive to matrix and self-shielding effects than passive assays. More analysis of the active data is required to explain the observed results.
Our PAN assay experience in the PDP tests to date is shown in Figure 5, along with the results of measurements made with the TGS and HENC. The PDP tests to date have concentrated on drum loadings in the range of 0 to 10 grams of weapons grade Pu (94% $^{239}\text{Pu}$, 6% $^{240}\text{Pu}$). A consequence of this is that the results reported so far have all been active assay results (the operating software forces this in its default mode). All of the results obtained with the PAN have been biased high. One reason why the PAN assay results are biased high may be that our calibration standard is too thick. We have used a depleted uranium bar provided with our fixed PAN system for active calibration. Unfortunately we have neglected to compensate for its ability to self shield. Self shielding would result in a reduced fission yield in our calibration standard for a given incident thermal neutron flux. Then, if the PDP standards are effectively "thinner" than our calibration standard we would obtain a larger fission yield for the same thermal neutron flux biasing our PDP results high. To test this possibility a series of Monte Carlo calculations will be run simulating neutron transport through the depleted uranium bar which we use to calibrate the active mode.
Figure 5 - Summary of PDP assay results for LANL's three assay systems. The data at actual grams equaling 1 and 9 grams are our best estimate (based on the concurrence of our three measurements) of what is in the drums. The data at 3.66 grams and 6.3 grams is accurately plotted based on the report of the first PDP round results.

This points to a general problem one has when assaying using the active mode. Without exact knowledge of the Pu particle sizes in actual waste, it is impossible to specify calibration standards which are equivalently self shielding.

The result of an active PAN assay is more strongly affected by moderation and absorption by the waste itself than passive assays are. At the same time as we are evaluating the $^{239}$Pu equivalence of our active mode calibration standard, we will be revisiting these and other effects to arrive at the appropriate calibration.
CURRENT ACTIVITIES

We are currently reviewing the entire calibration of the mobile PAN both in active and passive modes. Our first calibration was made using $^{252}$Cf for matrix corrections and PuC "pins" for zero matrix calibration in the passive mode. We plan to recalibrate using the PuC "pins" for matrix correction as well. The $^{239}$Pu equivalence of our depleted uranium standard will be referenced to a "thinner" PuO$_2$ standard for active assay calibration.

As we are acquiring the data needed for passive mode calibration, we will be mapping out in some detail the PAN response to point sources at various positions in a set of mock waste drums. Our existing $^{252}$Cf data indicates that in the course of acquiring this data, we will accumulate a fairly complete mapping of neutron coincidence count rate versus moderator index (Figure 6). A function can be fit to the distribution of results to correct for lost counts due to moderation of neutrons in the waste. This function can then be used to estimate the contribution from neutron moderation in the waste to the total uncertainty in a passive assay.
Figure 6 – Passive neutron coincidence count rate versus moderator index. The solid line represents what the PAN software is currently programmed to expect as the reduction in coincidence count rate due to moderation and correct for. Each symbol represents a particular mock waste drum.

FUTURE PLANS

Once we have re-evaluated our current calibration and made any indicated changes, we will begin assaying waste to meet the needs of a characterization audit scheduled for March 1997. After this audit we will complete implementation of an isotopics capability to support all three of our assay systems. This isotopics package will enable us to verify generator provided isotopics information, establish statistics on the variability of isotopics within a given material type and allow us to assay for $^{241}$Am by establishing a scaling factor to the Pu content of the waste, then assaying the Pu on the PAN.
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REFERENCES


