HIGH SENSITIVITY IMAGING PASSIVE AND ACTIVE NEUTRON (IPAN) TRANSURANIC ASSAY SYSTEM

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ABSTRACT

A group of nuclear scientists and engineers at the Los Alamos National Laboratory (LANL) developed and patented the original Passive Active Neutron (PAN) assay technology in the 1978-85 time period[1]. Several of these original inventors, led by John Caldwell and Walter Kunz, formed Pajarito Scientific Corporation (PSC) in 1986 as a technology transfer company specializing in implementation and further development of the PAN assay technology. This paper describes PSC's most recent developmental work in PAN assay technology—the use of neutron signal imaging to significantly improve assay sensitivity and accuracy.

The primary sources of error in PAN assay systems (as well as in all other NDA systems used to assay bulk wastes) are all directly or indirectly related to non-uniform measurement responses associated with unknown source spatial distributions. These errors can be significantly reduced when the exact spatial locations of sources are known. We accomplish this significant error reduction with our imaging technology—which we are implementing currently in 208 l drum and 1.5x1.5x2.5 m large box size assay systems. Initial customer delivery of both sized systems will occur in 1993. High resolution passive gamma ray detectors are integrated into two initial customers' systems as well.

We have performed an extensive series of fissile source distribution tests in 10 widely disparate waste matrices (40-50 separate distributions for each matrix, 200 sec measurement time) using PSC's prototype 208 l drum unit. In these tests we found an average improvement in assay accuracy of 300% for all matrices. This assay accuracy improvement is relative to a state-of-the-art PAN assay system that does not utilize source imaging. Our imaging hardware and analysis algorithm are robust and function well at low levels of fissile contamination. In a 300 kg concrete rubble matrix, for example, almost the same level of assay accuracy improvement was found for sources as small as 10 mg ^{239}Pu.

PSC has improved routine pulsed active (differential dieaway) measurement sensitivity by more than an order of magnitude in recent years. Our most recent work in this area is the use of active coincidence measurements to achieve a large signal-to-background improvement.

INTRODUCTION

Eight 208 liter drum-size and three large box size PAN units of the Los Alamos Group N-2 type were put into service at various USDOE sites during the time period 1982-86[4]—and are still in active operation at the present time. Much of the transuranic (TRU) wastes destined for ultimate disposal at the Waste Isolation Pilot Plant (WIPP) have been assayed in these units. The active assay in each of these 11 LANL, Group N-2 type PAN systems is performed with a Zetatron/MA-165C pulsed neutron generator. This is a very reliable, low background and long lived neutron generator system—suitable for routine high sensitivity waste assay measurements. Four or five year operating lifetimes for zetatron tubes have been common at USDOE sites. The field performance record of the complete assay systems has also been highly satisfactory.

The routine lower limit of detectability of the original 208 l systems is in the <10 mg ^{239}Pu range. These assay units also incorporated a sophisticated matrix correction formalism—based on an automatic measurement of each drum's average matrix moderator and absorber characteristics[2]. A recent systematic evaluation of this matrix correction formalism has been performed at the Idaho National Engineering Laboratory[3]. This evaluation intercompared assays obtained during a five year period of more than 5000 plutonium waste drums in segregated sets of 26 distinct matrix types. The intercomparisons were between the separate passive and active portions of the PAN system and SGS type passive gamma assays of the same drums performed at another USDOE site. The conclusion of the study is that ..."The Passive-Active Neutron Assay System is very versatile, yielding good results in the assay of nuclear waste of a wide variety of compositions[3]..."

The study identified some areas in which PAN technology improvements would be especially valuable. These included, as the primary source of active assay error, source location dependent matrix effects.

IMPROVED ASSAY ACCURACY USING IMAGING

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 or indirectly related to the non-uniform measurement responses associated with the unknown spatial distributions of the nuclear materials within the waste package. These errors vary greatly in magnitude from one waste matrix type to the next and can amount to a factor of two or more in difficult matrices. A number of years ago PSC initiated development of a new type of PAN system which addresses this primary source of assay error in a direct fashion. We call this approach **"Imaging Passive-Active Neutron" or IPAN**.

In this approach the IPAN system detectors are arranged in a configuration and the IPAN 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 significant improvement in assay accuracy relative to the original PAN systems and all present state-of-the-art PAN systems that do not use imaging. The extensive testing we have performed with our latest 208 l IPAN prototype[7] demonstrated a 300% (on average) improvement in assay accuracy relative to even the "best available" non-imaging PAN system.

The data are 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) is 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. Note that separate passive and active imaging measurements are performed—leading to separate processed images.

The imaging algorithm performs 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.

Since 1987 we have built and tested two generations of 208 l and one generation of 1.5x1.5x 2.5 m size IPAN units, both at our Los Alamos pulsed neutron test facilities and at a USDOE site. We have simultaneously developed our imaging acquisition software and analysis algorithm to a commercial status during this same time period.

DRUM SIZE IPAN UNIT AND DESCRIPTION OF TEST MEASUREMENTS

We have completed an extensive series of waste matrix measurements with our first "commercial" 208 liter size IPAN unit, as a condition of customer acceptance testing. These tests consisted of a systematic set of distributed fissile mass measurements within each of 10 mockup waste matrices placed inside typical USDOE standard 208 liter, mild steel drums. The matrices were chosen to mock-up a large variety of waste forms commonly encountered in the USDOE as well as international nuclear processing facilities. These included cellulose-combustibles, 5% to 75% PVC-combustible mixtures, low and high borax content glassware, 250 kg of iron-steel scrap, 250 kg of 5 - 10 cm diameter concrete rubble and a drum filled with 450 kg of solid concrete[7].

One of the customer acceptance tests required PSC to properly image low level signals (equivalent to 10 and 15 mg ^{239}Pu) in specific locations within a heavy waste matrix (250 kg of concrete rubble) during a standard 200 sec active assay measurement. In more than 90% of the test cases (ex: 10 mg source at bottom center; 10 mg source at middle outside plus 15 mg source at top middle; 10 mg source at top outside plus 15 mg source at bottom middle, etc.) the sources were imaged in their proper pixel locations. In addition, in a related series of 24 measurements with the 10 mg source placed at various locations throughout the concrete rubble drum, a 200% improvement in assay accuracy was obtained relative to non-imaging measurements in the same locations. These low signal imaging tests (10 mg ^{239}Pu corresponds to 3 nci/g in this matrix) demonstrate the robust nature of PSC's IPAN system.

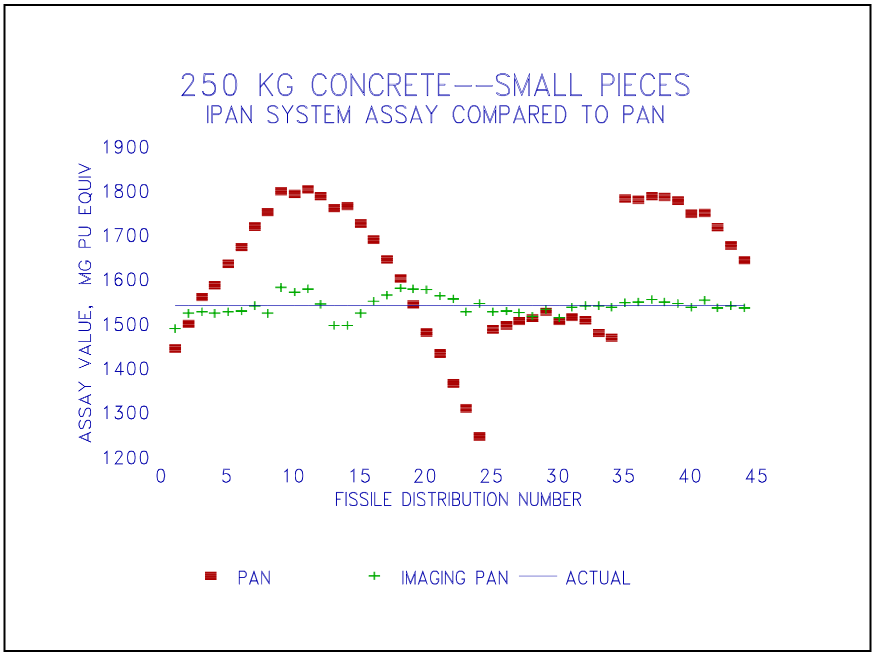

Figure 1: *Concrete Matrix Fissile Distributions*

Fig. 1 shows the results of more extensive and systematic fissile distribution measurement comparisons between IPAN and PAN ASSAYS in the same matrix[7]. The large general improvement in assay accuracy is apparent. Fig. 2 shows the compiled assay accuracy comparison results for all 10 test matrices[7]—with the observed average assay accuracy for the 40-50 test fissile mass distributions plotted as a function of matrix correction factor.

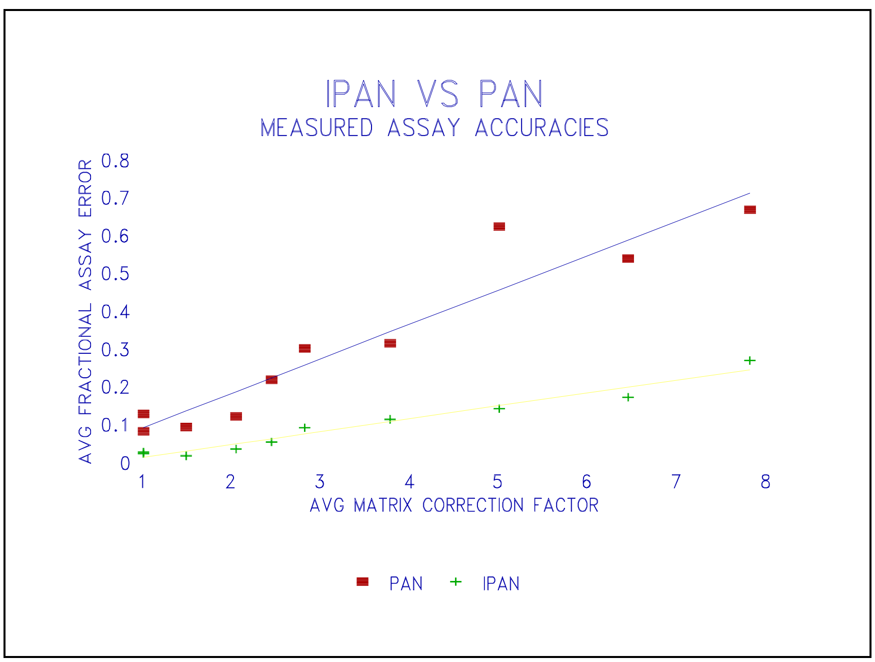

Figure 2: *10 Matrix Collective Distributions IPAN vs. PAN*

PSC'S IMPROVEMENTS IN ASSAY SENSITIVITY

One of the primary goals PSC identified for its PAN technology development program in 1987 was the significant improvement of minimum detectable fissile amounts. The original PAN units provided satisfactory sensitivity at the 10 mg ^{239}Pu level—but could not routinely be used for lower level assay situations. PSC's first generation improvements were motivated by and incorporated in the "INTEGRATED FIVE STATION NDA SYSTEM"[5,6] 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. The general level of quantitative ^{239}Pu assay sensitivity required by this D&D operation is <10 nci/g.

Incorporated in this system are two separate PAN units. One accommodates large glove boxes up to 1.5x1.5x3.0 m in size and one is used for assays of 208 l drums and small compressed waste bales. Both units incorporate PSC's improved (low background) Zetatron/MA-165C neutron generator housed within PSC's proprietary external "Moderating Assembly" (MA) neutron interrogation source. The elimination of "dark current" and "shot noise" neutron generator backgrounds is essential to the achievement of routine mg ^{239}Pu level assay sensitivity. The external MA addition produces a greater than 300% improvement in useful interrogation fluxes relative to the originally installed DOE site PAN systems. A third 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 three improvements (low noise pulsed neutron generator, increased useable neutron interrogation flux, lower detector backgrounds) results in a corresponding improvement in plutonium assay sensitivity—in both 208 l drum and large box size systems. At Erwin, for example, the (1.5x1.5X3.0 m) glove box PAN unit achieves routine 10 nCi/g performance and the drum-bale unit achieves a 2-4 nCi/g sensitivity—both of which represent about a factor of 10 improvement relative to the respective sizes of the original LANL PAN systems.

All three assay sensitivity improvements have been incorporated into the design of the 208 l and large box size **IPAN** systems discussed in this paper. We note also that the active coincidence/fission neutron multiplicity measurement capability (discussed below) is also incorporated into all of PSC's current IPAN systems.

PSC's most recent development is the addition of active coincidence measurements to achieve a large signal-to-background increase relative to the present state-of-the-art differential dieaway technology. This improvement is demonstrated in figures 3 and 4. Fig. 3 shows the usual gross active totals signal measured with PSC's prototype IPAN system. The gross active totals signal has been processed to remove all cosmic ray backgrounds. Note the "zero mass" active background. This residual response is due primarily to a small interrogation neutron penetration into the active detectors as well as to some matrix related singles neutrons response. This residual active background is what determines the active method lower limit of detection in state-of-the-art PAN systems.

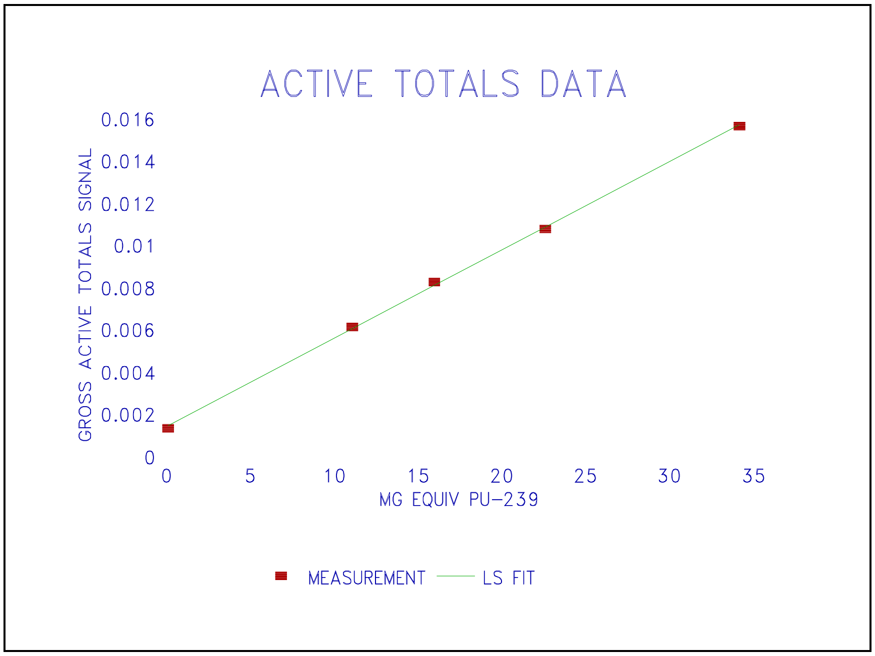

Figure 3: *Active Totals Data*

Fig. 4 shows the gross active coincidence signal measured simultaneously with PSC's prototype IPAN system. The gross active coincidence data has also been processed to remove all cosmic ray backgrounds and normalized to the same scale factor as the active totals data. Note first that the slopes of the active totals and active coincidence responses as a function of ^{239}Pu mass are identical. Note also that the "zero mass" response of the active coincidence measurement is virtually nothing. **Quantitatively the coincidence zero mass is 30 times lower than the totals zero mass.**

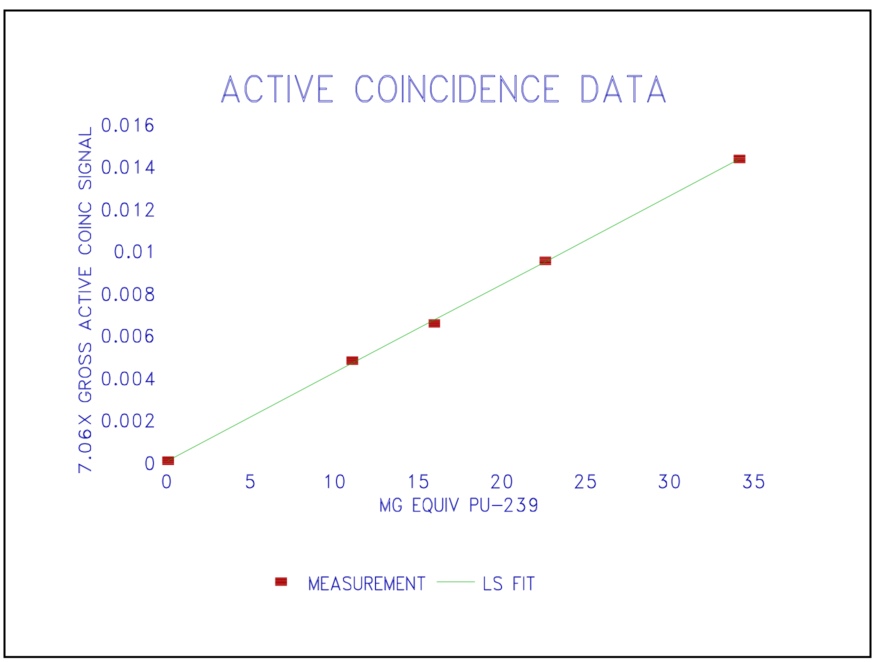

Figure 4: *Active Coincidence Data*

This latest development is particularly important for low level assays of normally difficult waste forms such as PVC and ferrous metals. Using active coincidence with our prototype 208 l IPAN system we have been able to demonstrate sub-mg ^{239}Pu assay sensitivities in both PVC and ferrous metals matrices. The use of a high performance neutron multiplicity sorter (discussed in a separate section) in conjunction with our active coincidence measurement also makes rapid, **mg level fissile co-assay of mixtures of ^{239}Pu and ^{235}U** a reality.

PROGRAMMABLE MULTI-CHANNEL COINCIDENCE MODULE—PMCCM

About one year ago PSC applied for and received a technology transfer patent license from LANL covering the commercialization of a just developed programmable multi-channel coincidence module[8]. We have successfully commercialized this module, including acquisition and analysis software, and incorporated it as a routine element in our IPAN systems. The module is described in the indicated LANL publication. Because it processes coincidence data through 15 parallel and synchronized shift registers this module reduces older "shift register coincidence unit" dead times by approximately a factor of 15. In addition this module obtains the "accidental coincidence" neutron data in an entirely different fashion from the older shift register modules—leading to a large reduction in statistical errors.

However, the most significant new feature provided by the PMCCM is that of routine, high count rate neutron multiplicity analysis. This feature makes possible **prompt fission neutron co-assay** in both passive (i.e., co-assay of ^{240}Pu and ^{244}Cm) and active modes (i.e., co-assay of ^{239}Pu and ^{235}U). PSC has demonstrated both passive and active co-assay with its 208 l IPAN system and has incorporated routine passive and active PMCCM measurements in all its commercial IPAN units.

COMBINED IPAN AND PASSIVE GAMMA SYSTEMS

Two of PSC's initial commercial IPAN installations include a combined IPAN and high resolution passive gamma feature. One system is for 208 l and 400 l size waste drums weighing as much as 1000 kg (Fig. 5) and the other is for 1.5x1.5x2.3 m boxes (Fig. 6) weighing as much as 7000 kg. Both systems will also feature a computer controlled drum/box conveyor system. All data—neutron and gamma—is acquired with integrated software and analyzed in a combined fashion. Both systems are scheduled for customer delivery in 1993.

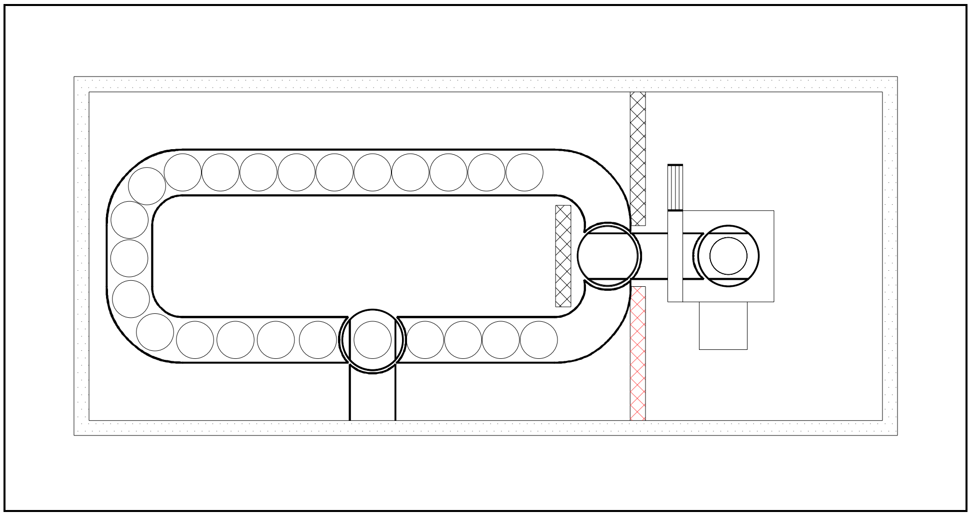

Figure 5: *Automated/Computer Controlled Drum Conveyor System*

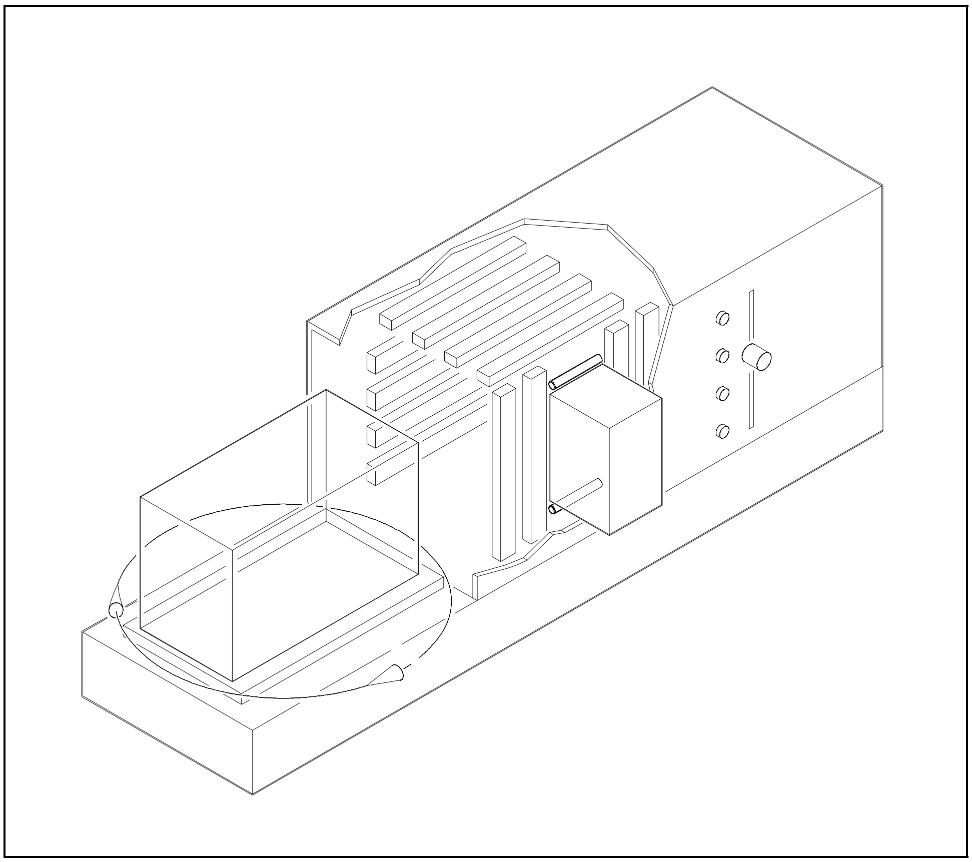

Figure 6: *Combined IPAN/GEA Crate System*

SUMMARY

We have demonstrated both improved assay sensitivity (as low as 0.2 mg ^{239}Pu 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 passive and active neutron detection efficiency can be utilized to provide a high sensitivity co-assay of ^{239}Pu and ^{235}U in the active mode and for ^{240}Pu and ^{244}Cm in the passive mode—through the use of our new high count rate PMCCM module.

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