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First Tests of A Portable Plutonium Mass Verification System

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Special Nuclear Material (SNM) accounting at some DOE facilities requires removal of samples from storage, transport to a counting facility, removal of the sample from its storage container, counting, repackaging, and transport back to the storage facility until selected in the next physical inventory. This procedure requires personnel time, radiation exposure, and increases the potential for diversion. If a verification can be performed within the storage vault or bunker and not require removal of the SNM from the container, personnel time and radiation exposure could be reduced, the sample would not have to leave the security of the storage vault, and costs for performing inventories could be reduced.

Lawrence Livermore National Laboratory (LLNL) is developing an instrument, based on established coincidence counting techniques, to yield a semi-portable Pu mass verification solution to the problem of in-vault verification. Commercially available instrumentation is being tested that will enable in-situ verification of Pu masses in sealed containers. The detector assembly includes both neutron and gamma detectors. The device will operate on battery power, and will be mounted on a cart for portability.

For mass verification of plutonium, two types of signal are analyzed. A gamma spectrum is analyzed for isotopic ratios using LLNL's MGAHI code. Neutron count rates and coincidence rates are measured to quantify the spontaneous fission rate within the Pu. Corrections for neutron scattering, alpha-neutron reaction sources, and induced fission rate must be made to establish the necessary accuracy for this analysis. The system is intended to be easy to set-up and operate.

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Introduction

Quantifying Plutonium

To measure an absolute amount of plutonium by non-destructive means typically requires two measurements. A gamma spectrum analysis can determine relative amounts of isotopes, and another measurement (specific power or neutron emission rate) is used to determine the quantity of one or more of the isotopes present. For this project, neutron coincidence counting is used to determine the amount of one of the plutonium isotopes. Together, the two measurements will enable the total amount of plutonium present.

The goals and challenges of this project

The purpose of this work is to estimate the amount of plutonium in a sealed container, at its storage location. The end result is a verification-level amount, so the accuracy requirement isn't too difficult to meet. However, other requirements pose problems:

- The samples are in sealed containers, so the exact distance from sample to detector is unknown.
- The samples are in low-Z packing materials, the densities of which can vary from container to container, so neutrons will be scattered differently from one container to the next.

- Characteristics of the neutron source terms (i.e., relative amounts of neutron fluence from alpha-neutron, spontaneous fission, or multiplication) can also vary from one sample to the next.
- The containers attenuate the gamma rays coming from the plutonium, so the lower energy gamma energies are unavailable for analysis.
- The samples will be analyzed while in their storage locations, so the background will be high, and contributions from nearby samples will have to be considered.
- The end user requires the apparatus to operate from battery-power.
- The end user has a twenty-minute target for count times.
- The user interface has to be easy to learn and use.

The ability to determine amounts of plutonium in sealed storage containers, while still in storage, offers several advantages. Not having to move the sample to the counting facility reduces handling, so radiation doses can be minimized. The elimination of movement of the samples would minimize chances of diversion or damage during transport, and would save time.

Work to date

This paper deals with the initial system tests on the neutron subsystem, as implemented on NIM electronics. It presents the results of this first phase of work.

Theory

The mass of one of the isotopes of plutonium present in the samples of interest to this project is proportional to the spontaneous fission rate of that isotope. The number of neutrons per spontaneous fission of plutonium is ~2. The spontaneous fission rate of the sample can be determined by the use of two counters to measure coincident neutrons from the sample. According to Knoll [1], the source strength, S, is determined by the following:

$$S = \frac{r_1 r_2}{r_{12} - r_{ch}} \qquad (1)$$

where r_1 is the singles count rate from detector 1, r_2 is the singles count rate from detector 2, r_{12} is the measured coincidence rate, and r_{ch} is the chance coincidence count rate, which can be calculated from the two counter's singles count rates and the time duration of the coincidence window, tau (τ), as follows:

$$r_{ch} = 2\tau r_1 r_2 \qquad (2)$$

The above equation for source strength was derived with the assumption that the radiation measured in coincidence was emitted isotropically. This is not the case for neutrons produced by fission, as can be seen in Figure 1 (from Reference [2]). To address this concern, the equipment has been designed to minimize the effect of the anisotropy.

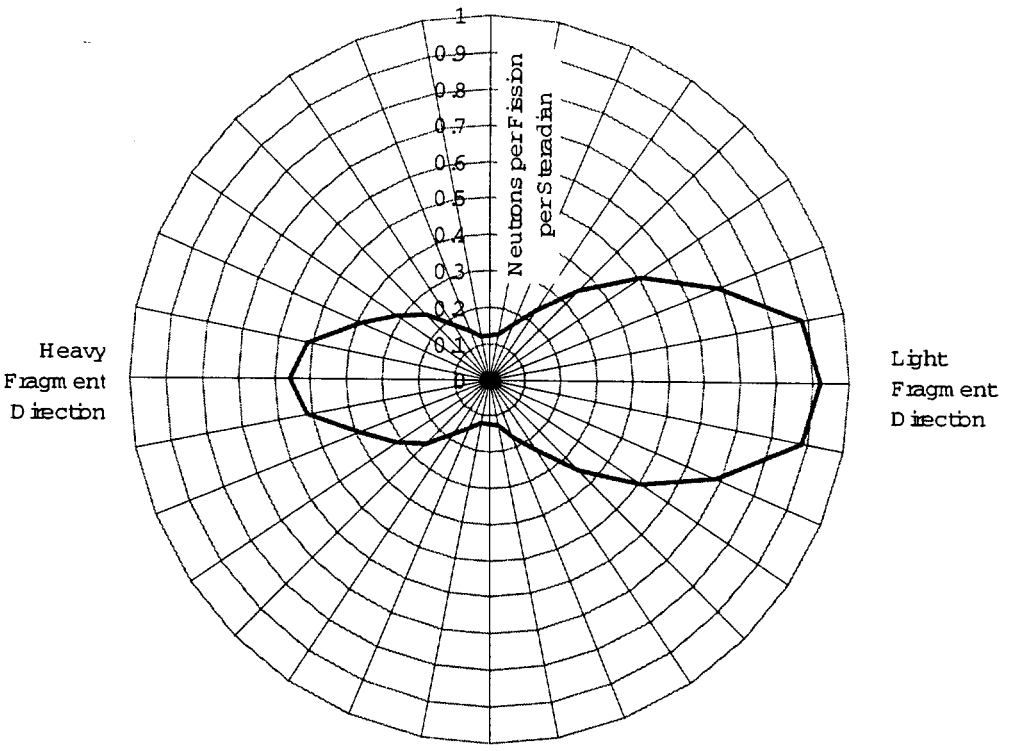

Figure 1. Angular Distribution of Neutrons from Spontaneous Fission (^{252}Cf)

Experimental

Equipment

As shown in equation (1), the two detector count rates and the coincidence count rates must be measured to calculate the source strength. Nuclear instrument module (NIM) electronics were selected to support the coincidence counting, as shown in Figure 2. Two trains of coincidence pulses are used, because this allows for correction of effects from scattering (packing) materials in the storage containers. This technique will be discussed later.

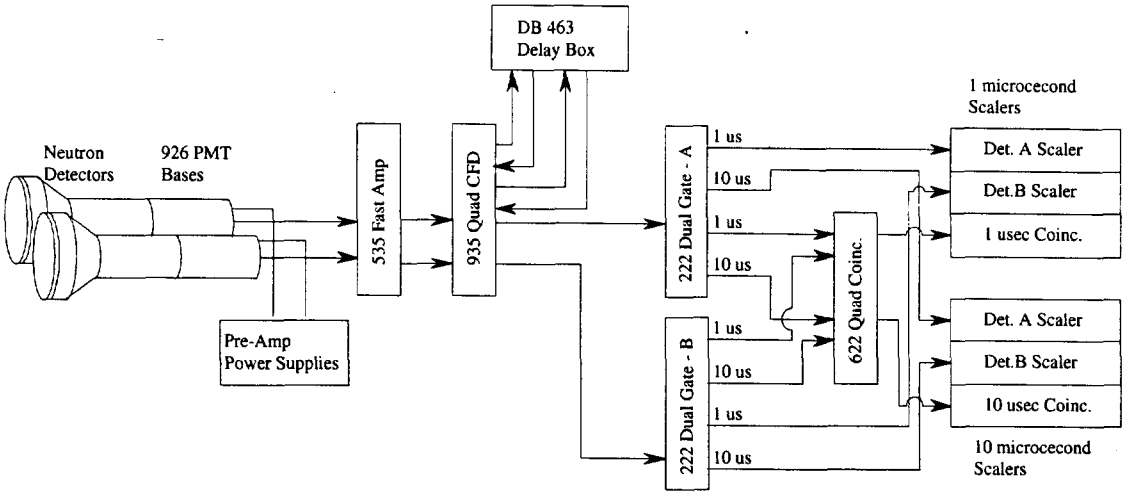

Figure 2. NIM-based Neutron Subsystem for the Portable Plutonium Mass Verification System.

The neutron detectors are ^{6}Li-doped, cerium activated glass scinitllators, available along with their photomultiplier tubes (PMTs), from Saint Gobain (Bicron). Each detector contains about a mole of ^{6}Li, and is 98% effective in detecting thermal neutrons perpendicular to the detector faces. A ^{6}Li-based neutron detector was chosen because of the high Q value for the ^{6}Li(n,t)α reaction. The high Q value of 4.78 MeV facilitates gamma discrimination. The detectors are also chosen for their compact size (to minimize the effects of the angular dependency of neutron emission probability), and ease of transportability. These detectors are also directional, which minimizes contributions to the count rates from samples other than the one being counted.

The electronics used in these first tests are standard NIM devices, available commercially. The PMT bases are PerkinElmer/Ortec 926 combination HV power supplies and preamplifiers. Power is supplied to these from the preamplifier power output from two spectroscopy amplifiers (the type providing the –12VDC is not important). The signals from the PMT bases go to a PerkinElmer/Ortec 535 fast amplifier, and thence to a

PerkinElmer/Ortec 935 quad constant fraction discriminator (CFD). The delay required for the CFD operation is provided by a PerkinElmer/Ortec DB 463 delay box. The logic-pulse output from the CFD goes to LaCroi 222 Gate and Delay Generators, one for each detector. From these two gate and delay generators, both a one-microsecond and a ten-microsecond logic pulse is sent to the scalers and to the coincidence amplifier. The coincidence amplifier is a LaCroi 622 Quad Coincidence Amplifier. It outputs two sets of coincidence pulses, one for the one-microsecond pulses, and one for the ten-microsecond pulses. Six Tennelec 526 Scalers count the single and coincidence pulses. Three of the six scalers count the one-microsecond pulses from the two detectors and the pulses from one-microsecond coincidences, and the other three are for the ten-microsecond pulses. Timing was verified using a Berkeley Nucleonics PB4 precision pulser.

The gamma isotopics system is a PerkinElmer/Ortec Safeguards detector run from a DigiDart™. The spectra are analyzed by MGAHI, a program for the analysis of shielded pltonium isotopics, developed by LLNL, and currently being licenced by PerkinElmer-/Ortec. The gamma subsystem will be further described in following papers. The MGAHI algorithm is described in reference [3].

Counting Parameters

Samples of known mass were counted at two DOE facilities. Materials measured included actual samples, like the target population for this measurement system, as well as unpackaged pieces and calibration sources. The count times were 20 minutes per sample, and the detector-to-sample distances were typically from 9 to 18 inches. Background singles and coincidence count rates were measure in the same locations as the sample counting, but with

the samples removed from the area. Singles and coincidence counts were recorded, as well as mass and amounts of scattering material (when known).

Results

Initial Calibration

For simple systems with known amounts of scattering materials, a relationship between the calculated source strength and mass was determined. A plot of calculated source strength versus mass, for a particular amount of scattering material, is shown in Figure 3. As can be seen, the error bars (propagated errors at 1-sigma) get smaller as the masses get larger. At the "masses of interest," the errors are near or within the accuracy specification requested by the end user.

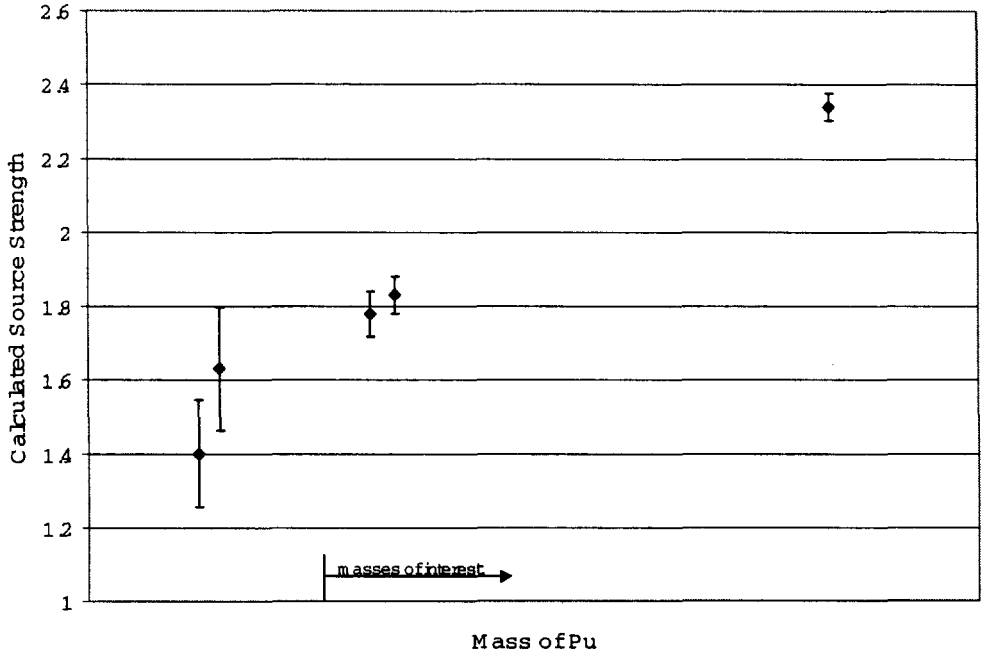

Figure 3. Relationship between Corrected Counting Data and the Mass of the Sample (for a constant amount of scattering material, as determined by the ten-microsecond count rates).

Scattering Corrections

These initial counting efforts showed the importance of being able to correct for the scattering effects of the packing materials. The curve described by the points in Figure 3 changes when a different amount of scattering material is between the sample and detector. The calculated mass of plutonium varied with the amount of scattering material. We needed to find a way determine the amount of scattering, and correct for it.

A difference between the calculated source strengths for the one microsecond pulses and the ten microsecond pulses provided a means by which the effective amount of scattering material could be measured. In Figure 4, the ratio of the calculated source strengths is shown as a function of polyethylene thickness. By computing this ratio for a sample in a container, the effective amount of neutron-scattering material can be determined.

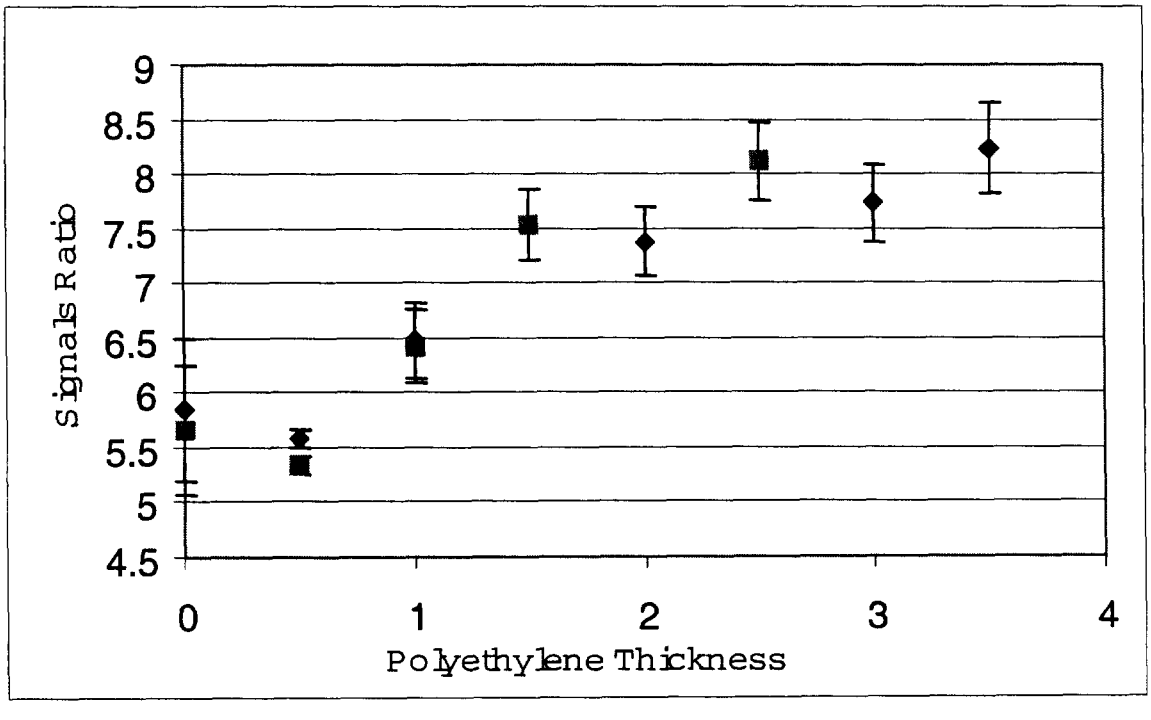

Figure 4. Ratio of calculated Source Strengths versus Polyethylene Thickness. (Squares are the results from one data set, diamonds from another).

Following the determination of the effective polyethylene thickness, a correction for the scattering effects is used. The scattering effects on the three count rates (the two detectors' singles rates and the measured coincidence rate) were determined in a series of experiments, and correction curves generated. In Figure 5, a set of correction curves is shown. Note that the correction curves are relatively flat above an effective polyethylene thickness of 1.5 inches. To use this to the advantage of the analyst, sufficient polyethylene can be placed in front of the detector to bring the total amount of scattering material (in the container, and right in front of the detectors) on to the flat portion of the correction curve. That way, the effect of errors in the determination of the amount of scattering materials between the source and the detectors can be minimized.

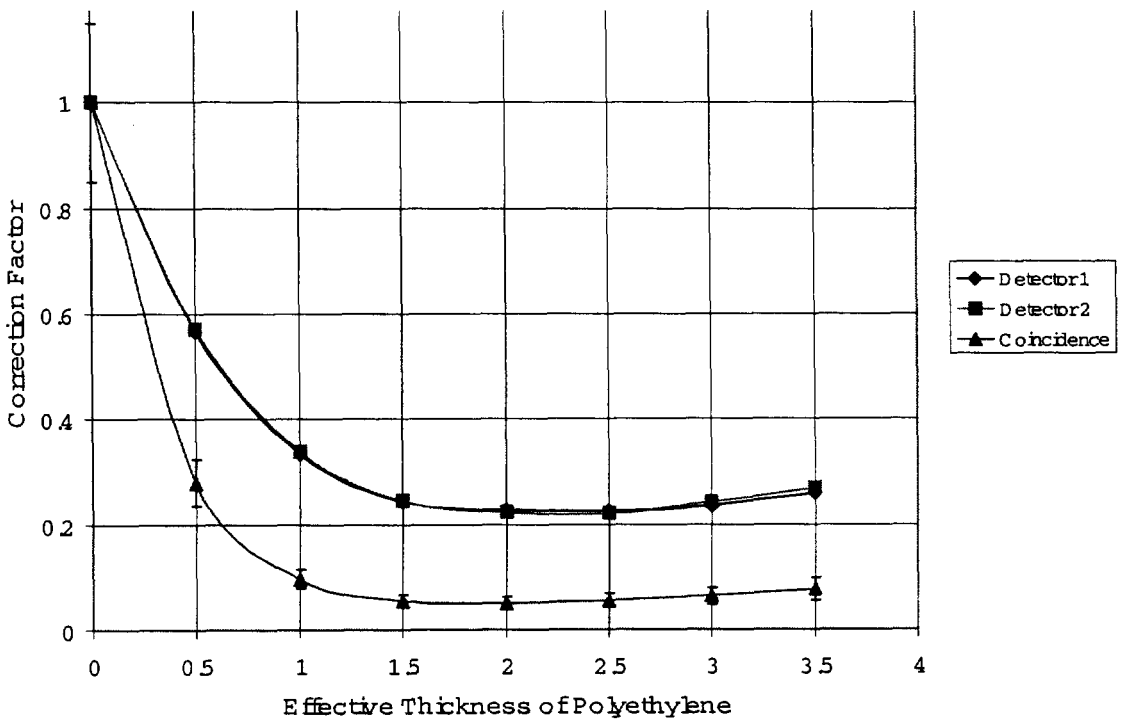

Figure 5. Correction factors for the 10 μs counts

To get a result, software calculates the effective amount of scattering material for the detector (as in Figure 4), and determines correction factors for the three count rates (Figure 5). With the corrected count rates, an accurate source strength (equivalent to the mass of the spontaneously fissioning isotope) is calculated.

Conclusions

We determined a relationship between the amount of neutron scattering material and system response. We also developed a technique for correcting the count rates for the scattering material. The initial results indicate that this is a satisfactory technique for verification of plutonium mass in sealed containers.

Future Work

A series of tests is being arranged at a DOE facility to determine the robustness of the method to actual conditions, and to further test this measurement technique. Counting parameters to be varied include detector to container distance, amounts of scattering material, and thickness of shielding. Samples chosen will vary in mass, as well as neutron source characteristics (alpha-neutron and multiplicity contributions). Results of the analysis of these data will be in a future paper.

The NIM-Based neutron subsystem is being replaced with a compact, low power system based on a National Instruments PXI™ computer and fast ADC. For this system, it is the current intent that neutron events will be stored in list mode, which would enable greater flexibility in data analysis (variable time windows for coincidence, possible multiplicity

corrections, et cetera). The PXI computer will also enable a much smaller system, capable of running both the software for neutron source strength determination and gamma spectroscopy software. This computer will be fielded with a touch-screen interface for the operator. Because the PXI computer is runs Microsoft Windows™, the gamma isotopic software will be run on the PXI, and the gamma subsystem will be integrated into the PXI-based neutron subsystem, so that total plutonium mass can be calculated.

References

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2. Bowman, H. R., Thompson, S. G., Milton, J. G. D., and Swiatecki, W. J., (1962) *Phys Rev.* 126, 2120
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