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A HIGH-EFFICIENCY, MODULAR SYSTEM FOR THE ASSAY OF PLUTONIUM IN LARGE WASTE PACKAGES

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A 4 π coincidence neutron counting system with nominal assay chamber dimensions of 1.2 by 1.2 by 2.4 m and a 14% detection efficiency has been designed, constructed, and used to determine the plutonium content in large crates of miscellaneous nuclear waste.

INTRODUCTION

A problem of great economic and environmental concern to the nuclear industry is the determination of the transuranic (TRU) nuclide content in bulk wastes. No instrumentation currently exists to screen such wastes in large packages at the required burial limit sensitivity of 10 nCi/gm per gram of waste. (A typical waste crate is 1.2 by 1.2 by 2.4 m and weighs 1000 kg; about 100 mg of plutonium in a 1000-kg waste package results in 10 nCi/gm.) Such bulk waste is currently either assumed to contain more than 10 nCi/gm, which entails placement in expensive retrievable storage, or it is grab-sample assayed assuming matrix homogeneity, a procedure that can lead to serious errors in TRU-content estimates. We report here on a direct, large-package TRU assay system that we have designed, built, and used to assay plutonium content in typical large waste crates.

Our design goals included (a) portability—we are scheduled to use this system in assay campaigns at locations several hundred kilometers apart, (b) overall detection sensitivity of 100-mg of plutonium—our system uses neutron coincidence counting of ^{239}Pu to 4 π neutron detection efficiency of about 14% is required for a 100-mg plutonium sensitivity, and (c) flexibility—the waste crates to be assayed in our program are not of standard size.

CONSTRUCTED DESIGN

We incorporated these design goals in a system composed of six large, stand-alone, detection modules that may be assembled into large 4 π counting geometries to accommodate an assortment of waste package sizes. Each detection module consists of a 1.25-m-thick polyethylene box (see enclosure in which high-efficiency ^3He proportional counters are placed. The side of each detection module facing the cavity is constructed of 1.27-cm-thick polyethylene.¹ Each detection module has two chambers separated by 2.54-cm-thick polyethylene and is reinforced structurally with aluminum rods and anastrol. Seven ^3He proportional counters (four in one chamber, three in the other) that are 2-atm fill pressure, 5 cm in diameter, and either 1.83 or 0.91 m long are contained within the module.

The moderator/detector configuration has been demonstrated experimentally to maximize counting efficiency per ^3He tube.^{2,3} Four of the detection modules have external dimensions of 2.49 by 1.22 by 0.53 m and two have dimensions of 1.22 by 1.22 by 0.53 m. Figure 1 depicts the six modules of our nominal 4 π configuration in which the large modules form the sides and the small modules form the ends of an assay chamber with internal dimensions of 1.2 by 1.2 by 2.4 m. Our measured 4 π detection efficiency in this configuration for a

Fig. 1.
Modular 4 π neutron assay chamber.

bare californium source is 14%. Alternate detection module configurations (with lesser 4 π detection efficiencies) are easily arranged to accommodate packages to dimensions of 2.4 by 2.4 by 2.4 m.

TABLE I. 4 π DETECTION EFFICIENCY (%)

Figure 2 plots the measured efficiencies for a bare ^{252}Cf source placed at different distances from one end of the

Fig. 2.
Efficiency of four configurations of the 1.2 by 1.2 by 2.4-m assay chamber for a bare ^{252}Cf source moved along the 2.4-m axis.

assembly. The highest efficiency and best linearity are obtained with the six-module assembly. The worst case was an open-ended cavity followed by a unit using three large modules for sides and a roof and 1.27-cm-polyethylene ends. These data were taken during the construction phase of this project. The poor intermediate results actually caused us to construct the two half-sized end modules rather than rely on passive albedo alone. As can be seen in Fig. 2, the end modules result in a considerable flattening of the detection response as a function of spatial position, as well as in a significant increase in total detection efficiency. With the six-module configuration, we observe a detection efficiency of $14 \pm 1\%$ independent of spatial location.

Separate counting electronics channels are provided for each chamber in each of the six modules for a total of 12 individual logic-pulse outputs. These 12 separate logic-pulse signals are routed to our LeCroy 3500 data acquisition system through the use of two Kinetic Systems 3610 hex-scalers. These 12 signals are recorded and stored separately on permanent disk files. The relative singles count rates from different portions of the 4 π system are used for geometric and matrix compensation algorithms.

For instance, the location of a source within the assay chamber can be found from the ratio of count rates from opposite side or end modules. Figure 3 shows the ratio of the observed count rates from the two end modules as a function of source position along the length of the assay chamber. The ratios of the count rates for the two sets of opposite side modules are shown in Fig. 4. As can be seen, these ratios are strong functions of source position. Under favorable conditions, the three ratios serve to determine a source hot spot location to within a few centimeters along each axis.

Coincidence Neutron Counting

In waste crates, even isotopes of plutonium, principally ^{239}Pu , undergo spontaneous fission reactions that result in the emission of time-correlated neutrons. That is, the neutrons from ^{239}Pu are given off in discrete bunches rather than one at a time. (Typically, there is also a large time-uncorrelated source present in HXU wastes, which is associated with (α, n) and other singles only backgrounds.) The time-correlated neutron fraction can be used reliably to estimate the plutonium content in a waste package; the uncorrelated fraction is strongly dependent on physical and chemical composition and does not, in general, lead to reliable plutonium mass estimates.

The technique to separate the time-correlated fraction from the uncorrelated fraction consists of using the first neutron detected to start a time-multiplexed scale (MVS) scan. If the first neutron is followed by a second detection (or

Fig. 3.
Ratio of the count rates from the end modules
for a source moved along the 2.4-m axis.

Fig. 4.
Ratio of the count rates from the side modules
for a source moved horizontally across the
1.2-m axis at the center of the chamber.
The upper curve is the ratio of the count rates
for the top and bottom modules for a source
moved vertically from the bottom.

third, etc.) from a spontaneous fission reaction, this detection will have a strong time correlation with the initial detection. If the initial neutron is not part of a correlated event, then subsequent detections recorded during the MCS scan will not display a time correlation with the initial event.

Figure 5 shows a first-neutron-generated MCS scan taken with our 1.2- by 1.2- by 2.4-m 4 π system with a ^{252}Cf spontaneous fission neutron source inside. The MCS was taken with a LeCroy 3500 data acquisition system using the LeCroy 3521 MCS module with a time-per-channel setting of 5 μs . As can be seen in Fig. 5, the MCS count time-history drops rapidly at times near initiation, reaching a nearly flat response at later times (after 1200 μs).

These data have been subjected to a least-squares fitting procedure using the functional form $Ae^{-\lambda t} + B$. This very simple function appears to fit the data quite well. Figure 6 shows the same data with the least-squares fit constant B ($B = 8836$) subtracted. As can be seen, the assumed single exponential results in an excellent fit over at least a factor of 2.5 decay from the initial count rate. We have used this fitting procedure successfully with a variety of spontaneous fission sources (such as ^{252}Cf and ^{244}Pu) covering a fairly large range of correlated-to-uncorrelated ratios and total count rates. In all cases, the assumption of a constant uncorrelated

Fig. 5.
Multiscale time-history showing the decay
of ^{252}Cf fission neutrons in the assay chamber.

Fig. 6.
Plot of Fig. 5 data with least-squares fit
parameter B ($B = 8836$) subtracted from each point.

background and a single exponential decay for the correlated signal has produced excellent fits. The exponential fall-off of the correlated component is readily explained by the detection neutronics of our system: the probability of detection of several neutrons emitted simultaneously should fall off exponentially in time, with a $T_{1/2}$ characteristic of a given ^3He counter and polyethylene geometric arrangement.

WASTE MATRIX EFFECTS AND COMPENSATION TECHNIQUES

All neutron detection systems suffer degradation of performance when waste matrix materials are placed between source and detector. Hydrogenous materials generally are responsible for the greatest degradation factors. Our system is designed to minimize this effect. We measure a $\pm 10\%$ flatness of response for moderated ^{252}Cf neutron sources covering a range from 0 to 7.5 cm of polyethylene thicknesses around the sources. For moderator amounts greater than 7.5 cm, we have developed two compensation techniques. One is based on the observed $T_{1/2}$ values in the least-squares fits to the first-neutron MCS data discussed previously. We observe, roughly, a 20-ns increase in $T_{1/2}$ for each additional centimeter thickness of polyethylene equivalent added around a spontaneous fission source.

A second compensation technique is based on the differential energy sensitivity of the count rates in the inner and outer chambers of each module. (Increasing the moderator thickness around a neutron source changes the emergent neutron spectrum.) We observe, roughly, a 7% change in the inner-to-outer-chamber ratio for each additional centimeter of polyethylene placed around a ^{252}Cf source.

WASTE CRATE ASSAY CAMPAIGN

We recently completed an assay campaign in which eight large wooden crates of miscellaneous LPO waste were assayed

using our large 4π passive counting system. These waste crates ranged in enclosed volume from 0.85 to 1.61 m^3 and required setting the system up in a configuration in which the assay chamber dimensions were 2.4 by 2.0 by 1.5 m and the detection efficiency was 11%. The ^{240}Pu isotopic composition in this waste was known (5.5% of the total plutonium) but the total plutonium in any crate was not known. Thus, by measuring the ^{240}Pu content with coincidence neutron counting, a total plutonium mass could be determined.

Our coincidence neutron measurements of these crates were successful (plutonium calibration was obtained using known standards) and led us to conclude that: one crate contained $< 10 \text{ nCi/g}$ total plutonium, three contained plutonium in the 40-60 nCi/g range, and the other four contained plutonium in amounts well above the 100- nCi/g range. The estimated assay accuracy for these measurements was about $\pm 50\%$ for the $< 10\text{-nCi/g}$ crate and about $\pm 20\%$ for the other seven. We also determined that our ^{240}Pu detection limit in a crate was about 5 mg.

CONCLUSION

The large 4π passive neutron counting system that we have designed, built, and used assays or screens large LPO waste crates at the 10 nCi/g sensitivity level. The assay system accommodates a considerable variety of waste crate sizes; assay chambers of different internal dimensions are readily set up using the six self-supporting, individual detection modules from which the system is constructed. We carried out detailed measurements for two typical assay chamber configurations: (a) 2.4 by 1.2 by 1.2 m, 14% detection efficiency, and (b) 2.4 by 2.0 by 1.5 m, 11% detection efficiency. Neither configuration required support structures beyond the modules themselves.

We experimentally demonstrated a ^{240}Pu coincidence neutron detection sensitivity of 5 mg and a detection flatness of response of $\pm 10\%$ (or better) for all spatial locations within the assay chamber and for source moderation conditions ranging from 0 to 7.5 cm of equivalent polyethylene thickness. We used the system in an assay campaign in which individual container total plutonium amounts ranging from less than 100 mg to about 1000 g were determined, a 10^4 dynamic range.

Waste matrix effects on assay accuracy were investigated. The system requires no corrections for moderation effects up to 7.5-cm-equivalent thickness of polyethylene. Two independent techniques for moderation compensation beyond this thickness were developed and demonstrated.

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