LA-UR- 11-04081

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Author(s):	Sy Stange, Douglas Mayo, Rollin Lakis, and M. William Johnson
Intended for:	Institute of Nuclear Materials Management 2011 Annual Meeting

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EST. 1943

Form 836 (7/06)

Development of a Neutron POD Spectrometer

Sy Stange, Douglas R. Mayo, Rollin E. Lakis, and M. William Johnson

Los Alamos National Laboratory, Los Alamos, New Mexico

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Abstract

Los Alamos National Laboratory has a long history of research, development, test and evaluation, implementation, and installation of radiation detection equipment. The Nuclear Nonproliferation Division has done so in support of domestic and international safeguards, emergency response, nondestructive analysis, and criticality safety. One of the systems that we have developed and deployed for use in the field is the NPOD-III. The system is designed to perform correlated neutron measurements coupled with totals neutron counting, multiplicity, Feynman variance-to-mean, and Rossi-alpha analyses to determine the mass, multiplication, and alpha-n ratio, but many more capabilities have yet to be explored. We are developing an algorithm and determining the applicability of using the current design as a low-resolution neutron spectrometer, that may differentiate between different impurities present with alpha emitting materials. We are initially focusing on well-known 10-g PuO_2 standards that contain specific impurities. In addition we will be comparing other sources that have different chemical forms and impurity contents. A brief description of the detector, experimental constraints, and experimental results will be presented.

1 Introduction

No single tool is universally employed for neutron spectrometry. Rather, a variety of techniques have been developed to meet the requirements of specific applications. [1] These include threshold radioactivation, proton recoil spectrometry, Bonner spheres, plastic and liquid scintillators, and ionization chambers. However, few of the existing neutron spectrometry methods are well-suited to field measurements, which require a detector that is portable and produces prompt results. Some techniques, such as threshold radioactivation, have analysis procedures that are too time-consuming for field measurements. Others, including liquid scintillators, contain flammable materials, making their use potentially hazardous in uncontrolled environments. In addition, many neutron spectrometers possess undesireable levels of γ-ray sensitivity. The objective of this project was to develop a low-resolution neutron spectrometry system that would make use of portable ^{3}He-based detectors that are already present in the field for other

measurements. Such a system would provide additional information for field and treaty verification measurements, and would also be useful for facility measurements in which a portable system is needed.

A neutron spectrometry system would assist in the identification of impurities present in special nuclear materials (SNM). Nuclear radioisotopes, like all other nuclei with mass number A greater than $\sim$150, are unstable against α-decay. When low-Z impurities are present, α-particles may undergo (α, n) reactions with light nuclei. The average energies of neutrons produced through (α, n) reactions are a few MeV and are characteristic of the target nucleus. The concept on which our neutron spectrometer is based involves positioning detectors, consisting of ^{3}He tubes embedded in polyethylene slabs, so there are significant differences in the distances between individual tubes and a source of (α, n) neutrons. By analyzing the response profiles of the detectors to neutrons with known energies, it may be possible to develop an algorithm capable of analyzing the response profile of an SNM source of unknown composition and identifying the impurities present.

2 Experimental Methodology

The neutron spectrometer utilizes Neutron POD-III (NPOD) detectors, which are well-characterized systems that are frequently used in the field. An NPOD consists of a polyethylene slab with 15 10-atm ^{3}He tubes in embedded in the polyethylene in two rows, as shown in Figure 1. Data output is provided by an ethernet connector. The data stream contains 17 potential channels, 15 corresponding to the individual ^{3}He tubes, one dedicated to a 100 Hz clock tick, to be used in synchronizing multiple NPODs, and one pass-through channel.

Standard 10-gram PuO_2 sources with different impurities, as well as PuBe and AmLi, were used as sources of (α, n) neutrons to evaluate the suitability of the system for neutron spectroscopy. The impurities found in the PuO_2 sources and the average energies of neutrons produced through (α, n) reactions with 5.2-MeV α-particles (the average energy of α-particles emitted by plutonium [2]) are given in Table 1. The composition of source 67-000 was the subject of some uncertainty; institutional knowledge gave the composition as PuF_4, but it was unclear whether this was an impurity in a PuO_2 source or the complete composition. The average neutron energies expected from the PuO_2 sources ranged from 1.0 to 2.9 MeV; the inclusion of a PuBe source, which produces (α, n) neutrons with an average energy of 4.2 MeV, and an AmLi source, which emits (α, n) neutrons with an average energy of 0.3 MeV, served to extend the energy range.

As shown in Figure 1, the neutron spectrometry configuration used two NPODs, one placed with its narrow end facing the source and the other placed with its face directed towards the source. Both detectors were placed at a distance of 25 cm from the source with the detector centers 148.5 cm above the floor. A Shielded Neutron Assay Probe (SNAP) was also used for neutron counting. Its front face (excluding the polyethylene cover) was positioned 100 cm from the center of the source. Two 15-minute measurements were performed for each source, one with the polyethylene moderator plate attached to the SNAP detector and one without the moderator. The source was placed on a stand 142.5 cm above the floor. The data stream from each NPOD was collected in a binary file, which listed the timestamp and ^{3}He tube of each interaction and the 100 Hz clock ticks. In addition, both pass-through channels were used.

Type	Source ID	Impurity	Avg. Neutron Energy for 5.2 MeV Alphas [2]	Neutron Yield per 10^6 5.2 MeV Alphas [2]
PuO_2	67-000	PuF4*	1.2	5.9
	86-000	Oxide	1.9	0.059
	87-000	Al	1.0	0.41
	88-000	Mg	2.7	0.89
	90-000	B	2.9	17.5
	91-000	Fluoride	1.2	5.9
Other	PuBe	Be	4.2	65
	AmLi	Li	0.3	1.13

Table 1: Sources measured using the neutron spectrometer. Institutional knowledge about the composition of source 67-000 was ambiguous.

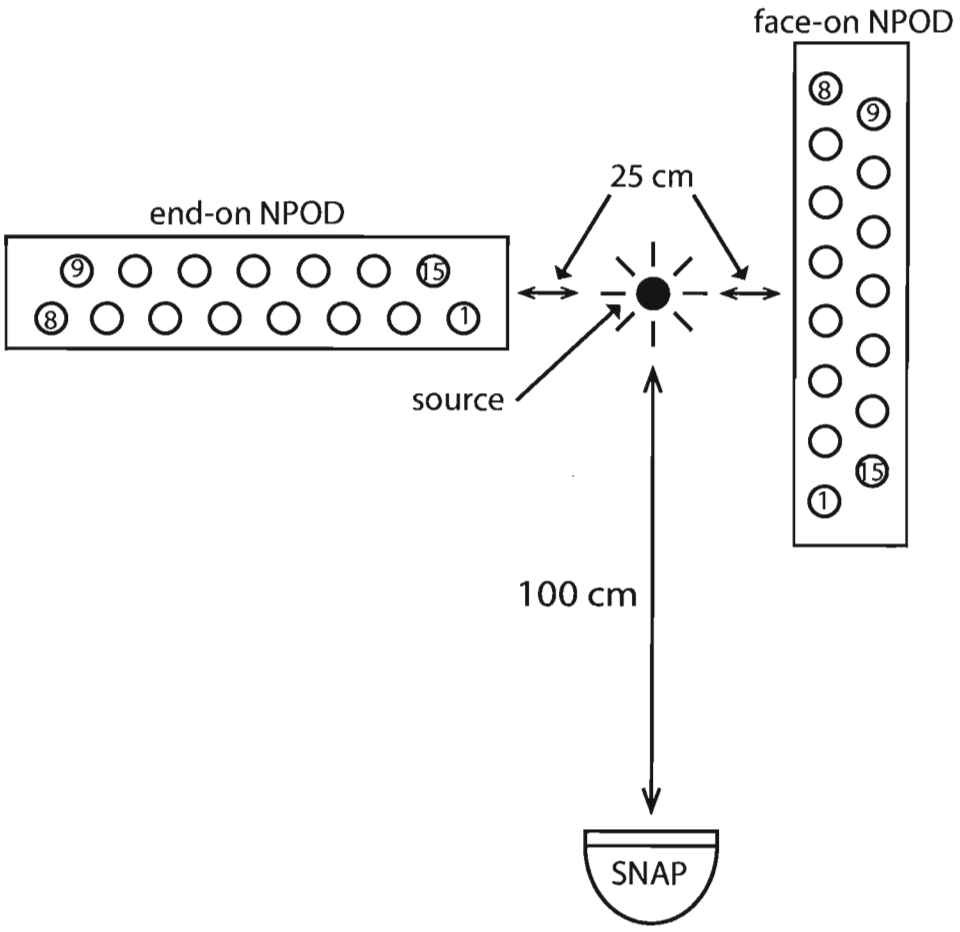

Figure 1: Experimental configuration of the two NPODs and single SNAP detector used for spectrometry. Channel numbering scheme of the NPODs is also shown.

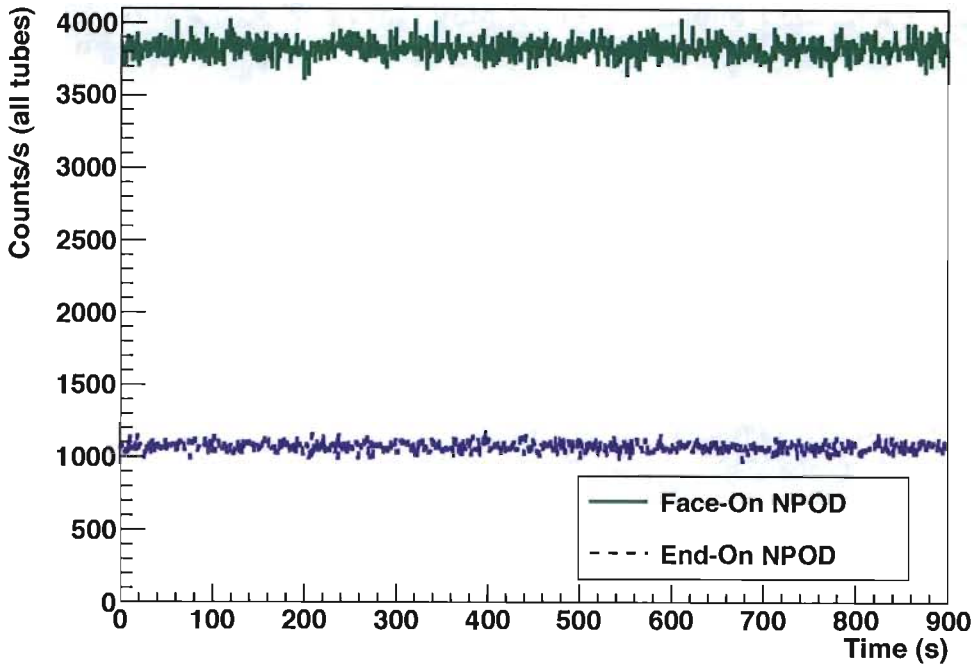

Figure 2: Total count rate observed in face-on and end-on NPODs during one 15-minute measurement of source 90-000, PuO_2 with boron impurities.

The clock output of one of the NPODs was sent into the pass-through channel of the other, to provide a single record of the timing of the system. The other pass-through channel was used to collect the SNAP detector output.

3 Results and Analysis

Due to the greater solid angle seen by the face-on NPOD, its total count rate was significantly higher than the count rate observed in the end-on NPOD for all (α, n) sources. A typical example can be seen in Figure 2. The ratio between end-on and face-on count rates was typically close to 0.25, but differed slightly for the sources measured, as shown in Figure 3, providing a qualitative indication of the neutron energy. The ratio measured for the PuF_4 source was slightly higher than the ratio for the PuO_2 with fluoride impurity, suggesting that the two sources do not possess identical compositions.

Since neutron stopping distance is dependent on energy, analysis of the variation in response rates among the ^{3}He tubes was expected to differentiate between low-energy neutrons, such as those emitted by (α, n) reactions on lithium and aluminum, and high-energy neutrons, such as those emitted by boron and beryllium. The response profiles of the individual ^{3}He tubes in both NPODs can be seen in Figures 4 and 5. These figures list the tubes in geometrical (see Figure 1) rather than numerical order and are organized from lowest-energy neutrons (AmLi, at 0.3 MeV) to highest-energy neutrons (PuBe, at 4.2 MeV).

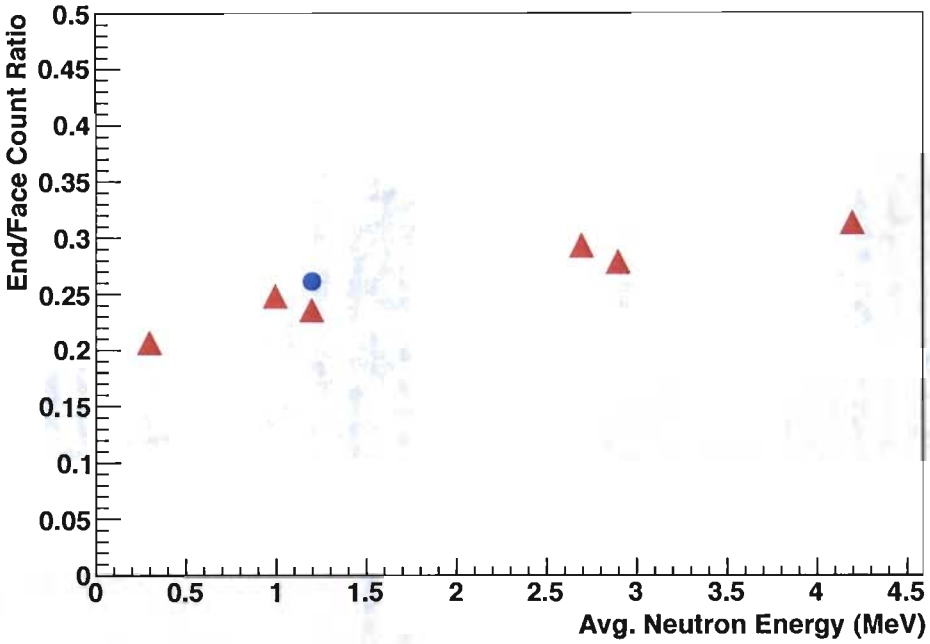

Figure 3: Ratio between end-on and face-on NPOD count rates for PuO_2, AmLi, and PuBe sources as a function of average energy of (α, n) neutrons. The circular point at 1.2 MeV represents the ratio for the PuF_4 source.

Several phenomena are noticeable in the response profiles. The largest number of neutron interactions occur not in the ^{3}He tubes that are closest to the source, but in the tubes behind them, since neutrons are more likely to be captured following moderation. Thus, tube 12 detects more counts than tubes 4 and 5 for the face-on NPOD, and tube 15, rather than tube 1, detects the largest number of counts for the end-on NPOD. The effect of moderation may be seen generally for the face-on NPOD by comparing the responses of the front tubes (numbers 1–8) with the responses of the back tubes (numbers 9-15) in Figures 4 and 5. With the exception of the AmLi profile, all sources show consistently higher counts in the back ^{3}He tubes. The AmLi source produces a different profile because its average neutron energy is sufficiently low to allow neutrons to be captured after few scattering interactions.

The response profiles for the end-on NPOD fall off quickly after the first few ^{3}He tubes, as neutrons escape or are captured in the polyethylene. This effect appears to be energy-dependent – as the neutron energy increases, a smaller fraction of neutrons are captured in the first two tubes (numbers 1 and 15) and more neutrons are detected in more-distant tubes.

4 Conclusions and Future Work

Measurements using two NPODs have been performed on eight (α, n) sources to assess the potential of the NPOD system as a neutron spectrometer. Initial examinations of the profiles of both detectors indicate that neutron energy affects the ratio in which neutrons are detected

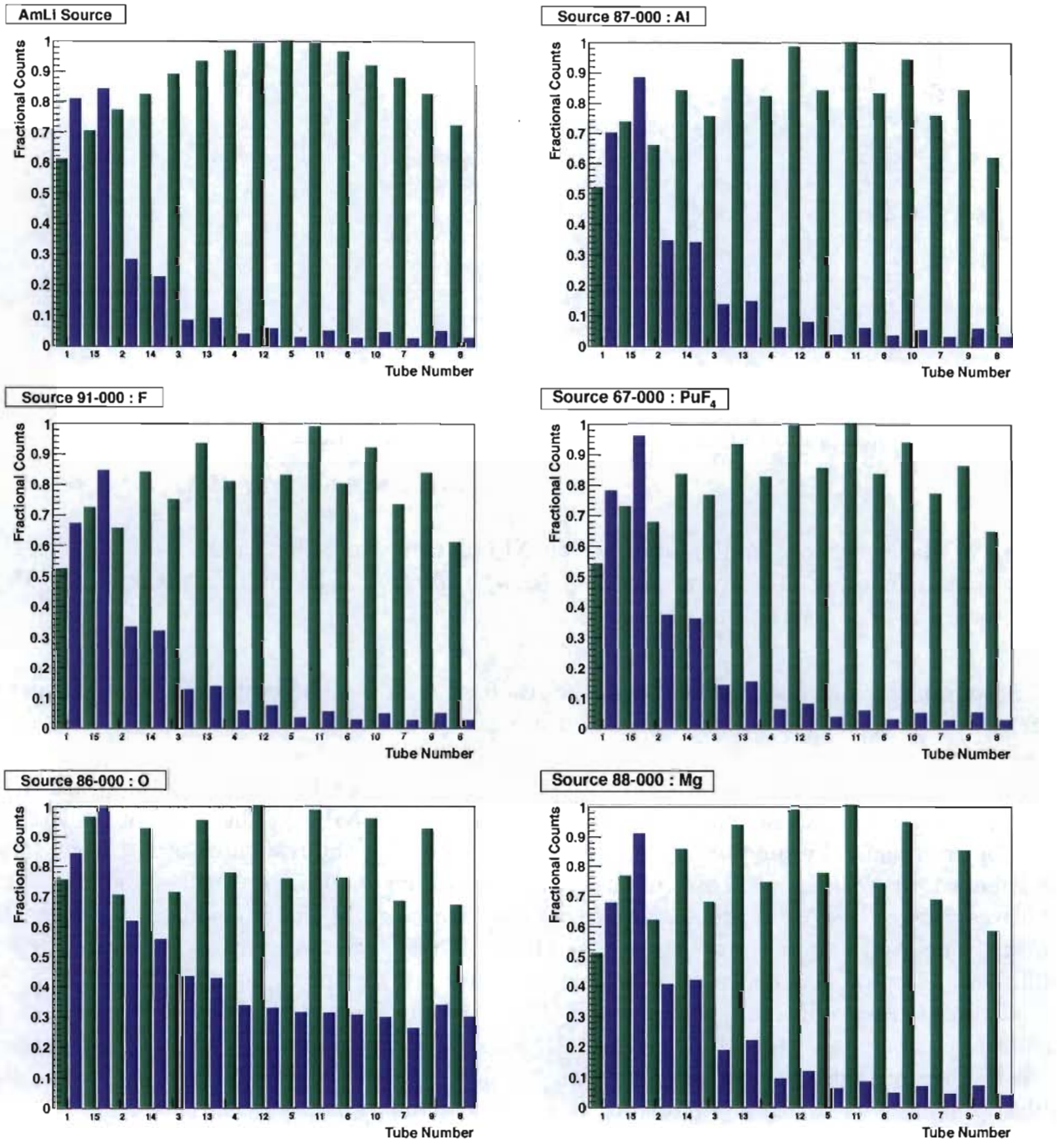

Figure 4: Normalized profiles for both NPOD detectors. Green lines represent head-on detector; blue lines represent end-on detector. Profiles are displayed in order from lowest-average-energy neutrons (AmLi) to highest (PuBe, in Figure 5).

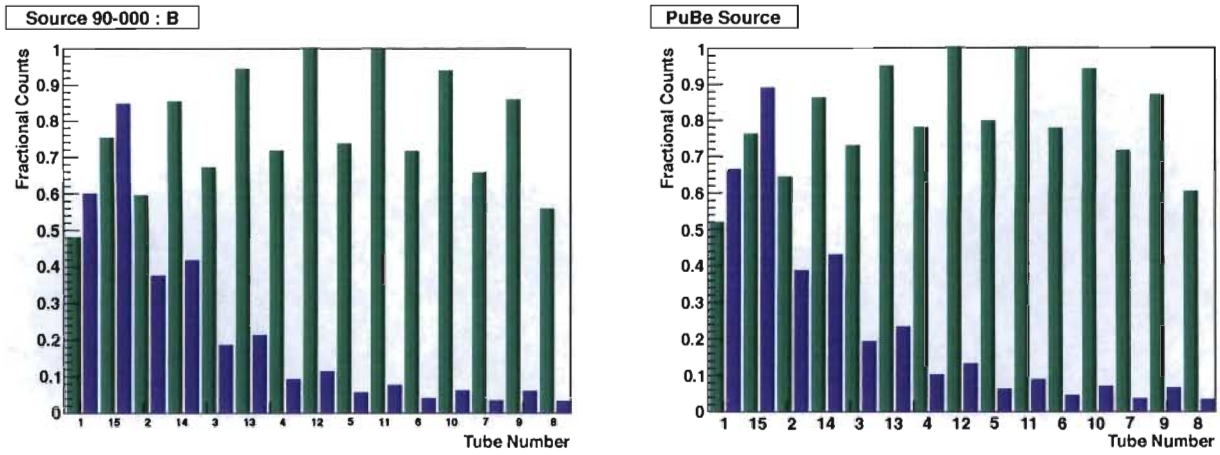

Figure 5: Normalized profiles for both NPOD detectors. Green lines represent head-on detector; blue lines represent end-on detector. Profiles are displayed in order from lowest-average-energy neutrons (AmLi, in Figure 4) to highest (PuBe).

in the two NPODs, as well as the identity of the individual tubes in which detections occur.

Analysis methods for the NPOD data that are under consideration primarily involve the ratios of ^{3}He tubes. For example, Figures 4 and 5 suggest that the difference between the face-on detector's front and back tubes increases with neutron energy, particularly for tubes located near the detector center. Ratios such as this one will be analyzed to determine whether their relationship to neutron energy can be quantified. Other potential analysis methods include the use of doubles data from the NPODs and the incorporation of data collected by the SNAP detector. Additional measurements will also be performed to improve the breadth of the data set, among them, measurements of an additional source, PuO_2 with silicon impurities, which has an average neutron energy of 1.2 MeV.

References

[1]F. D. Brooks and H. Klein. Neutron spectrometry — historical review and present status. Nucl. Instr. Meth. Phys. Res. A **476**:1-11, 2002.

[2]N. Ensslin, in: D. Reilly, N. Ensslin, H. Smith, Jr. (Eds.), Passive Nondestructive Assay of Nuclear Materials (NUREG/CR-5550), U.S. Government Printing Office, Washington, D. C., 1991, pp. 337.