Characterization of a Portable Neutron Coincidence Counter

Angela Thornton and W. Charlton
Texas A&M University
College Station, TX 77843

Abstract

Neutron coincidence counting is a technique widely used for qualitative and quantitative analysis of nuclear material. Because different isotopes possess different coincident neutron characteristics, the coincident neutron signature can be used to identify and quantify a given material. In an effort to identify unknown nuclear samples in field inspections, a new portable neutron coincidence counter has been developed. An in-depth analysis has been performed to establish whether the nuclear material in an unknown sample could be quantified with some confidence. The analysis was performed by comparing the true measurements of the system to the calculated output produced using MCNPX and the neutron coincidence point model. Based on the analysis, it is evident that this new portable system can play a useful role in identifying nuclear material for verification purposes.

Introduction

Neutron coincidence counting is an important passive, nondestructive-assay method that can be used to characterize nuclear material in bulk samples, namely plutonium. Because the number of neutrons emitted simultaneously during the fission process is particular to each isotope, the coincidence neutrons create a useful signature that can be used to describe the isotopes in a sample. Coincidence measurements can be done regardless of background or (α, n) neutrons making them less sensitive to external factors.

For plutonium, ^{238}Pu, ^{240}Pu, and ^{242}Pu all have dominant spontaneous fission yields producing coincidence neutrons. The measurement of these spontaneous fission neutrons, however, is complicated by the presence of induced fission neutrons. Induced fission is dependent on the fissile isotopes, primarily ^{239}Pu, and is caused by the absorption of neutrons in the sample. Both spontaneous fission and induced fission produce coincidence neutrons. Because these neutrons are emitted practically simultaneously, they create a useful signature that can be used to quantify a particular nuclear material[1,2].

The Portable Neutron Coincidence Counter was developed by Los Alamos National Laboratory to aid inspectors in field measurements of bulk samples. In order to benchmark and characterize the detector, a computer model was created using the Monte Carlo N-Particle eXtended (MCNPX) code[3] and its embedded advanced multiplicity capabilities. The results determined using MCNPX were then compared to the results calculated using the traditional Neutron Coincidence Point Model. This paper discusses both these results and their implications.

Background

The PNCC consists of four individual high-density polyethylene slabs. The slabs are approximately 7-in. (length) x 10-in. (height) x 3-in. (width) and weigh approximately 8 lbs. each. Four ^{3}He tubes are embedded in each polyethylene slab. Each 10-atm tube has a 1-in. diameter and 7-in. active length that run the height of the slab. Figure 1 shows a drawing of an individual slab. Other features include a low profile junction box to house the electronics package, onboard high voltage, and the ability to operate individually or combined in chain.

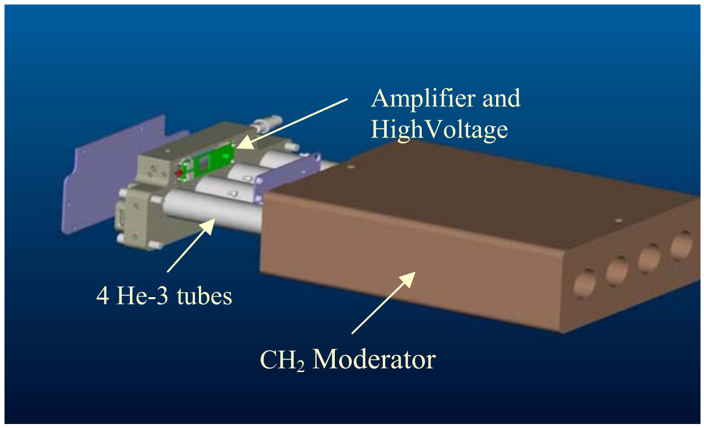

Figure 1. Individual slab of PNCC.

For the measurements described here, four slabs were arranged in a well-counter-type design. All four polyethylene slabs were sitting on two additional inches of polyethylene to lessen the effects of environmental factors such as the table material and proximity of the floor. This configuration is shown in Figure 2.

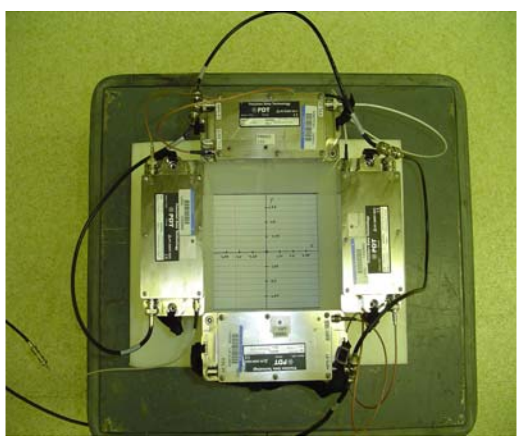

Figure 2. Experimental setup.

Using a small ^{252}Cf source, a number of detector characteristics were measured and/or calculated. These characteristics include the system efficiency, high voltage plateau, die-away time, and optimal gate length determination. These determined properties are listed in Table 1[1].

Table 1. Detector Characteristics.

Parameter	Value
Pu Efficiency	8.9%
High Voltage	1660 V
Die-Away Time	85 μs
Gate Length	64 μs

Experimental Data Analysis

After characterization, the detector system was set up, as shown in Figure 1, to measure the coincidence signature from four known plutonium oxide standards. IAEA Neutron Coincidence Counter software (INCC 5.04), which is a program that aids in collecting neutron coincidence data, was used to record the measurements.

The plutonium oxide samples were centered inside the sample area and counted for 15 minutes using 90 cycles of 10 seconds each. The average measured values are given in Table 2. The background rate, which was approximately 145 cps, has been accounted for in the INCC software. The outer packaging dimensions of the standards used were measured to be 5.25” × 6”.

Table 2. Measured data for plutonium standards.

Source ID	Singles Count Rates (cps)	1 σ Uncertainty	Doubles Count Rates (cps)	1 σ Uncertainty	Measured Detector Efficiency
LAO251C10	3721	4.80	132.4	2.17	8.2 %
LAO252C10	7017	2.98	267.8	2.03	8.4%
LAO255C10	12191	4.10	493.1	3.64	8.6%
LAO256C10	8369	3.63	328.7	2.46	8.4%

Because the original isotopic composition for each standard is known, the numbers of spontaneous fission and (α, n) neutrons are easily calculated using equations from page 484 of the Passive Nondestructive Assay of Nuclear Materials[2] (PANDA) manual:

$$\text{SF Yield (n/s)} = 1020 * (2.54*f_{238} + f_{240} + 1.69*f_{242}) \qquad \text{Eq. (1)}$$

$$(\alpha, n) \text{ Yield (n/s)} = 13400*f_{238} + 38.1*f_{239} + 141*f_{240} + 1.3*f_{241} + 2.0*f_{242} + 2690*f_{Am\text{-}241} \qquad \text{Eq. (2)}$$

In these equations, f_n is the isotopic weight percent of the nth plutonium isotope and $f_{Am\text{-}241}$ is the isotopic weight percent of the ^{241}Am in the sample[2]. The singles count rate for each sample was divided by the total neutron yield to determine the efficiency of the system. These values can also be seen in Table 2. Note that they are comparable to the

8.9% system efficiency determined using the small ^{252}Cf point source. The differences are mostly likely due to the uncertainties in the sample geometries.

MCNPX Calculations
New advanced multiplicity capabilities have been embedded inside MCNPX 2.5.0 f. These new capabilities allow for direct simulation of the detector response through the use of a ^{3}He capture tally. This simulation eliminates the need to use the traditional Point Model technique.

The PNCC was modeled in MCNPX. The model geometry mimicked the measurement set-up. Figure 3 shows the model of the system. The polyethylene slabs, ^{3}He tubes, and junctions boxes are all included in the model. The plutonium standards used in the experiments were modeled based on their weight and an approximate density of 0.9 g/cc.

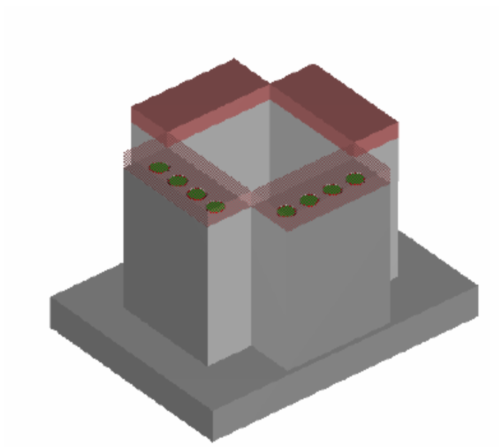

Figure 3. MCNPX model of the PNCC.

The true density of the material in the standards is not known with high certainty. Thus an assumed density of 0.9 g/cc was used in the model. The uncertainty in this parameter will pose some concern in the accuracy of the results, but a sensitivity analysis in relation to this parameter showed a small effect. The parameters dictating the sample geometry can be found in Table 3. Here the mass refers to the total mass of the sample, the radius refers to the radius of the inner canning, the height is the height of the inner canning, and the fill height refers to the fill height of the material in the sample. The source definition in the MCNPX model was changed accordingly.

Table 3. Sample Dimensions.

Sample ID	**Mass (g)**	**Density (g/cc)**	**Volume (cc)**	**Radius (cm)**	**Height (cm)**	**Area (cm*2)**	**Fill Height (cm)**
LAO-251	195.	0.9	216.7	5.4356	12.7	92.8207	2.33
LAO-252	365.1	0.9	405.6	5.4356	12.7	92.8207	4.37
LAO-255	616.8	0.9	685.3	5.4356	12.7	92.8207	7.38
LAO-256	436.5	0.9	485.0	5.4356	12.7	92.8207	5.23

The MCNPX simulation was split into two input decks: a spontaneous fission deck and an (α, n) deck. The neutron spectrum for the (α, n) deck was generated by the computer code SOURCES[4]. The two input decks maintained a consistent geometry and each contained F8 ^{3}He capture tallies. For the spontaneous fission deck, the calculated singles and doubles efficiencies were output. For the (α, n) deck, the singles efficiency was output. These were then multiplied by the appropriate fission yields to obtain the calculated count rates. The results of these tallies are listed in Table 4.

Table 4. MCNPX results.

Sample ID	SF Count Rate (cps)	Alpha Count Rate (cps)	Total Count Rate (cps)	Doubles Count Rate (cps)
LAO-251	2787	1186	3973	151
LAO-252	5226	2208	7434	301
LAO-255	9068	3863	12931	559
LAO-256	6297	2652	8949	373

Point-Model Calculations

Neutron Coincidence Point Model calculations were performed in order to compare the fundamentals of the point-model equation to the MCNPX code. The point-model equations for the singles and doubles are given below:

$$S = m * F * \varepsilon * \nu_{s1} * M * (1+\alpha)\,, \qquad \text{Eq. (3)}$$

$$D = m * F * \frac{\varepsilon^2}{2} * f_d * M^2 * (\nu_{s2} + \frac{(M-1)}{(\nu_{i1}-1)} * \nu_{s1} * \nu_{s2} * (1+\alpha))\,, \qquad \text{Eq. (4)}$$

where m = ^{240}Pu effective mass
F = spontaneous fission rate
ε = efficiency
M = leakage multiplication
α = (α, n) to SF neutron ratio
f_d = doubles gate fraction
ν_{s1}, ν_{s2} = first, second reduced moments of SF neutron distribution
ν_{i1}, ν_{i2} = first, second reduced moments of induced fission neutron distribution

For this model, $m, F, \nu_{s1}, \nu_{s2}, \nu_{i1}$, and ν_{i2} are all known parameters whereas M, ε, α, and f_d are all parameters obtained using MCNPX. In other words, a Monte Carlo model must be created to obtain some of the information required to use the point model. The main objective of this research is to show that MCNPX is now capable of performing all the necessary calculations and can directly simulate detector response so that the use of the Neutron Coincidence Point Model can be bypassed[5].

In the point model calculations performed for this analysis, M, ε, α, and f_d were obtained from both the traditional F4 tally and the new F8 ^{3}He capture tally. Equations 3 and 4 were then used to calculate the singles and doubles rates given by the Neutron Coincidence Point Model. The results based on the traditional F4 and new F8 tallies are given in Table 5. As can be seen, only minor differences exist.

Table 5. Neutron Coincidence Point Model data.

Standard ID	Calculated Singles From F4 (cps)	Calculated Doubles From F4 (cps)	Calculated Singles From F8 (cps)	Calculated Doubles From F8 (cps)
LAO-251	4180	137	4182	132
LAO-252	7813	264	7812	254
LAO-255	13560	473	13614	465
LAO-256	9405	323	9401	310

Data Comparison

The MCNPX and point model results are given again in Table 6, along with the original measured data. The point model results shown here are the results of using the traditional F4 tallies. From this data, it is obvious that the MCNPX simulation better calculates the singles and the point model better calculates the doubles. The point model data for singles differs from the measured values by roughly 10% while the MCNPX data differs by less than 7%. For the doubles, the MCNPX data differs from the measured values by about 14% while the point model data differs by less than 5%.

Table 6. Data Summary.

Sample ID	Measured Singles	Measured Doubles	Point Model Singles	Point Model Doubles	MCNPX Singles	MCNPX Doubles
LAO-251	3721	132	4180	137	3973	151
LAO-252	7017	268	7813	264	7434	301
LAO-255	12191	493	13560	473	12931	559
LAO-256	8369	329	9405	323	8949	373

Because the coincidence signature is unique to a specific isotope, the doubles rates are of the most importance. Figure 4 shows a plot of the doubles rate as a function of Pu_{eff} mass. It is obvious that the Neutron Coincidence Point Model does a better job estimating the doubles rates. The MCNPX simulations appear to show a consistent positive bias in the doubles count rate which could be due to uncertainties in the neutron multiplicity data used in MCNPX.

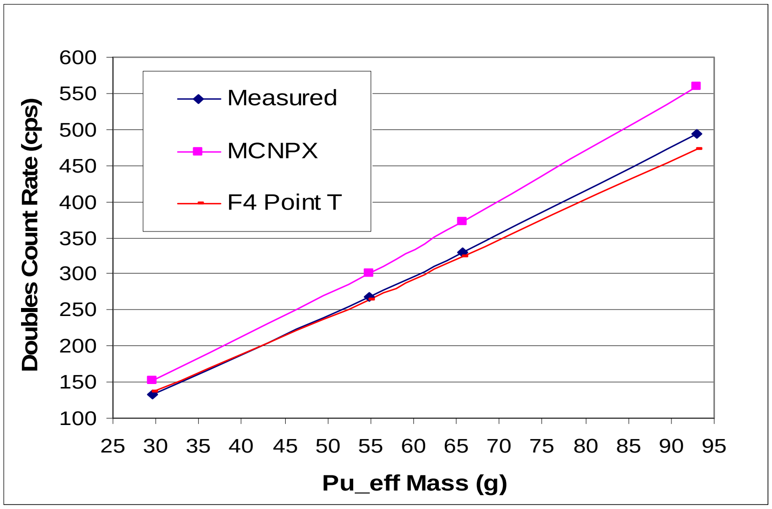

Figure 4. Doubles rate as a function of Pu_{eff} mass.

Sensitivity Analysis

In an attempt to identify unknown parameters in the MCNPX model that could account for the bias seen in Figure 4, a short sensitivity analysis was performed. The dependence of sample density on detector response was studied and shown to have small effects for densities between 0.5 and 1.0 g/cc. A more in-depth study was then performed to determine how varying water content in a given sample would alter the results measured by the detector system. Figure 5 shows the results of this analysis. Based on the MCNPX calculation without water, the differences in doubles rates are practically negligent with water contents up to 5%.

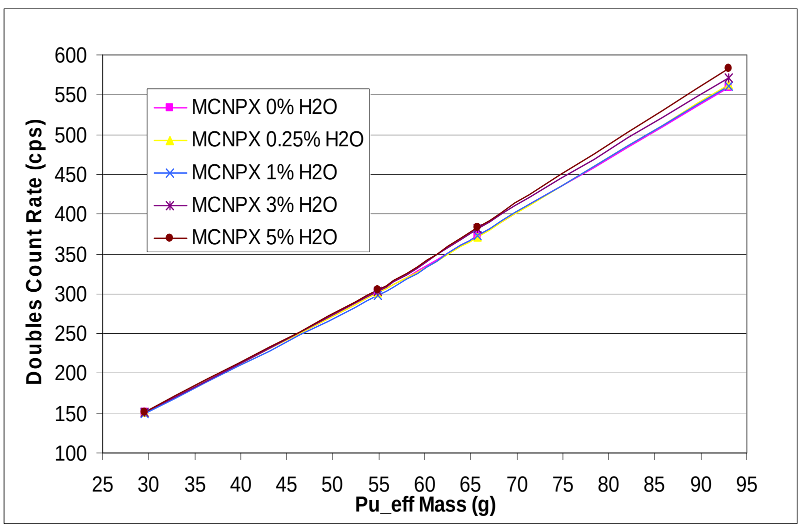

Figure 5. Double rates vs. Pu_{eff} for various water contents.

When water is added to a sample it causes additional absorption (loss of neutrons) but also moderates more neutrons making them more likely to be detected and more likely to be multiplied in the sample. These competing effects due to the additional water in the sample seem to offset each other. The doubles rates seem to be driven primarily by the number of neutrons created. Using MCNPX, the numbers of neutrons created and lost were recorded. For the larger standards, the numbers of neutrons created and lost did not change significantly until the water added approached 10%. For the smallest sample, these numbers did not significantly change until the water added approached 5%. Therefore the sensitivity analysis concludes that for small variations of water in the samples the results will not be compromised.

Conclusion

Overall, the Portable Neutron Coincidence Counter will be a useful tool for aiding in nuclear safeguards. The analysis of this detector should be verified by performing more tests and measurements of more known samples. The known $^{240}Pu_{eff}$ mass of these samples should then be compared to the $^{240}Pu_{eff}$ given by this model.

Although the Neutron Coincidence Point Model has proved to be a more accurate method for neutron coincidence measurements, it is evident that MCNPX can be a viable tool that should be utilized for the purposes of neutron coincidence measurements. Not only is MCNPX a practical tool, it is an efficiency way to directly simulate detector response. More work should be done to investigate why the doubles rates directly calculated by MCNPX are slightly higher than expected.

Acknowledgement

We would like to acknowledge Los Alamos National Laboratory for their contributions to this project.

References

1. Angela Thornton, H. Menlove, W. Charlton, M. Miller, and C. Rael, "Characterization of a Portable Neutron Coincidence Counter," Los Alamos National Laboratory document LA-UR-06-4608 (2006).

2. Doug Reilly et al., "Passive Nondestructive Assay of Nuclear Materials," NUREG/CR-5550 U.S. Government Printing Office, Washington, D.C. (1991).

3. Denise Pelowitz, Ed., *MCNPX User's Manual,* Version 2.5.0, Los Alamos National Laboratory document LA-CP-05-0369 (2005).

4. W. B. Wilson, R.T. Perry, W.S. Charlton et al., "SOURCES: A Code for Calculating (α, n), Spontaneous Fission, and Delayed Neutron Sources and Spectra," Los Alamos National Laboratory document LA-UR-03-1208 (2003).

5. N. Ensslin et al., "Application Guide to Neutron Multiplicity Counting," Los Alamos National Laboratory report LA-13422-M (1998).