SAND2009-7594C

Plutonium Sphere Multiplicity Simulations with MCNP-PoliMi

E.C. Miller[1*], S.D. Clarke[1], S.A. Pozzi[1] and J. Mattingly[2]
[1]*University of Michigan, Ann Arbor, MI, 48109*
[2]*Sandia National Laboratories, Albuquerque, NM 87185*
**ericcm@umich.edu*

ABSTRACT

In order to improve the characterization methods for fissile materials, effort must be made to validate the computer programs that are used to simulate the behavior of these systems. For this work, measurements of a 4.4-kg sphere of weapons grade plutonium metal were taken. A detector system of ^{3}He tubes was used to measure the multiplicity of the system. The experiment was then modeled using the MCNP-PoliMi code and the simulated results were compared to the measured results via the Feynman-Y metric. MCNP-PoliMi is able to correctly predict the measured value of the Feynman-Y vale within 5% for all of the moderated cases and within 22% for the bare sphere.

I. INTRODUCTION

The recent concern regarding the potential proliferation of nuclear material for use in a nuclear-based weapon has increased the demand for methods to detect, characterize, and track such materials. One method that can be used to quickly characterize an unknown fissile sample is neutron multiplicity analysis. This method relies on detecting fissile material by detecting correlated neutrons, which can be used to distinguish fissile events from the random events that are characteristic of the terrestrial background. This is a well-established assay in the field of nuclear materials, control, and accountability [1].

The objective of our current work is to benchmark the ability of the MCNP-PoliMi code to simulate the neutron multiplicity distributions in a heavily moderated system. To perform this benchmark, simulations of a polyethylene moderated plutonium sphere are being performed using MCNP-PoliMi. This work is of great importance to the non-proliferation community because showing that neutron multiplicities can be accurately simulated will improve our ability to model the characterization and detection of fissile material.

MCNP-PoliMi is a modified version of the code MCNP-4C [2]. Several changes have been implemented in MCNP-PoliMi that improve its ability to simulate correlated events. The full fission multiplicity distributions have been implemented and several specific spontaneous fission and (alpha, n) source definitions have been included.

The simulated data is post-processed to include the effects of dead time associated with the ^{3}He tubes and their associated electronics. The post-processor also applies a shifting window that (at regular intervals) determines the number of events present. This time window was varied from 16 µs to 4096 µs in 16-µs increments. The events counted in the time window are used to establish a neutron multiplicity distribution.

II. EXPERIMENT

The experimental measurements were made by Sandia National Laboratories personnel at the Nevada Test site. The experiment consisted of a 4.483-kg weapons-grade plutonium metal sphere placed 50 cm from a bank of ^{3}He detectors. The plutonium sphere has a composition as shown in Table 1. The plutonium was α-phase and had a density of 19.60 g/cm^{3}. The experimental setup is shown in Figure 1.

Table 1. Isotopic and source information for the weapons-grade plutonium sphere

Isotope	Mass (g)	Mass %	Neutrons/ Second	% of Total Neutrons
Pu-239	4200.70	93.70	91.58	0.03
Pu-240	266.46	5.94	271785.32	99.18
Pu-241	7.40	0.16	0.37	1.35E-04
Am-241	4.58	0.10	5.40	1.97E-03
Pu-242	1.26	0.03	2158.97	0.79
U-235	1.19	0.03	0.00	1.3E-07
U-238	0.83	0.02	0.01	4.11E-06
Total	4482.4	---	274041.0	---

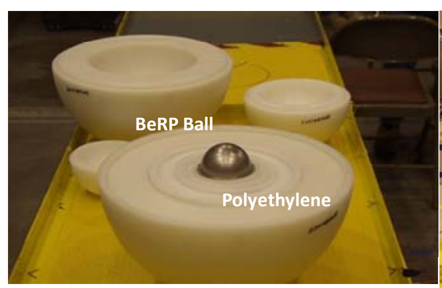

Figure 1: Pictures of the experimental setup
Left: Plutonium Sphere and polyethylene shells
Right: The NPOD detector

To increase the moderation, the plutonium sphere was surrounded with custom-built polyethylene shells. These shells allowed for the plutonium sphere to be surrounded by 1.27 cm, 2.54 cm, 3.81cm, 7.62 cm, and 15.24 cm of polyethylene.

The detector bank consisted of 15 ^{3}He detectors arranged in two rows in a polyethylene matrix, with eight detectors on the front row and seven detectors in the back row. The ^{3}He detectors had a length of 42.164 cm and a radius of 1.27 cm, including the detector housing and had a pressure of 10 atm.

The entire detector bank was surrounded with 0.0762 cm of cadmium. This detector bank was placed on a table that is 1 meter from the concrete floor.

In addition to the measurements taken with the plutonium sphere, the measurements were also repeated using a point source of ^{252}Cf. This californium source had an activity of 57,046 spontaneous fissions events per second. This source was placed in the same configurations using the polyethylene moderators as the plutonium sphere.

III. MODELING WITH MCNP-POLIMI

In most Monte Carlo codes a single source particle is simulated for each simulated history. This considerably complicates the simulation of a multiplicity measurement, where the numbers of neutrons produced in an event are extremely important. MCNP-PoliMi is one of the few Monte Carlo codes with the ability to simulate multiple source neutrons, and for this reason this code was chosen.

In addition to the ability to handle multiple source neutrons, MCNP-PoliMi also has full multiplicity and energy distributions built into the code for several common isotopes of interest such as ^{252}Cf, ^{240}Pu, ^{235}U, and ^{238}U. Typical Monte Carlo codes sample the number of neutrons released from fission by taking the integer values on either side of the ν bar value while preserving the average number of neutrons released [3]. For example, a ν bar of 3.7 would release 4 neutrons 70% of the time and 3 neutrons 30% of the time, preserving the average behavior. In MCNP-PoliMi the full neutron distributions are implemented for the induced fission distributions. These distributions are based on two papers by Terrell [4] and Zucker and Holden [5].

In addition to the full multiplicity distributions being implemented for spontaneous fission sources, MCNP-PoliMi also offers improved handling of the physics simulations, correcting the sampling order to better simulate actual events. This allows for meaningful information to be drawn from individual particle histories.

The experimental geometry was built in MCNP-PoliMi as shown in Figure 2.

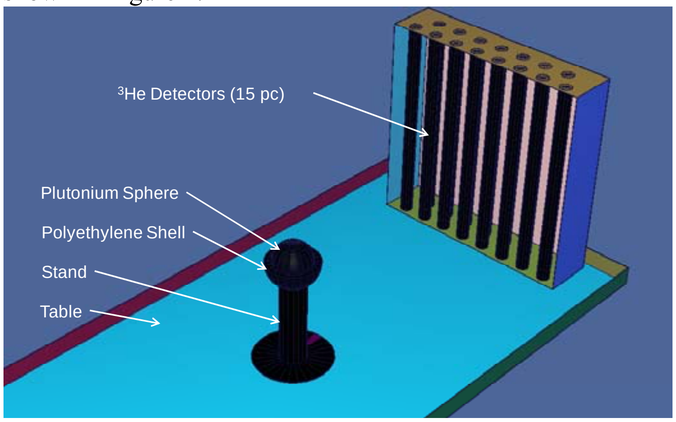

Figure 2. The setup of the experimental geometry. The top portion of the polyethylene sphere, the front face of the detector and the polyethylene matrix of the detector have been voided to show the geometry

The plutonium sphere source was modeled using the built-in spontaneous fission source for ^{240}Pu. Other neutron-producing isotopes in the sphere were ignored. As shown in Table 1, the ^{240}Pu provides over 99% of the total source activity.

The experiment was modeled as close to the physical experiment as possible. Nearby objects such as the table and the concrete floor provide surfaces for the neutrons to scatter on and this effect will influence the results. The walls and ceiling were far enough from the measurement system that their effects can be ignored. The model also included the surrounding air.

To reproduce the multiplicity measurement as faithfully as possible, the plutonium source in MCNP-PoliMi was distributed over time. This time distribution was essential for simulating multiplicity measurements because neutrons from separate spontaneous fissions can interact with a detector at the same time. Using the information in Table 1, the number of neutrons emitted in a given time can be calculated. This number is used to determine the number of neutrons to simulate when the source is distributed over this determined time.

MCNP-PoliMi provides an output file which contains information about individual particle track information in a given cell. All of the information about events occurring in the ^{3}He tubes are post-processed using a FORTRAN script. This post-processing code applies a 4μs dead time to each of the detectors. Once all of the simulated events that would be removed by the dead-time effects of the detector system occur, the post-processor also determines the multiplicity of the sample. This is determined by first sorting all accepted events in time. Then a counting window (or gate) of a fixed width is open at time=0. The first time window chosen was 16 μs.

These windows are opened incrementally through the list of accepted pulses. The number of events that occur in each window are counted and placed in a histogram. The histogram that is constructed represents the neutron multiplicity distribution for that fixed counting window. The process is then repeated for increasingly larger counting windows. In this work, the counting windows ranged from 16 μs to 4096 μs in 16 μs increments. A schematic of this process is shown in Figure 3.

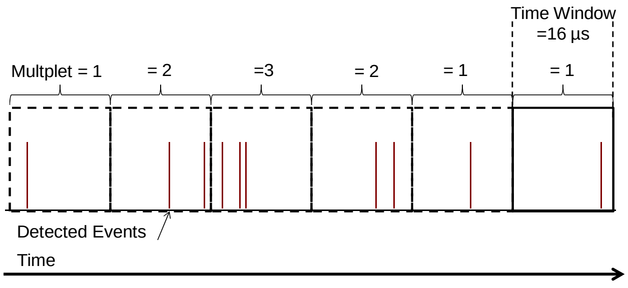

Figure 3. Schematic diagram of the multiplicity counting process

IV. THE FEYNMAN-Y METRIC

To compare the simulated results to the measured data, the Feynman-Y was chosen. This metric determines the deviation of the variance of the neutron multiplicity distribution in excess from the variance of a normal Poisson distribution, as shown in Equation 1 [3].

$$Y = \frac{\sigma^2}{\mu} - 1 \qquad \text{[eq. 1]}$$

This Feynman-Y is calculated for each of the different counting windows. The Feynman-Y metric is useful because most background radiation is random (except neutrons from cosmic interactions). Random events will have a Poison distribution and because the Feynman-Y is a measure of the deviation from this random distribution, it measures the level of correlation in a given sample. This level of correlation can be related to the multiplicative or fissile nature of the sample.

V. VERIFICATION OF THE MODELED GEOMETRY

To verify the modeled geometry in MCNP-PoliMi, the simpler measurement of the ^{252}Cf was modeled. The ^{252}Cf source was modeled using the built-in spontaneous fission source available in MCNP-PoliMi. MCNP-PoliMi offers the option of simulating the spontaneous fission source term with a degree of anisotropy. This level of anisotropy has been demonstrated for ^{252}Cf in experiments. However, the isotropic source term results in the most accurate simulation of the measured data. Once this conclusion was reached, all further modeling was done using an isotropic source term.

Once modeled, the neutron multiplicity distributions and the Feynman-Y were calculated for the simulated data and compared to the measured results. As can be seen in Table 2, the mean and variance of the simulated neutron multiplicity distributions are within 4% of the measured values. The Feynman-Y is also predicted very closely. The percent difference appears abnormally large due to the calculation of percent difference of extremely small numbers.

From these results it is appears that the geometry dimensions and materials are modeled accurately.

To verify that the plutonium source was accurately modeled the k_{eff} was calculated and compared to the results of a benchmark experiment [7]. The results of this comparison are shown in Table 3. As shown in Table 3, the k_{eff} is accurately reproduced by MCNP-PoliMi, with all of the simulations predicting the k_{eff} within 1%.

This is an indication that the plutonium sphere source and composition are realistic representations of the physical sample.

Table 2. Comparison of neutron multiplicity distributions for the ^{252}Cf simulation to measured data

Poly Thickness (cm)		Measured	MCNP-PoliMi	% Difference
0.0	Mean	7.9403	7.9915	0.64
	Variance	8.1660	8.2138	0.59
	Feynman-Y	0.0284	0.0278	-2.10
1.27	Mean	8.4559	8.7210	3.14
	Variance	8.7403	9.0125	3.11
	Feynman-Y	0.0336	0.0334	-0.66
2.54	Mean	8.5372	8.6568	1.40
	Variance	8.7926	8.9212	1.46
	Feynman-Y	0.0299	0.0305	2.07
3.81	Mean	8.0608	7.8656	-2.42
	Variance	8.3255	8.0502	-3.31
	Feynman-Y	0.0328	0.0235	-28.51
7.62	Mean	4.5156	4.5156	0.00
	Variance	4.5985	4.5478	-1.10
	Feynman-Y	0.0184	0.0175	-4.53
15.24	Mean	1.0987	1.0987	0.00
	Variance	1.1026	1.0806	-2.00
	Feynman-Y	0.0035	0.0027	-24.08

Table 3. K_{eff} Results for the Plutonium sphere compared to benchmark experiments

Polyethylene Thickness (cm)	Benchmark	MCNP-PoliMi	% Difference
0.0	0.7805	0.7740	-0.835
1.27	0.8253	0.8237	-0.194
2.54	0.8665	0.8668	0.037
3.81	0.8999	0.8991	-0.089
7.62	0.9364	0.9348	-0.176
15.24	---	0.9381	---

VI. PLUTONIUM SPHERE RESULTS

With the MCNP-PoliMi geometry verified and the plutonium source term compared to the expected k_{eff} the plutonium sphere and polyethylene shell experiments were modeled. The results for the Feynman-Y for the bare sphere case are shown in Figure 4.

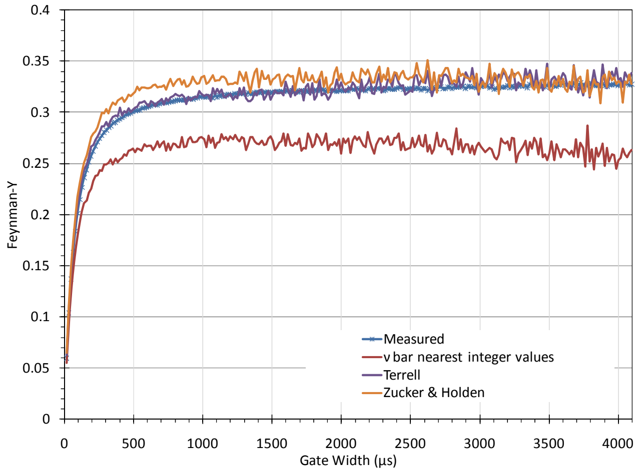

Figure 4. Feynman-Y results for the bare plutonium sphere showing the effect of induced fission distributions

In Figure 4, the measured Feynman-Y is well-predicted using the induced fission distributions as described by Terrell. The sampling of integer values of ν bar under-predicts the measured results as expected. The distribution predicted by Zucker and Holden provides an adequate prediction of the Feynman-Y but it appears to be less accurate than the result obtained with the Terrell distribution. When the neutron multiplicity distribution at a gate width of 4096 µs is examined, there is a slight shift in the simulated mean compared to the measured mean. The neutron multiplicity distribution for the bare plutonium sphere case is shown in Figure 5.

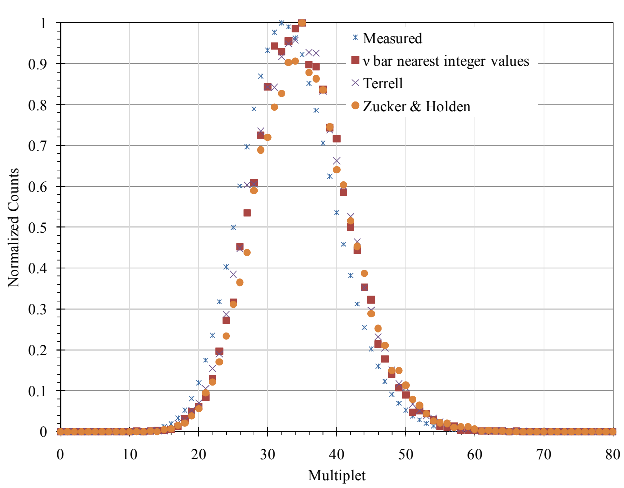

Figure 5. Neutron multiplicity distribution for the bare plutonium sphere

When the polyethylene shells are added to the model, the simulated Feynman-Y using the Terrell and Zucker and Holden distributions begin to deviate from the measured values. Surprisingly, when the less physical distribution of the integer values around ν bar is used, they provide the most accurate prediction of the measured result. This effect is shown in Figure 6 for the plutonium sphere moderated with 2.54 cm of polyethylene.

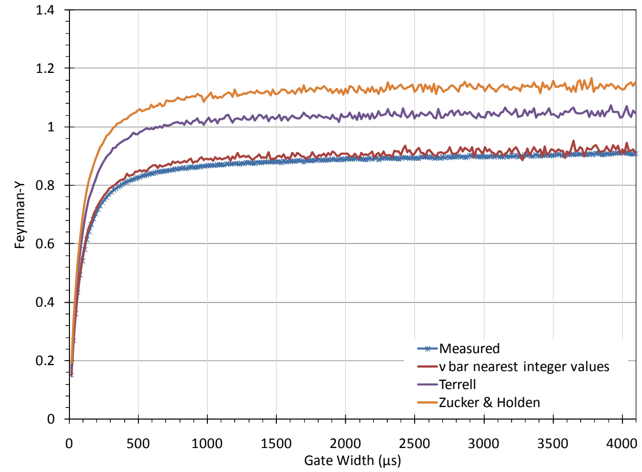

Figure 6. Convergence of the Feynman-Y for the 2.54 cm moderated plutonium sphere

The neutron multiplicity distribution for the 2.54-cm moderated case also displays a more pronounced shift in the mean than was seen in the bare plutonium sphere case. The neutron multiplicity distribution using a 4096 µs gate width is shown in Figure 7.

These effects of the ν bar integer values most accurately predicting the measured results were seen for all of the moderated cases. The shift in the mean value of the distributions was also observed. A summary of the key parameters of the distributions for the plutonium sphere are shown in Table 4, which demonstrates that the Terrell distribution predicts the measured Feynman-Y within 0.2%. However, as moderation is added, the prediction of the Feynman-Y deviates by as much as 21%. Conversely, the ν bar integers are off by 20% for the bare sphere case.

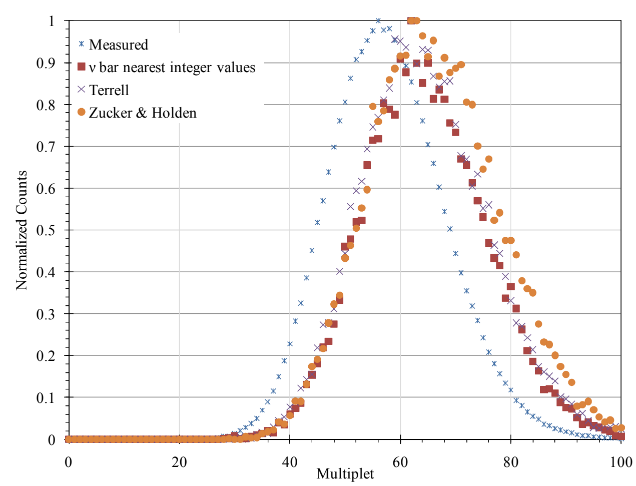

Figure 7. Neutron multiplicity distribution for the 2.54 cm moderated plutonium sphere

However, the deviation in the Feynman-Y improves to within 5% for the moderated cases. The Zucker and Holden distributions follow the same trend as the Terrell distribution, but do not predict the measured results as accurately as Terrell.

The mean values of the simulation appear to vary evenly for all three distributions. In all cases they are approximately the same percentage greater than the mean value determined by the measurement. This indicates that the difference in the variance of these distributions is the cause for the variability in the predictions of the Feynman-Y.

This effect is shown in Table 4. The variance between the distributions can vary greatly even for the same thickness of polyethylene. In the case of the 7.62-cm moderated plutonium sphere, the variance varies from 5% with ν bar integers to 34% with Zucker and Holden.

Table 4. Summary of key parameters for the comparison of the neutron multiplicity distributions

Polyethylene Thickness		Measured	ν bar integers	% Difference	Terrell	% Difference	Zucker and Holden	% Difference
0.0	Mean	31.3945	32.1804	2.50	32.1570	2.43	32.6052	3.86
	Variance	41.6569	40.5461	-2.67	42.6898	2.48	44.1867	6.07
	Feynman-Y	0.3269	0.2600	-20.45	0.3275	0.19	0.3552	8.67
0.5	Mean	42.1144	44.9157	6.65	44.8054	6.39	45.7511	8.64
	Variance	65.1203	68.3311	4.93	70.7907	8.71	76.3712	17.28
	Feynman-Y	0.5462	0.5213	-4.55	0.5800	6.18	0.6693	22.53
1.0	Mean	54.6182	59.6387	9.19	59.5678	9.06	61.1395	11.94
	Variance	104.1455	114.5891	10.03	122.6351	17.75	130.4507	25.26
	Feynman-Y	0.9066	0.9213	1.62	1.0586	16.76	1.1335	25.02
1.5	Mean	66.1439	71.2778	7.76	71.2884	7.78	73.6661	11.37
	Variance	159.5502	174.6478	9.46	189.7433	18.92	206.3002	29.30
	Feynman-Y	1.4120	1.4502	2.71	1.6615	17.67	1.8003	27.50
3.0	Mean	56.7284	59.7204	5.27	59.8426	5.49	62.8649	10.82
	Variance	155.1344	162.4197	4.70	185.3554	19.48	208.0389	34.10
	Feynman-Y	1.7326	1.7195	-0.75	2.0972	21.04	2.3091	33.27
6.0	Mean	13.7935	13.9788	1.34	13.9154	0.88	14.7215	6.73
	Variance	20.1730	20.1020	-0.35	21.1307	4.75	23.6180	17.08
	Feynman-Y	0.4624	0.4380	-5.28	0.5184	12.12	0.6041	30.66

VII. CONCLUSION AND FUTURE WORK

This work has shown that the model of the experimental system is accurate as is demonstrated by the ability of the code to correctly predict the mean and variance results of the ^{252}Cf measurements. The accuracy of the plutonium sphere has been verified by the comparison between the simulated value for the k_{eff} and the benchmarked values.

The MCNP-PoliMi code is capable of accurately simulating Feynman-Y of the bare plutonium sphere using the induced neutron distributions presented by Terrell. This value was simulated within 2.43% of the measured value.

However, using the Terrell distributions, the simulated prediction of the Feynman-Y for the moderated cases can vary by as much as 20%. Conversely, using the standard MCNP induced fission treatment, the simulated Feynman-Y of the bare plutonium sphere under-predicts the measured value by 20.45%. Yet using the standard MCNP induced fission treatment results in the Feynman-Y of the moderated cases being within 5.28% of the measured value.

Future work on this project will investigate the cause for the deviation between the prediction of the Feynman-Y for the bare and moderated spheres depending on the choice of distribution that is added.

References

1. N. Ensslin, W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell, J.E. Stewart. "Application Guide to Neutron Multiplicity Counting," LA-13422-M, 1998
2. S.A. Pozzi, E. Padovani, and M. Marseguerra. "MCNP-PoliMi: A Monte Carlo Code for Correlation Measurements," *Nucl. Instr. and Meth. A*, 513, pp. 550–558 (2003).
3. D-5 MCNPX Development Team, "MCNPX User's Manual, Version 2.5.0," *Los Alamos National Laboratory,* LA-CP-05-0369 (2005).
4. Terrell, James. "Distributions of Fission Neutron Numbers," *Physical Review*. Vol. 108, No. 3, 1957
5. M.S. Zucker and N.E. Holden, "Energy Dependence of the Neutron Multiplicity ρ_ν in Fast Neutron Induced Fission of $^{235,238}U$ and ^{239}Pu," *Brookhaven National Laboratory*, BNL-38491, 1986
6. John Mattingly and Eric Varley. "Synthesis of the Feynman-Y Neutron Multiplicity Metric using Deterministic Transport, invited," *Transactions of the American Nuclear Society*. Vol. 98, pp. 572-574. 2008
7. Timothy E. Valentine, "Polyethylene-Reflected Plutonium Metal Sphere Subcritical Noise Measurement," *Oak Ridge National Laboratory*, SUB-PU-MIXED-00.