

LA-UR-12-*00585*

*Approved for public release;
distribution is unlimited.*

Title: An Evaluation of Monte Carlo Simulations of Neutron Multiplicity Measurements of Plutonium Metal

Author(s): John Mattingly, Eric Miller, C.J. Solomon, Ben Dennis, Amy Meldrum, Shaun Clarke, Sara Pozzi

Intended for: 2012 ANS Annual Meeting
Chicago, IL
June 24-28



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

An Evaluation of Monte Carlo Simulations of Neutron Multiplicity Measurements of Plutonium Metal

John Mattingly^A, Eric Miller^B, C.J. Solomon^C, Ben Dennis^B, Amy Meldrum^B, Shaun Clarke^B, Sara Pozzi^B

^A North Carolina State University, 2500 Stinson Dr, Raleigh, NC 27695-7909, john_mattingly@ncsu.edu

^B University of Michigan, 2355 Bonisteel Blvd, Ann Arbor, MI 48109, ericcm@umich.edu

^C Los Alamos National Laboratory, PO Box 1663, Los Alamos, NM 87545, c.solomon@lanl.gov

INTRODUCTION

Neutron multiplicity counting experiments with subcritical assemblies of polyethylene-reflected plutonium metal were conducted at the Nevada National Security Site in 2009. [1] These experiments were applied to evaluate the accuracy of Monte Carlo simulations of neutron multiplicity measurements of highly multiplicative assemblies of plutonium. Simulations were performed using two different, independently developed Monte Carlo codes. Both codes exhibited near identical errors in the calculated neutron multiplicity distribution. A sensitivity study was conducted to identify the source(s) of the observed errors. This evaluation examined all the plausible sources of error, including the Monte Carlo simulation logic, the material and geometry models, detector dead-time, plutonium density, and the induced fission neutron multiplicity (\bar{v}) of ^{239}Pu . The results of this evaluation seem to indicate that small errors in ENDF/B-VII evaluation of ^{239}Pu \bar{v} may be the primary cause of the observed error in the calculated neutron multiplicity distribution. [2]

EXPERIMENTS AND SIMULATIONS

Neutron Multiplicity Counting Experiments

The source was a 4484 g sphere of alpha-phase plutonium metal (nominally 19.66 g/cm³) containing 93.3% ^{239}Pu and 5.9% ^{240}Pu by mass. [3] The plutonium metal sphere was clad in a 0.3 mm thick stainless steel shell. Six measurements were performed with the plutonium source (a) bare and (b) reflected by 5 nesting spheres of high density polyethylene (HDPE, nominally 0.96 g/cm³) with total thicknesses of 12.7, 25.4, 38.1, 76.2, and 152.4 mm. The plutonium source and HDPE reflectors are shown in Fig. 1.

MCNP5 calculations estimated the effective neutron multiplication factor k_{eff} ranged between 0.78 for the bare case and 0.94 for the most reflected case, i.e., neutron multiplication $M = 1/(1 - k_{\text{eff}})$ ranged between approximately 4.5 and 16.7. [3]

Each of the 6 configurations of the plutonium source was measured using a portable neutron multiplicity counter constructed from 15 ^3He proportional counters. The counters contained 101.3 MPa of ^3He gas with a

small quantity of CO₂ quench gas. Each counter was 381 mm long and 25.4 mm in diameter. The 15 counters were embedded in 2 rows in an HDPE moderator 430 mm wide, 422 mm tall, and 102 mm thick. The front row of 8 counters was positioned 19 mm back from the front face of the moderator, and the rear row of 7 counters was positioned 61 mm back from the front face of the moderator. The moderator was wrapped in 0.8 mm of cadmium to minimize the instrument's sensitivity to neutrons reflected by, e.g., the floor. The multiplicity counter was positioned 500 mm from the center of the plutonium source in all 6 measurements. The neutron multiplicity counter and plutonium source are shown in Fig. 2.

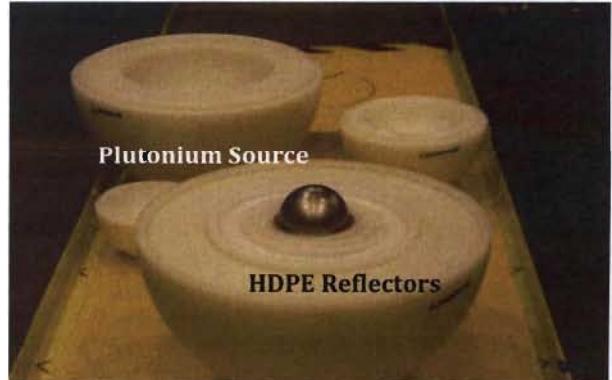


Fig. 1. The plutonium source and HDPE reflectors.

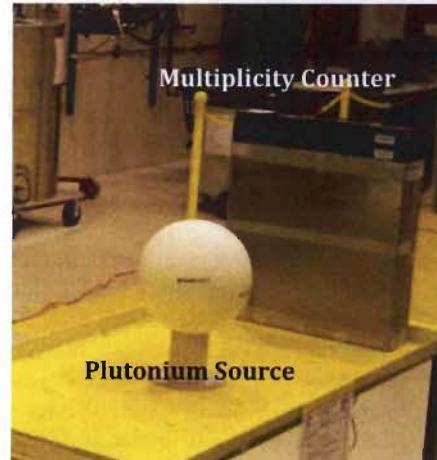


Fig. 2. The plutonium source and neutron multiplicity counter.

The multiplicity counter recorded neutron detection events in list mode: the channel number of each counter registering a neutron count was recorded in 1 μ s intervals. The measured neutron multiplicity distribution was accumulated by recording the number of coincident counts for a fixed coincidence gate width. A coincidence gate width of 4096 μ s is used throughout this paper.

Monte Carlo Simulations

Monte Carlo models of each of the 6 measurements were constructed, and the experiments were simulated using MCNPX-PoliMi, a modification of MCNPX 2.7.0 maintained by the University of Michigan (UM) and the Polytechnic of Milan, Italy. [4, 5] MCNPX-PoliMi simulates spontaneous and induced fission events by sampling the full neutron multiplicity distributions.

A “multiplicity patch” to MCNP5 1.6.0 was also independently developed by Los Alamos National Laboratory (LANL) that similarly samples the full spontaneous and induced fission neutron multiplicity distributions. [6]

MCNPX-PoliMi employed Terrell’s model of the spontaneous and induced fission neutron multiplicity distribution, adjusted for the evaluated value of \bar{v} contained in the cross section library. [7] The MCNP5 multiplicity patch employed a different model of the neutron multiplicity distributions that preserves the factorial moments. [8] All simulations shown in this paper employed the ENDF/B-VII cross section library. [2]

The input decks for both sets of simulations were also independently developed by UM and LANL.

Both MCNPX-PoliMi and the MCNP5 multiplicity patch tallied $^3\text{He}(\text{n},\text{p})$ reactions in the proportional counters in list mode: the cell number and time of each detection event was recorded. Post-processors to accumulate the multiplicity distribution of neutron detection events were independently developed by UM and LANL.

RESULTS

“Baseline” Simulations

Monte Carlo simulations of each of the 6 measurements yielded substantial errors in the calculated neutron multiplicity distribution. Fig. 3 illustrates the error for the plutonium source reflected by 38.1 mm of HDPE. The mean (i.e., the first moment) of the calculated multiplicity distribution is 15% higher than the measured mean. The variance (i.e., the second central moment) of the calculated distribution is 35% higher than the measurement. Observe that, as Fig. 3 shows, near-identical errors in the simulations were observed using MCNPX-PoliMi and the LANL MCNP5 multiplicity

patch. Furthermore, both Monte Carlo codes (and their associated post-processors) exhibited near-identical errors for all 6 measurements. In every case, the calculated distribution was displaced to the right of and wider than the measured distribution. In other words, the mean and variance were consistently overestimated by the calculations. In addition, the magnitude of the relative error in of the calculated mean and variance increased with increasing neutron multiplication.

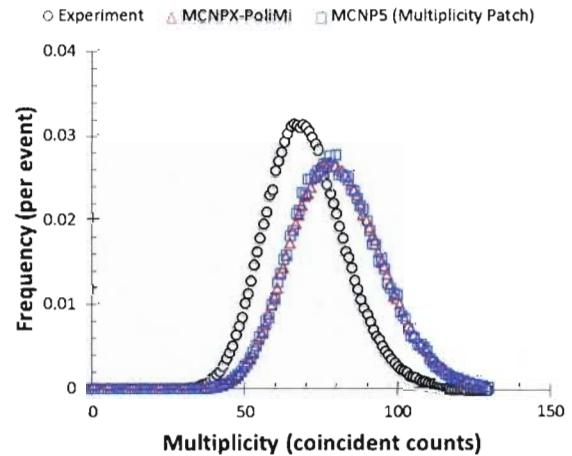


Fig. 3. The neutron multiplicity distribution measured for the plutonium sphere reflected by 76.1 mm of polyethylene using a 4096 μ s coincidence gate width; simulations of this experiment using MCNPX-PoliMi and a multiplicity patch to MCNP5 yield near-identical errors.

Recall that both simulation codes, and their respective post-processors, were independently developed by UM and LANL. Consequently, it is unlikely that the simulation or post-processing logic implemented by either code is the cause of the observed errors.

Sensitivity Study

Other potential sources of error in the Monte Carlo simulations include:

- Errors in the material and/or geometry models of (a) the plutonium source, (b) the HDPE reflectors, or (c) the neutron multiplicity counter;
- Inadequate treatment of dead-time in the ^3He proportional counters and associated electronics;
- Inaccuracy in the plutonium source density; and/or
- Errors in the evaluated values of fundamental fission physics parameters, e.g. ^{239}Pu \bar{v} .

The sensitivity of the calculated neutron multiplicity distribution was evaluated for each of the preceding factors.

In addition to the 6 measurements of the plutonium source, 6 similar measurements of a ^{252}Cf point source

were conducted where the source was suspended in the center of each HDPE reflector. Fig. 4 compares the measured and calculated neutron multiplicity distribution for the ^{252}Cf source reflected by 38.1 mm of HDPE. The MCNPX-PoliMi simulation shown in Fig. 4 is very similar to the measured neutron multiplicity distribution. The calculated mean and variance of the neutron multiplicity distribution differ from the measurement by only 2.5%. Similar favorable comparisons were observed for all 6 cases. Consequently, it appears there are no substantial errors in the material and geometry models for the HDPE reflectors or the multiplicity counter.

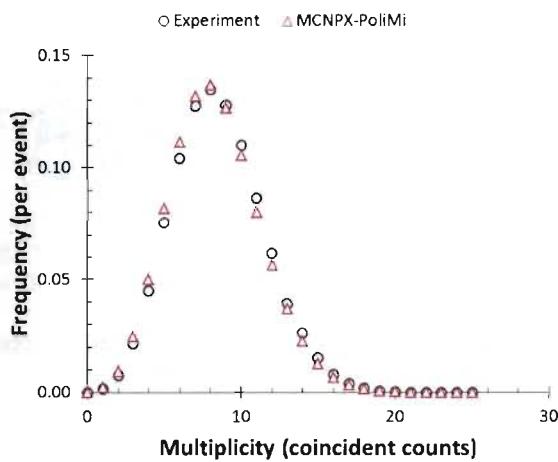


Fig. 4. The neutron multiplicity distribution measured for a californium-252 source reflected by 76.1 mm of polyethylene using a 4096 μs coincidence gate width; simulation of this experiment using MCNPX-PoliMi produces essentially the same distribution.

Therefore, the errors in the Monte Carlo simulations most likely resulted from errors in (a) the material model of the plutonium source; (b) the source-detector distance; (c) the detector dead-time; and/or (d) the simulation of fission chain reactions in the plutonium source. UM conducted sensitivity studies to evaluate each of the preceding potential sources of error.

The mass and isotopic composition of the plutonium source were measured very precisely during its fabrication. [1] However, the actual density of the plutonium source is uncertain (due to thermal expansion via heat generation resulting from alpha-decay). The density of the plutonium sphere was varied in MCNPX-PoliMi simulations from its maximum theoretical density to the minimum density that would cause the plutonium sphere to fill the entire volume interior to the steel cladding. No single consistent change in the plutonium density was found to correct the errors in the MCNPX-PoliMi simulations of all 6 measurements. Consequently,

inaccuracy in the simulated plutonium density is unlikely to be the cause in the simulation errors.

Furthermore, note that altering the plutonium density is equivalent to changing the plutonium cross sections by a scalar multiplicative factor. Therefore, if there are errors in the plutonium cross sections, it is unlikely they can be corrected by a scalar multiplicative factor.

The source-detector distance is known with ± 5 mm tolerance. The source-detector distance was increased by as much as 30 mm in MCNPX-PoliMi simulations, but no plausible error in the modeled source-detector distance adequately corrected the simulated neutron multiplicity distribution. Consequently, it is highly unlikely that errors in the modeled source-detector distance caused the observed errors in the Monte Carlo simulations.

The detector dead-time was measured to be approximately 5 μs per channel. In the baseline simulations, both post-processors treated each channel as dead for 5 μs following a neutron count to account for the effect of dead-time on the accumulated neutron multiplicity distribution.

In the sensitivity study, the modeled detector dead-time was increased up to 80 μs . However, only implausibly enormous errors in the dead-time (greater than 40 μs) corrected the calculated multiplicity distribution, and no consistent dead-time correction could be found for all 6 measurements. Therefore, incorrect treatment of dead-time is not responsible for the error in the calculated neutron multiplicity distribution.

After eliminating the preceding potential sources of error (or at least establishing that they are extremely unlikely), the sensitivity of the calculated neutron multiplicity distribution to the evaluated value of ^{239}Pu induced fission \bar{v} was evaluated. For this 93%- ^{239}Pu metallic source, very slight errors in the value of induced fission \bar{v} can induce substantial errors in the calculated neutron multiplicity distribution, and those errors will become more significant with increasing neutron multiplication. This trend is consistent with the observed trend in the calculated neutron multiplicity distribution.

In the sensitivity study, the MCNPX-PoliMi simulations were repeated with the value of ^{239}Pu \bar{v} decreased relative to its ENDF/B-VII evaluated value by 1% to 3%.

A scalar multiplicative reduction (over all incident neutron energies) of 1.1% was found to consistently correct the calculated neutron multiplicity distribution such that it compared favorably with the measured distribution for all 6 measurements. In terms of the variance of the distribution, the errors in the calculated distribution ranged between only -11.6% and +9.5%. Errors in the mean ranged between -9.1% and +6.7%.

Fig. 5 depicts the multiplicity distribution calculated by MCNPX-PoliMi for a reduction of only 1.1% in ^{239}Pu \bar{v} . The calculated multiplicity distribution closely matches the measured distribution for this case, and the

same correction to \bar{v} induces the calculated distribution to compare favorably to the measurement in all of the other 5 cases. Consequently, a single small scalar multiplicative correction to the ENDF/B-VII evaluated value of ^{239}Pu \bar{v} appears to be sufficient to correct the MCNPX-PoliMi simulations for all 6 measurements.

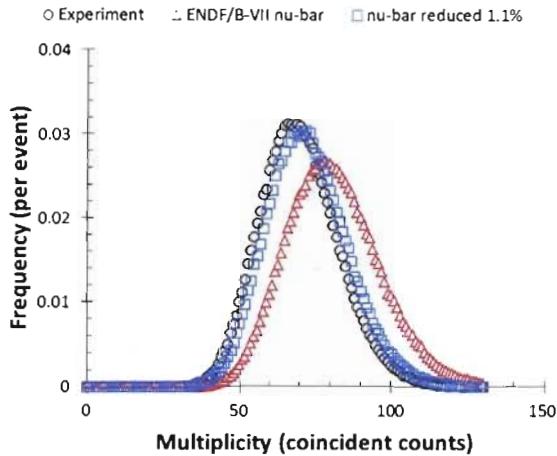


Fig. 5. The neutron multiplicity distribution calculated for the plutonium sphere reflected by 76.1 mm of polyethylene using a 4096 μs coincidence gate width; a reduction of 1.1% in the ENDF/B-VII evaluated value of ^{239}Pu \bar{v} yields a calculated neutron multiplicity distribution very close to the measurement

CONCLUSION

Monte Carlo simulations of neutron multiplicity counting experiments with highly multiplicative, subcritical assemblies of polyethylene-reflected plutonium metal exhibited significant errors in the calculated multiplicity distribution. The observed errors in the calculated multiplicity distribution do not appear to result from (a) logic employed in the simulation of fission chain reactions, (b) errors in the material and/or geometry models of the experiments; or (c) multiplicity counter dead-time. Instead, a sensitivity analysis of the preceding factors indicates that the errors in the calculated multiplicity distributions may result from very small errors in the fundamental fission physics parameters for ^{239}Pu , namely, the evaluated value of the induced fission prompt neutron multiplicity (\bar{v}) of ^{239}Pu .

A simple scalar adjustment of -1.1% applied to the ENDF/B-VII evaluated value of ^{239}Pu \bar{v} for all incident neutron energies was found to consistently correct the calculated neutron multiplicity distribution for every reflected configuration of the plutonium source. Although this correction factor indicates the magnitude of the possible error in ^{239}Pu \bar{v} , we acknowledge that a simple scalar multiplicative factor is probably not the

correct functional form for a correction to \bar{v} . In future work, we plan to investigate energy-dependent adjustments to \bar{v} .

REFERENCES

1. J. MATTINGLY, "Polyethylene-Reflected Plutonium Metal Sphere: Subcritical Neutron and Gamma Measurements," SAND2009-5804-R2, Sandia National Laboratories (2009).
2. P.G. YOUNG, et al., "Evaluation of Neutron reactions for ENDF/B-VII: $^{232-241}\text{U}$ and ^{239}Pu ," *Nuclear Data Sheets*, **108**(12), 2859 (2007).
5. E. BRANDON, "Assembly of ^{239}PU Ball for Criticality Experiment", CMB-11-FAB-80-65, Los Alamos National Laboratory (1980).
3. X-5 MONTE CARLO TEAM, "MCNP — A General Monte Carlo N-Particle Transport Code, Version 5," LA-UR-03-1987, Los Alamos National Laboratory (2003).
4. S.A. POZZI, E. PADOVANI, and M. MARSEGUEARRA, "MCNP-PoliMi: A Monte Carlo Code for Correlation Measurements," *Nucl. Instrum. Meth. A*, **513**, 550 (2003).
5. D. PELOWITZ (editor), "MCNPX™ User's Manual, Version 2.7.0," LA-CP-11-00438, Los Alamos National Laboratory (2011).
6. C.J. SOLOMON, "Polyethylene Reflected Pu Multiplication Inference Simulations," LA-UR-11-03933, Los Alamos National Laboratory (2011).
7. J. TERRELL, "Distributions of Fission Neutron Numbers," *Phys. Rev.*, **108**(3), 783 (1957).
8. J.P. LESTONE, "Energy and Isotope Dependence of Neutron Multiplicity Distributions," LA-UR-05-00288, Los Alamos National Laboratory (2005).