

## Stochastic Models for Fast Neutron Multiplicity Counting of Plutonium

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**Abstract** - This paper compares simulated prompt, time-correlated, neutrons emitted by plutonium metal plates and detected by organic scintillators to experimental data, measured at the Zero Power Physics Reactor of Idaho National Laboratory. These experimental results are compared to simulations with the MCNPX and MCNPX-PoliMi Monte Carlo codes. The number and energy of the neutrons emitted by each fission was simulated using a standard bounded integer approach and two more advanced models.

### I. INTRODUCTION

Neutron multiplicity counting is a well-established measurement modality for characterizing and verifying fissile material [1]. It has been used for many years both in passive systems for plutonium, and in active systems for uranium. The technique is applied to the assay of materials such as fresh and spent nuclear fuel, scrap nuclear metal, and MOX fuel. The instruments are traditionally based on thermal neutron detection using <sup>3</sup>He neutron capture detectors; however, <sup>3</sup>He has recently entered a state of severe scarcity, resulting in the need of new instruments for the verification of fissile mass in international safeguards.

At the University of Michigan we simulated, designed, and built a measurement system to be used as an alternative to <sup>3</sup>He-based systems for the assay of special nuclear materials [2,3]. The new system uses multiple organic scintillation detectors, including EJ-309 liquid and new stilbene scintillators [4], and digital acquisition with on-the-fly data processing [5]. This new approach provides shorter die-away time, faster sample assay times, and more accurate energy information.

In this paper, the stochastic simulations used to model an experiment performed on plutonium metal at Idaho National Laboratory in 2015 is described. We evaluate the performance of different fission models and compare them to experimental data for several experimental configurations.

### II. DESCRIPTION OF THE ACTUAL WORK

#### 1. Monte Carlo Fission Model

Early versions of Monte Carlo codes were developed during the Manhattan project to simulate criticality in fissile systems [6]. For this reason, they employed a significant assumption to the number of neutrons emitted from fission: only an integer number of neutrons immediately surrounding the average (nubar) could be emitted. For example, if a fission occurred with nubar of 2.6, the code could only emit 2 neutrons (40% of the time) or 3 neutrons (60% of the time). This so-called “bounded-integers”

approach was maintained for several decades, until Valentine at ORNL introduced full multiplicity distributions in MCNP-DSP [7]. More advanced fission models are being developed in the MCNPX-PoliMi code [8].

#### 2. Experiments at Idaho National Laboratory

We conducted extensive experiments on well-known fissile samples at Idaho National Laboratory. The composition of the plutonium metal plates is listed in Table 1.

Table I. Composition of one plutonium metal plate, PANN series.

Isotope	Mass (g)
Pu-238	2.33E-04
Pu-239	98.89
Pu-240	4.70
Pu-241	0.04
Am-241	0.23
Al nat	1.16

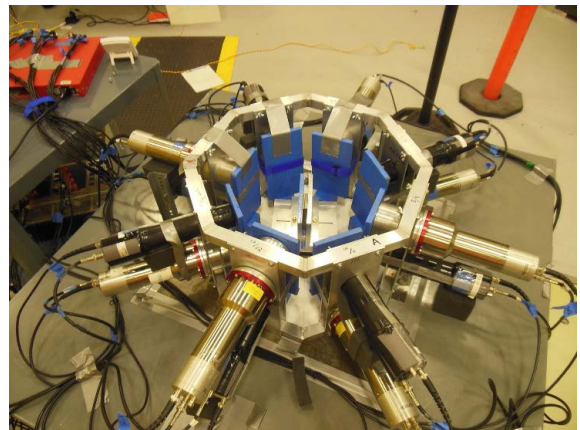


Fig. 1. Photograph of the experimental setup showing the Fast Neutron Multiplicity Counter and the plutonium plates (center) at Idaho National Laboratory, 2015.

The fast neutron multiplicity counter consists of 16 detectors (8 liquid scintillators and 8 stilbene detectors), arranged in two rings. The detectors are read-out by a fast digitizer, model V1730 manufactured by CAEN (14 bits and 500 MHz). The pulse processing includes pulse shape discrimination and time correlations between detectors. This approach allows us to analyze time correlations between neutrons and gamma rays. In this paper, we focus on the neutron-neutron time correlations. The acquisition system achieved a readout rate of approximately 60 MBps, with a negligible dead-time, corresponding to approximately 140,000 waveforms per second. We implemented an acquisition-in-coincidence readout logic, which drastically reduced the readout throughput due to random background pulses. This readout logic validates and transmits to the workstation only those pulses which occur within a 40 ns coincidence window. However, all the pulses detected in coincidence by top-bottom nearest neighbor detectors were rejected. This readout method is meant to reject most of the spurious coincidence events due to cross-talk.

### III. RESULTS

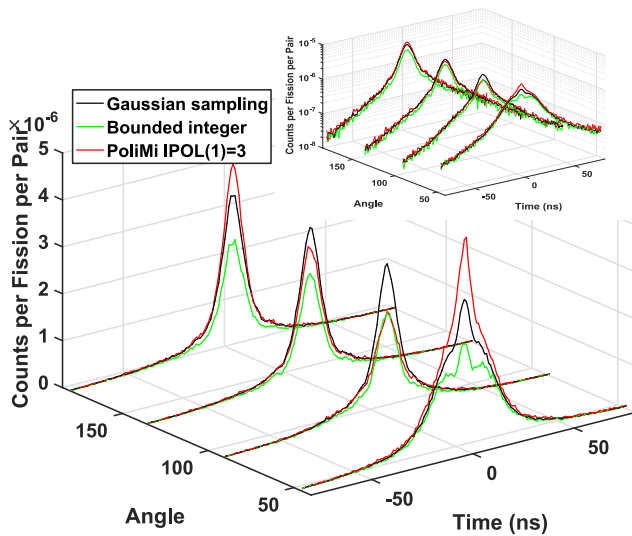


Fig. 2. Neutron-neutron cross-correlation functions for the detector pairs as a function of angle between detectors for a  $^{240}\text{Pu}$  point-like source, simulated using MCNPX-PoliMi model (red line) and the bounded integer model (black line).

Fig. 2 shows the simulated neutron-neutron, cross correlation functions as a function of time delay for all detector pairs for a  $^{240}\text{Pu}$  point-like source. Three models were used to simulate the fission emitted neutrons. The first approach relies on MCNPX-PoliMi, where the angular distribution of neutron emission depends on the direction of the light fission fragment. The first parameter of IPOL card in MCNPX-PoliMi allows selecting one of the available built-in sources. In this case IPOL(1) = 3 was used, which corresponds to a  $^{240}\text{Pu}$  source and the energy of the emitted neutrons depends on the multiplicity [9]. The second

approach relies on a bounded integers model, which assumes an integer number of neutrons per fission. In this case the neutrons are emitted isotropically, and their energy does not depend on the multiplicity. The bounded integer treatment is obtained in the MCNPX 2.7.0 code simulating a spontaneous fission source and an integer number of neutrons emitted per fission, i.e. for Pu-240 (spontaneous fission nu<sub>bar</sub> 2.16), the number of neutrons is two 84% of the time and three 16% of the time (SDEF PAR=SF and PHYS:N FISM=4 FMULT 94240 SFNU=0 0 0.840 1.0). The FMULT card controls the bounded integer treatment and only applies to spontaneous fission. The third approach relies on Gaussian sampling, where nu<sub>bar</sub> is sampled from a Gaussian distribution and the sampling is corrected to preserve the average multiplicity [10]. The bins of the cumulative probability distribution used in this case are: 0.063 0.295 0.628 0.881 0.980 0.998 1.000, from 0 to 6 neutrons [11].

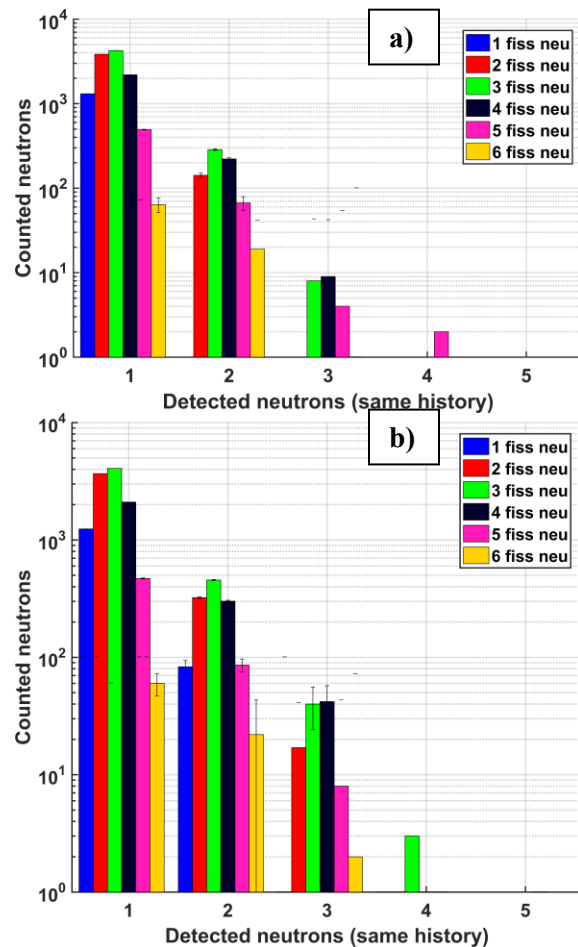


Fig. 3. Simulated  $^{240}\text{Pu}$  neutrons detected in correlation (MCNPX-PoliMi model). The contribution due to each multiplicity group is color coded. Cross-talk events are rejected in Fig.3.a.

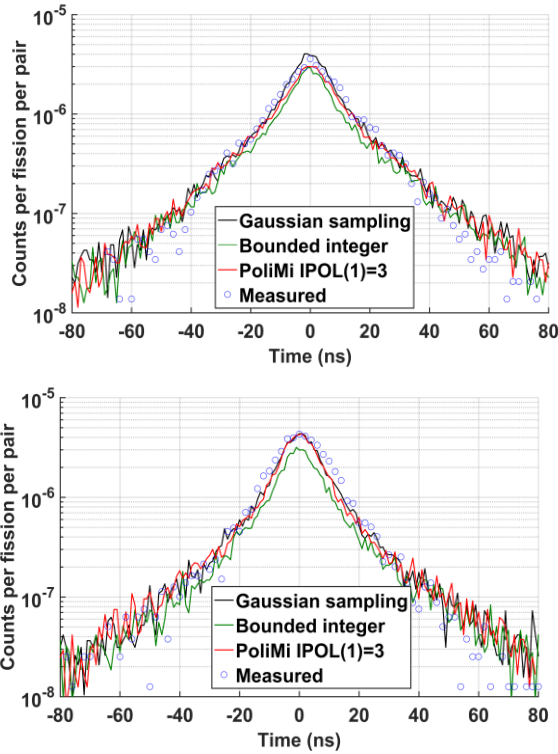


Fig. 4. Time distribution of simulated and measured neutron-neutron correlations emitted by one plate for all detector pairs, for angles of (top) 90 deg and (bottom) 180 deg.

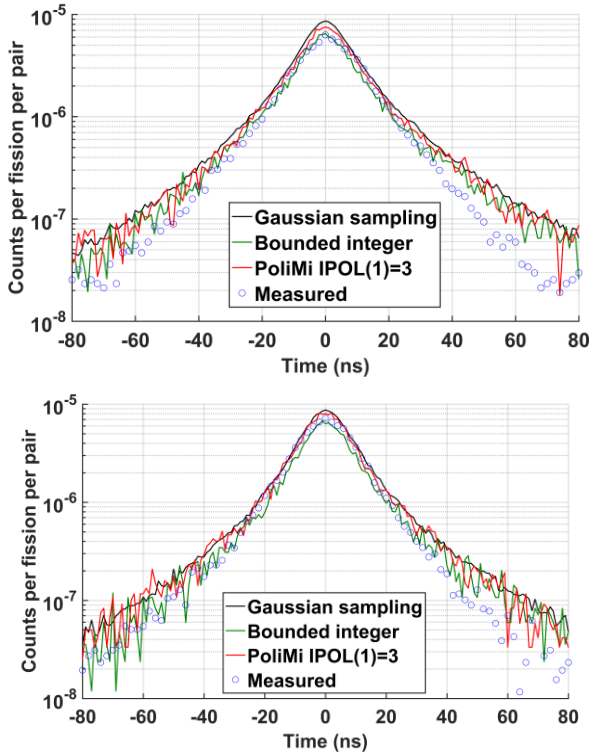


Fig. 5. Time distribution of simulated and measured neutron-neutron correlations emitted by nine plates for all detector pairs, for angles of (top) 90 deg and (bottom) 180 deg.

detector angles of 45, 90, 135 and 180 deg. The magnitude of the distribution simulated using the MCNPX-PoliMi model is higher for 180 degrees, compared to closer angles, because neutrons are more likely emitted in the direction of the fission fragments. As expected, both the Gaussian sampling and the bounded integer treatment yield a fairly constant magnitude of the cross-correlation distribution, as a function of angle. A broader cross-correlation distribution can be noticed at 45 degrees, which is due to the contribution of cross-talk neutrons, i.e. neutrons detected within the coincidence window because of scattering between two adjacent detectors. The bounded integer model underestimates the magnitude of the distribution simulated using both the MCNPX-PoliMi model and the Gaussian treatment. This is probably due to the fact that double neutrons are likely detected by the well counter not only from fissions that emit 2 or 3 neutrons, but also from fissions emitting higher order multiplets, as shown in Fig. 3. Fig. 3 shows the distribution of neutrons detected in coincidence, by discriminating them by the number of emitted neutron in the respective fission reaction which produced them. In Fig. 3a, coincidence due to scattering between detectors, i.e. spurious coincidences due to cross talk, have been removed.

Fig. 4 shows the measured and simulated cross-correlation function for pairs of detectors at 180 and 90 degrees for a single plutonium metal plate; Fig. 5 shows the same results for an assembly of nine plates. The underestimation of correlated counts by the bounded integer model is more relevant for a single plate, compared to 9 plate-assembly, because of the higher contribution of induced fissions in this last case and self-shielding effects. In Fig. 4, a slight asymmetry can be noticed in the cross-correlation plot, this may be due to a misalignment of the single plate.

#### IV. CONCLUSIONS

We compared simulated and experimental results on correlated neutrons detected by pairs of detectors placed in a well counter around plutonium metal plates. We found a reasonable agreement between simulated and measured results both using a Gaussian sampling model for neutron multiplicity and a more complex energy-dependent treatment, implemented by MCNPX-PoliMi. Conversely, correlated counts are significantly underestimated when a bounded integer approach is used to predict the number of emitted neutrons by each fission event. The simulated results showed that the dependence of the number of correlated detections as a function of angle is well captured only by the MCNPX-PoliMi model. The angular dependence is more evident when a point-like source is measured.

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