Correlated Neutron Emissions from ^{252}Cf

Sara A. Pozzi, Brian Wieger, Andreas Enqvist, Shaun D. Clarke, Marek Flaska, Matthew Marcath, and Edward Larsen

University of Michigan, Department of Nuclear Engineering and Radiological Sciences
Ann Arbor, Michigan

Robert C. Haight

Los Alamos National Laboratory
Los Alamos, New Mexico

and

Enrico Padovani

Polytechnic of Milan, Department of Energy
Milan, Italy

Abstract — *This paper presents new experimental results of correlated, prompt neutron emission from the spontaneous fission of ^{252}Cf. Specifically, we present correlated-neutron emission probabilities and average energies for two detected neutrons as a function of the angle between the two neutrons. Experimental results are compared to several Monte Carlo models that include the number, energy, and angular distributions of prompt neutrons from fission.*

I. INTRODUCTION

Fission generates multiple prompt neutrons emitted in coincidence: In ^{240}Pu spontaneous fission, >70% of the fissions emit more than one neutron; for ^{252}Cf spontaneous fission, that number is 97% (Ref. 1). The vast majority of fissions are binary fission, where most prompt neutrons are subjected to a Lorenzian increase in their velocities due to emission from accelerating or fully accelerated fission fragments.[2] This effect causes an angular dependence (anisotropy) of the emission probabilities and energy-angle correlations of neutrons emitted in a fission event. These effects have not yet been quantified in detail. Prior work has examined the theory of neutron and photon emission from excited fission fragments[3–5] and of the ratio of pre-scission to post-scission prompt neutrons emitted from a ^{252}Cf source.[6,7] Very few experiments have been performed to measure the neutron-neutron angular and energy correlations.[8]

Correlated neutrons emitted from individual fission events have been investigated since the beginning of nuclear-related activities following the discovery of fission.[9] Research efforts have continued on this topic to measure these nuclear data for various applications.[10] Correlated neutrons can be measured using multiple-detector systems, and this technique has been applied widely in nonproliferation and safeguards applications.[11–14] In one approach, the angular anisotropy of neutron emission in fission is used to determine the ratio of (α,n) to spontaneous fission activity in a fissile sample.[15] These detection systems are used to characterize special nuclear materials (plutonium and highly enriched uranium), which undergo spontaneous and induced fission and thus generate correlated neutrons.

The present work gives experimental results on pairs of correlated prompt neutrons from fission. Detection probability and average detected energy are presented as a function of angle between the neutrons. Specifically, we present new measurements of the number and average energy of correlated neutrons detected using liquid scintillators placed at several angles about a ^{252}Cf source. These experimental results are compared to simulations performed with the MCNPX and MCNPX-PoliMi Monte Carlo codes.

II. MEASUREMENT SETUP

1 The measurement setup (see Fig. 1) consists of an array of 14 EJ-309 liquid scintillation detectors, arranged in a ring around a ^{252}Cf source directly on a thin metal table. Each cell is cylindrical and has dimensions of 7.62-cm diameter $\times$ 5.08-cm length and is read out by 7.62-cm-diam ET-Enterprises 9821B photomultiplier tubes (PMTs). The distance between the source and the front face of each of the detectors was set to 20 cm. The setup allows the measurement of coincident neutrons for angles between detectors of 26, 51, 77, 103, 128, 153, and 180 deg. Because of the dimensions of the active volume of the detectors, the variance of the angle of correlation is $\sigma^2 \approx 6.5^2$. Small pieces of low-density polyurethane foam (ρ = 0.0256 g/cm^3) were placed between the detectors for spacing and for source support.

The liquid scintillators are sensitive to fast neutrons and gamma rays from the source. Two CAEN V1720 digitizers (12-bit dynamic range and a sampling frequency of 250 MHz) were used in this measurement. The digitized pulses were recorded and analyzed in time coincidence using a 60-ns time window, whereas the pulses themselves were recorded in a 480-ns time window to allow for accurate pulse-shape discrimination (PSD).

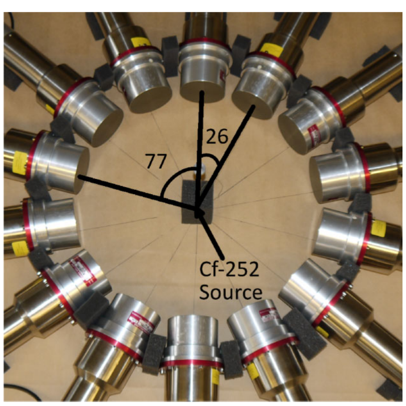


Fig. 1. Photograph of the experimental setup for the ^{252}Cf measurements. A ring of 14 liquid organic scintillators is placed around a source. Two angles are explicitly shown: 26 and 77 deg.

The detectors were first calibrated using a ^{137}Cs source to ensure a uniform response. Figure 2 shows the results of the calibration for the 14 detectors used in the experiment. A threshold of 40 keVee was chosen to ensure good PSD. For this liquid scintillator, this threshold corresponds to $\sim$500 keV of neutron energy deposited. The ^{252}Cf was placed at the center of the assembly, and data were acquired for a measurement time of 5 h with source 1 and 100 h with source 2.

Two ^{252}Cf sources were measured independently with this setup. The manufacture date and intensity at the time of the measurement were 1994 and 56 000 fissions/s for source 1, and 2005 and 4800 fissions/s for source 2. Sources 1 and 2 are not packaged in the same way. Source 1 has a smaller casing than source 2, i.e., 1-cm diameter and 2-cm length for source 1, and 3.2-cm diameter and 2-cm length for source 2. In both cases, the casing has a modest effect on the outgoing neutrons from fission. The two sources were manufactured at Oak Ridge National Laboratory and were initially loaded with $\sim$82% ^{252}Cf and $\sim$10% ^{250}Cf by weight (Ref. 16). Because of the longer half-life of ^{250}Cf when compared to ^{252}Cf, the fraction of fission neutrons originating from ^{250}Cf will increase with time.[17] Based on the initial composition and branching ratios for these sources, $\sim$14.5% and 1.8% of fissions are from ^{250}Cf for sources 1 and 2, respectively.

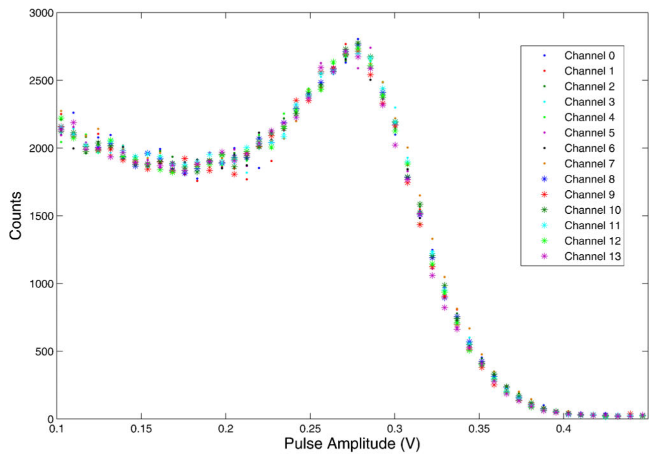
III. RESULTS

The measurement of correlated neutron emissions from fission events using liquid scintillators requires PSD and fast timing analysis. Sections III.A through III.D describe the analysis applied to the digitized data and the results obtained following that analysis.

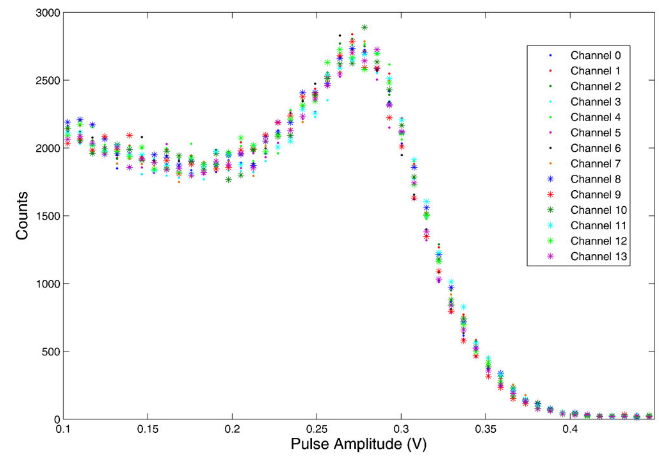
III.A. Pulse-Shape Discrimination

Three optimized, off-line, digital PSD algorithms were applied to the measured data.[18] Figure 3 shows the results of these algorithms applied to ^{252}Cf data acquired with one of the detectors used in the experiment. The black lines show the discrimination curves used to separate neutrons from gamma rays. All detectors yielded similar results. Figure 3a shows the digital equivalent of PSD by charge integration, which gives good discrimination over a wide range of pulse heights; however, at small pulse heights, the discrimination becomes poor. The PSD methods shown in Figs. 3b and 3c improve the discrimination of small-amplitude pulses by using a different metric. When used in series, the three methods provide a lower misclassification rate than that obtained when using the charge integration method alone.

A time-of-flight experiment conducted at Los Alamos Neutron Science Center[19] (LANSCE) was used to quantify the effect of applying the three PSD methods.

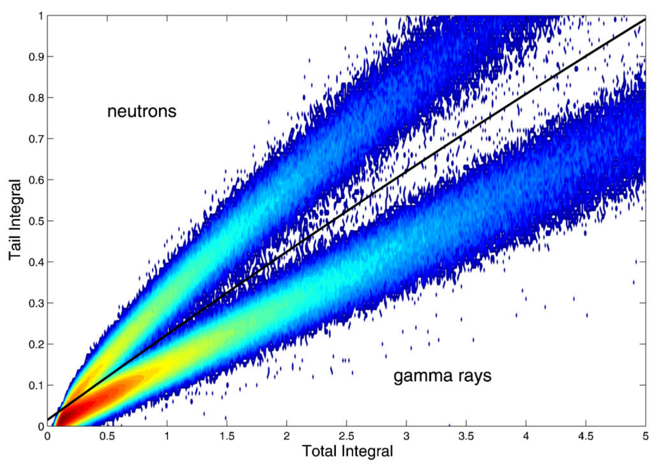


(a)

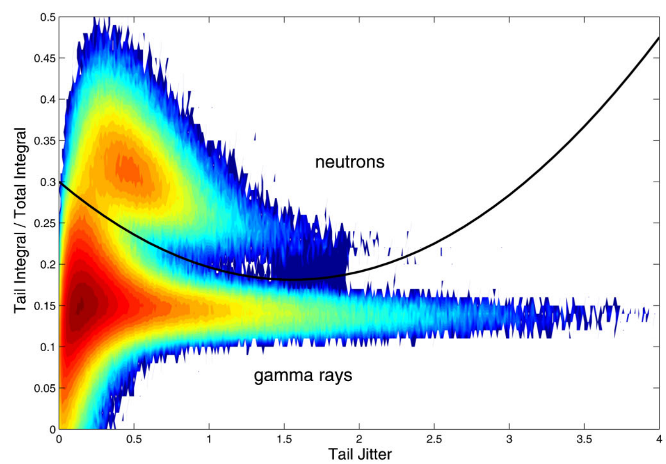


(b)

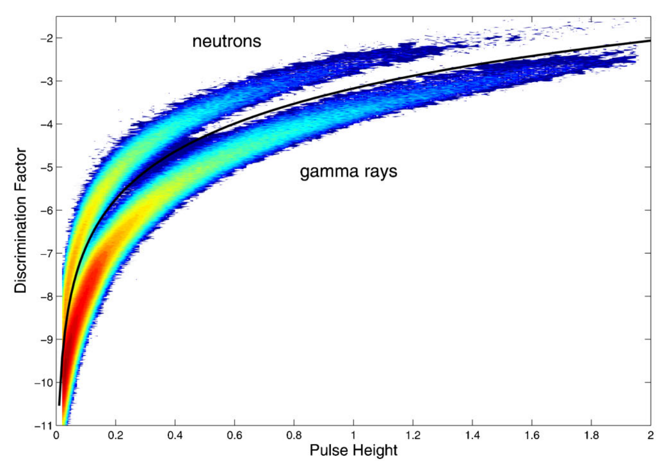
Fig. 2. Cesium-137 calibration of 14 liquid scintillation detectors at (a) the beginning and (b) the end of the data acquisition.



(a)



(b)



(c)

Fig. 3. Pulse-shape discrimination techniques applied to ^{252}Cf data: (a) charge integration, (b) pulse-tail analysis, and (c) simplified digital charge collection.

A discussion of the PSD algorithms shown in Figs. 3a and 3c can be found in Refs. 20 and 21, respectively. The PSD method in Fig. 3b uses the fluctuations in the data in the tail of each pulse: Neutron pulses have slightly more jitter in their tails. The normalized difference in adjacent pulse-tail points is calculated and plotted versus the ratio of tail-to-total integrals; the tail and total ranges are the same as used in the charge integration PSD method shown in Fig. 3a. The results from that experiment are shown in Fig. 4, where neutrons appearing in the gamma-ray peak result from misclassification of photons to neutrons. The misclassification rate is 1.51% when one PSD algorithm is applied and is reduced to 0.88% when the three algorithms are applied. The misclassification rate for gamma rays appearing in the low-energy neutron region is 0.69% with one PSD algorithm and 0.16% with the three algorithms. This improvement is obtained without removing any real neutron counts.

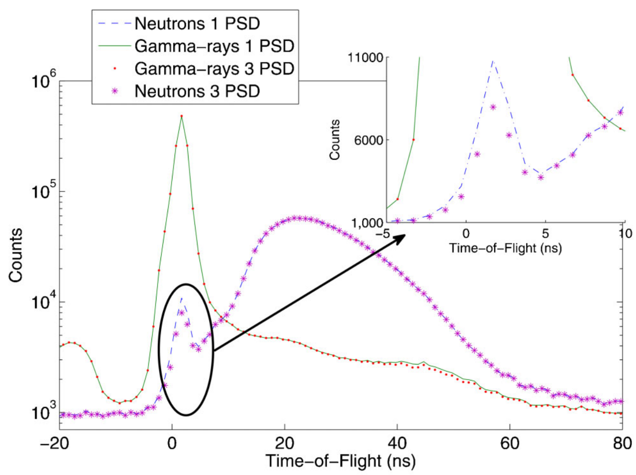
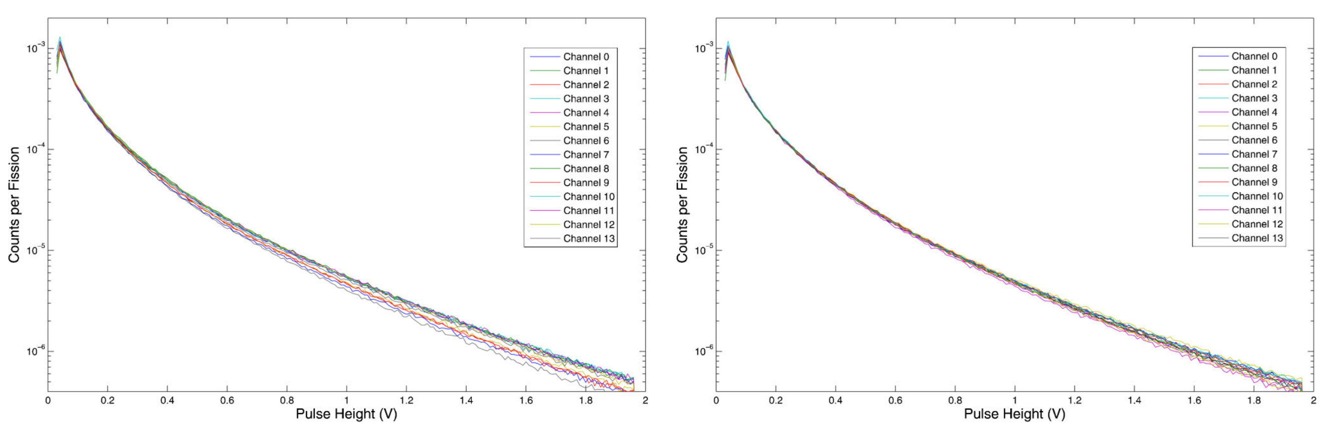


Fig. 4. Time-of-flight distribution obtained at LANSCE (Ref. 19) and close-up showing improvement in photon misclassification after applying one or three PSD algorithms.

After PSD, pulse-height analysis was applied to the neutrons. Figure 5 shows the neutron pulse-height distributions from the individual detectors. The total number of neutrons detected per detector was 1 to 2
1.1×10^7 with source 1 and 1.8 to 1.9×10^7 with 3
source 2. The results show generally good agreement among pulse-height distributions measured in the 14 channels. Source 1 results show a slightly wider distribution in pulse heights, especially for the larger pulse heights. The spread in pulse heights is not believed to be due to a time-related change in the detection system. The measurement with source 2 was significantly longer than that with source 1, and the data did not show this problem. For all measurements, the detectors and high-voltage supply had ample time to stabilize, and care was taken in positioning the source at the center of the assembly. Instead, we believe the spread in pulse heights to be caused by the way the source is packaged inside of its container. Further investigation of this effect is underway. Source 2 results show very good consistency among channels.

III.B. Neutron-Neutron Angular Correlations

Time-dependent cross-correlation functions were measured for pairs of detectors at different angles. In this setup, the angles between pairs of detectors are 26, 51, 77, 103, 129, 154, and 180 deg. If neutrons were emitted isotropically at each fission event, and if cross talk did not exist for this setup, the cross-correlation functions would be identical for detector pairs placed at each of the angles listed above. Because neutrons are not emitted isotropically—rather, preferentially in the direction of the



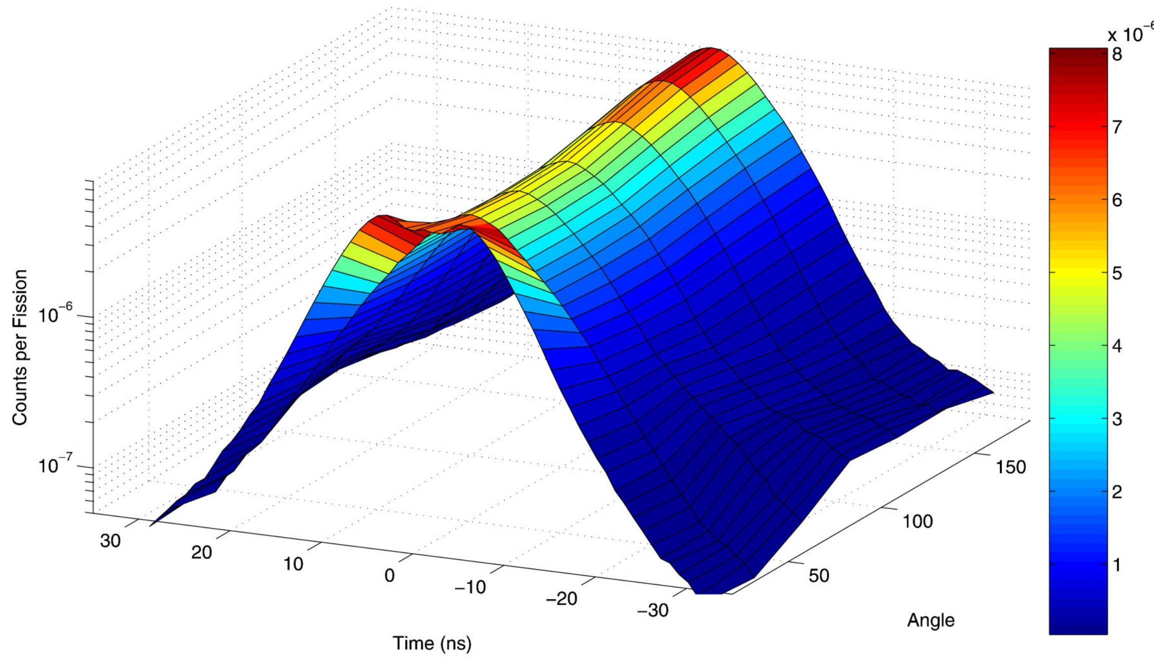
(a) (b)

Fig. 5. Neutron pulse-height distributions for (a) source 1 and (b) source 2.

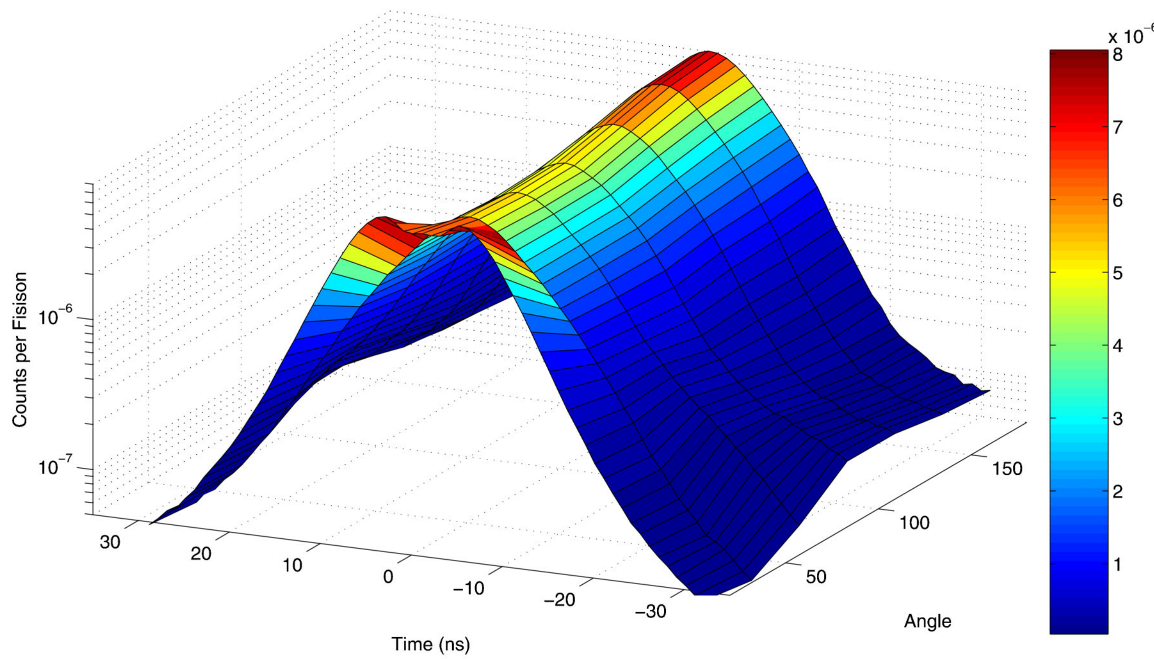
light and heavy fission fragments—and because cross talk exists, the measured cross-correlation functions have a strong dependence on angle.

Cross talk occurs when the same neutron is detected by two or more detectors. It affects primarily the pairs of detectors at the smallest separation angle, 26 deg, because these detectors are neighbors and do not have material between them to prevent a neutron from scattering from one detector to the other.

Figure 6 shows the measured neutron-neutron, cross-correlation functions as a function of time delay for all detector pairs for source 1 (Fig. 6a) and source 2



(a)



(b)

Fig. 6. Measured neutron-neutron cross-correlation functions for the detector pairs as a function of angle between detectors for ^{252}Cf spontaneous fission of (a) source 1 and (b) source 2.

(Fig. 6b). The result shows that correlated neutron detections are more prevalent at small and large angles and less prevalent at ~90-deg angles. This result is in agreement with the fact that in binary fission, neutrons are emitted from fission fragments that are accelerating (or are fully accelerated) in opposite directions. Two neutrons could be emitted from opposite fragments (favoring correlations at 180 deg) or from the same fragment (favoring correlations at 26 deg, in our setup); emission at correlation angles of 90 deg is less probable. This fact is corroborated by our measurement results, which show that for a threshold of 40 keVee (~475-keV neutron energy deposited in the scintillator), the probability of correlated neutron counts at 77 deg is ~71% ± 1% of that of a correlated neutron count at 180 deg. For a threshold of 100 keVee (~830-keV deposited neutron energy), this probability decreases to ~63% ± 1.2%. The error here represents the variation in the amount of correlated neutrons at each angle due to the variance in the angle of correlation. A weighted sine function was fit to the data, and values using the extreme angles of correlation were calculated. These new values give a range of ratios that account for the width of our detectors and give the error on the ratio between the 180- and 77-deg detectors. For both thresholds, the number of correlated neutrons measured at angles of 103, 129, and 154 deg increases gradually, leading up to the 180-deg maximum. Likewise, the number of correlated neutrons measured at angles of 77, 56, and 26 deg increases gradually.

Table I shows the total number of correlated neutron counts per fission for the two ^{252}Cf sources for the seven angles between detectors investigated in this study.

The side "wings" measured at the 26-deg angle result primarily from cross-talk events. Results for other angles do not show this effect, indicating the expected reduction of cross talk for detectors placed at large angles.

Comparison of Figs. 6a and 6b shows good consistency of experimental results for the two sources that were used in the experiment.

III.C. Comparison with Monte Carlo Simulations

We simulated the experiment using MCNPX (Ref. 22) and MCNPX-PoliMi v. 2.0.5 (Ref. 23). These codes offer several models of neutron emission from spontaneous fission events. In the MCNPX code, the neutrons are emitted isotropically, and their energy distribution is not dependent on the number of neutrons emitted in a given fission event. In MCNPX-PoliMi, there are several fission models available for the emission of fission neutrons from spontaneous fission. For all of the models tested in this work, the angular distribution of neutron emission in the laboratory frame of reference depends on the direction of the light fission fragment (which is selected isotropically). In MCNPX-PoliMi, the selection of a built-in source is performed by using the first parameter of the IPOL card.

TABLE I

Correlated Neutron Counts per Detector Pair per Fission for the Two ^{252}Cf Sources and 40- and 100-keVee Thresholds Versus the Angle Between Detectors

	Angle Between Detectors (deg)						
$\times 10^{-4}$	25.7 ± 6.5	51.4 ± 6.5	77.1 ± 6.5	102.8 ± 6.5	128.5 ± 6.5	154.3 ± 6.5	180 ± 6.5
Source 1, 40 keVee	1.43 ± 0.001	0.86 ± 0.0008	0.83 ± 0.0008	0.85 ± 0.0008	0.97 ± 0.0008	1.12 ± 0.0009	1.19 ± 0.0012
Source 2, 40 keVee	1.43 ± 0.0008	0.86 ± 0.0006	0.84 ± 0.0006	0.86 ± 0.0006	0.97 ± 0.0006	1.11 ± 0.0007	1.12 ± 0.001
Source 1, 100 keVee	0.63 ± 0.0007	0.41 ± 0.0005	0.38 ± 0.0005	0.38 ± 0.0005	0.46 ± 0.0006	0.56 ± 0.0006	0.60 ± 0.0009
Source 2, 100 keVee	0.64 ± 0.0005	0.41 ± 0.0004	0.39 ± 0.0004	0.39 ± 0.0004	0.47 ± 0.0004	0.56 ± 0.0005	0.60 ± 0.0007

For IPOL(1)=10, the energy distribution does not depend on multiplicity; for IPOL(1)=1, the energy distribution does depend on multiplicity.[23] The MCNP model accounted for the detectors, the PMTs, the foam between detectors, and the immediate surrounding geometry (walls, floor, and table); it did not include the source packaging.

Figure 7 shows the measured and simulated time distribution of neutron-neutron correlations for detector angles of 77 deg (Fig. 7a) and 180 deg (Fig. 7b). A difference in the shape and magnitude is observed: The wider cross-correlation in the case of neutrons emitted at 77 deg compared to 180 deg indicates a larger distribution of neutron energies. The magnitude of the cross-correlation for 180 deg compared to 77 deg indicates the greater probability of neutron emission in the direction of the fission fragments. Figure 7c shows the relative difference of the time-of-flight distributions of Figs. 7a and 7b, showing a close to 50% increase of correlated pairs of neutrons at ~0-ns time delay for the 180-deg angle compared to the 77-deg angle. The opposite effect can be seen at time delays of approximately ±23 ns, where the probability of correlated pairs is greater by 50% for the 77-deg angle compared to the 180-deg angle.

Figure 8 shows the correlated neutron counts as a function of angle. The measurement results are the integral of the data shown in Fig. 6. As expected, the results from sources 1 and 2 are essentially the same. The results of simulations using several fission models are compared to the measurement results. The results show that the minimum number of correlated neutrons was measured for an angle between detectors of 77 deg. The maximum number of correlated neutrons was measured for an angle of 26 deg. The correlated detections at 26 deg include a large contribution from cross-talk events. We characterized these events using MCNPX-PoliMi and found that for this experimental setup and source, ~33% of the total events for the 26-deg angle are cross-talk events. The correlated events for the 180-deg angle are not affected by cross talk; only a fraction of 1% of correlated events is attributable to cross talk.

A comparison of the simulations with measured data shows that the MCNPX treatment results in a flat (uncorrelated) number of correlated neutron counts as a function of angle, with the exception of the 26-deg angle, where cross-talk contributes heavily to coincidences. The simulation results presented here include the cross-talk contribution. The disagreement observed between MCNPX-PoliMi simulations and measurements for the correlated pairs detected at an angle of 26 deg could be the result of (a) an excessive "boost" to neutrons emitted from one of the fission fragments in our models, resulting

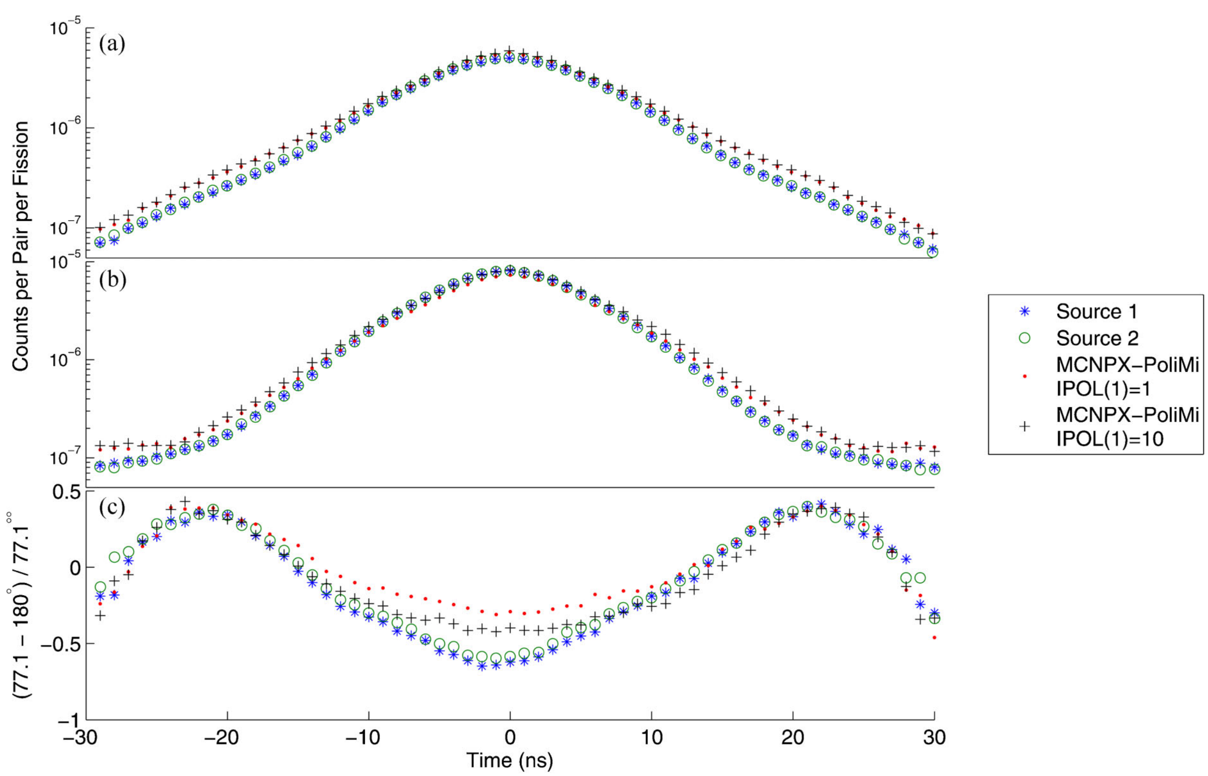


Fig. 7. Time distribution of neutron-neutron correlations for angles of (a) 77 deg and (b) 180 deg and (c) the relative difference between them. Detection threshold is 40 keVee.

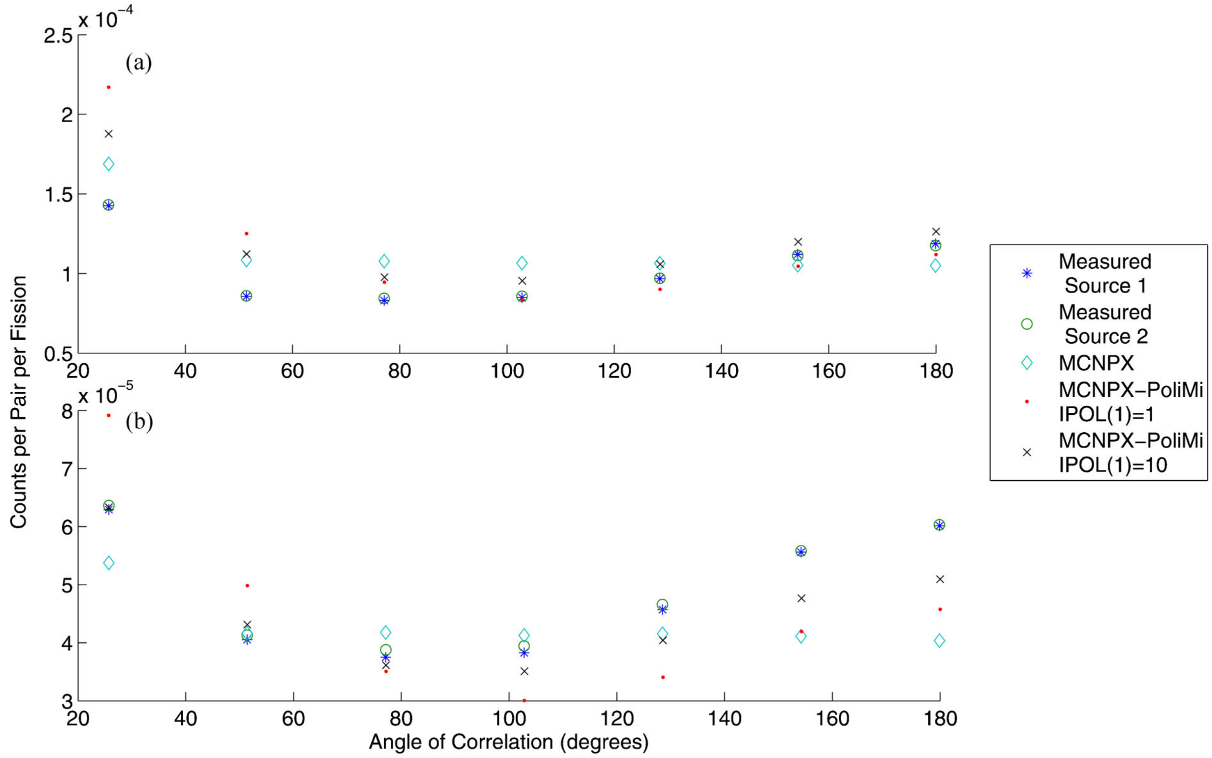


Fig. 8. Measured neutron-neutron correlated counts as a function of angle between detectors for ^{252}Cf spontaneous fission for (a) 40-keVee threshold and (b) 100-keVee threshold. Vertical, statistical error bars are not shown as they are smaller than the symbol used in both simulation and measurement results.

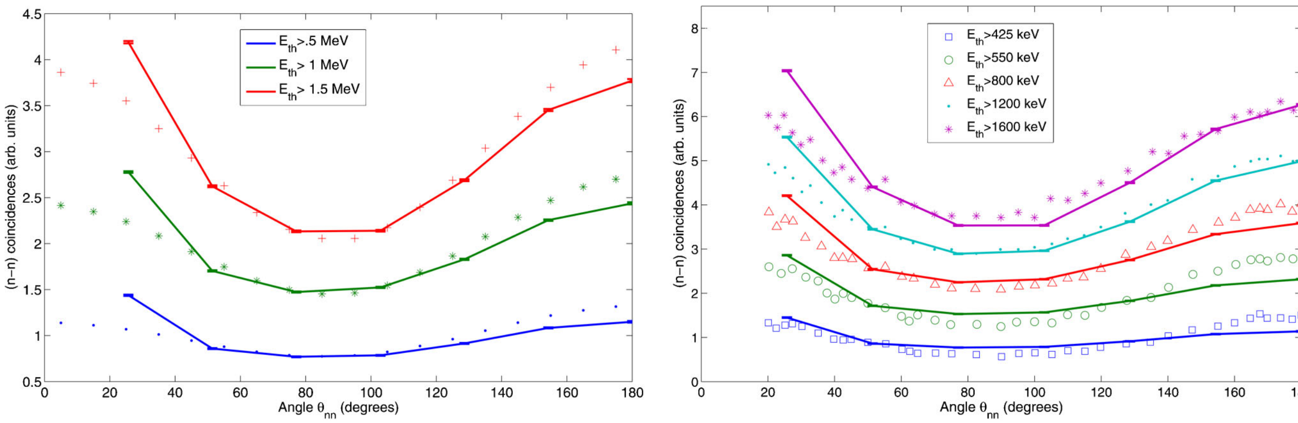
in a higher energy, thereby increasing the probability of the neutron overcoming the detection threshold in both true coincidence events and in cross-talk events and/or (b) inaccurate modeling of the cross-talk effect, which could be due to an incomplete modeling of the detector geometry and materials.

All of the MCNPX-PoliMi treatments show a dependence of the number of correlated neutrons with angle that agrees well with the measured data, with differences among each other of a few percent. The best agreement is obtained with IPOL(1)=10. It is at first surprising to find that the simpler IPOL(1)=10 model yields a slightly better agreement with experimental data than the more complex IPOL(1)=1 model. We believe this effect to occur because the IPOL(1)=10 model uses simpler, but more validated, integral values of nuclear data, such as the neutron energy spectrum. The IPOL(1)=1 model is considerably more complex and relies on newly developed multiplicity-dependent energy spectra. This new model is very much a work in progress, and new experiments will be required to fully validate and improve this more complex model.

Figure 9 shows the correlated neutron counts as a function of angle for several neutron energy thresholds. Figure 9a compares our measurement results with theory.[3] The results show generally good agreement except at small angles between detectors. Here, the disagreement can be explained by cross talk: It affects the measurement but not the theory prediction. Figure 9b compares previous measurements[8] performed with stilbene detectors, shielded with borated polyethylene and lead to prevent cross talk, using various neutron energy thresholds. Good agreement is generally seen between the data presented in Ref. 8 and the current work. The low-angle data show the reduction in cross talk achieved by the shielding used in Ref. 8. The generally good agreement between data sets is encouraging because we used a completely independent approach, including a different detection medium, experimental setup, and analysis approach.

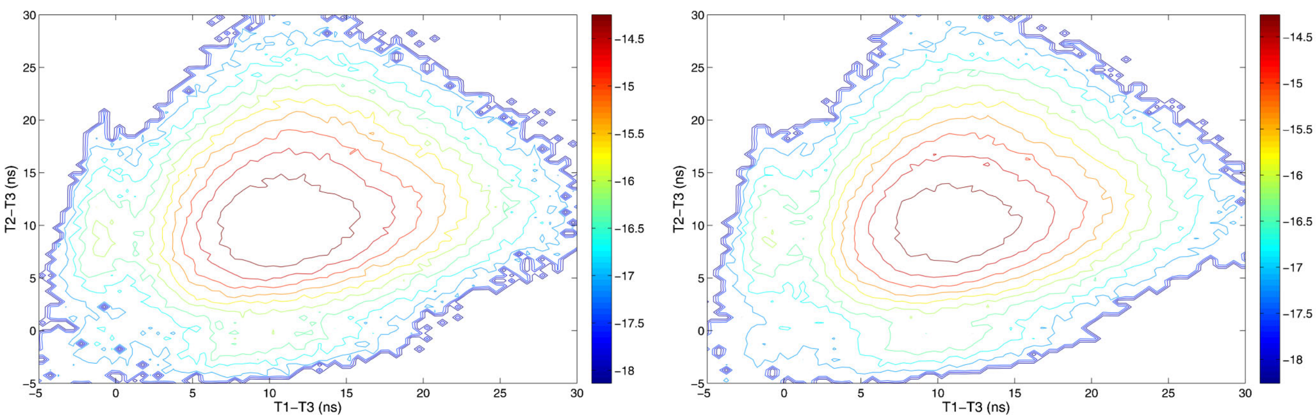
III.D. Neutron-Neutron Energy-Angle Correlations

Figure 10 shows the measured bi-correlation[24] function for gamma-ray neutron-neutron detections in three detectors. The gamma-ray detection provides timing for the fission event and allows the estimation of neutron energy for the two neutrons detected by time of flight. The neutron energy is estimated in the energy range of ~0.5-MeV (lower threshold energy) up to ~5-MeV neutron energy deposited (upper threshold energy). Higher-energy neutrons can also be detected when they deposit less than their full energy in the detector.



(a) (b)

Fig. 9. Measured [this work (solid line)] neutron-neutron correlated counts as a function of angle between detectors for ^{252}Cf spontaneous fission for various energy thresholds compared to results in the literature (symbols): (a) Vogt and Randrup[3] and (b) Petrov et al.[8]



(a) (b)

Fig. 10. Measured gamma-ray neutron-neutron correlated counts: (a) source 1 and (b) source 2 (log correlated counts per fission).

Figure 11 shows the average energy detected for the two neutrons as a function of the angle between them for two detection thresholds. Results show that a variation of ~10% in the average detected neutron energy exists for neutrons emitted at various angles, with the higher neutron energies corresponding to the 180- and 27-deg angles.

Simulation results show generally good agreement with the measured results. MCNPX results show no correlation of neutron energy and angle, whereas all MCNPX-PoliMi treatments show the correct trend between correlated neutron energy and angle. Simulation results with MCNPX-PoliMi treatment IPOL(1)=1 were within 3% for the 40-keVee threshold and within 5% for the 100-keVee threshold. IPOL(1)=10 results were also within a few percent of the measured results, though a slight bias toward higher energies was observed.

IV. CONCLUSIONS

We presented new experimental results on correlated neutrons detected by pairs of detectors placed at a number of angles about a ^{252}Cf spontaneous fission source. The results showed a clear dependence of the number of correlated detections as a function of angle, with the greatest number of correlated detections occurring at the smallest angle (26 deg) and largest angle (180 deg). The ratio of correlated detections at 77 ± 6.5 deg versus 180 ± 6.5 deg was 0.71 ± 0.01 for a measurement threshold of 40 keVee and 0.63 ± 0.012 for a threshold

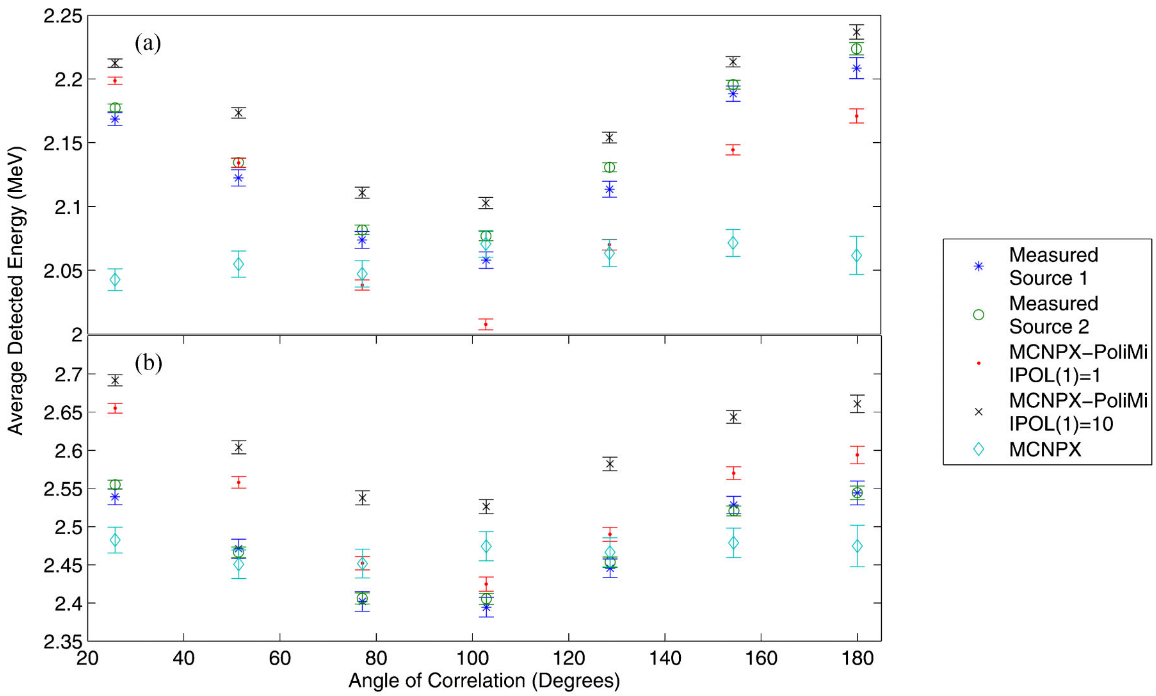


Fig. 11. Average detected energy of neutron-neutron correlated counts as a function of angle between detectors for ^{252}Cf spontaneous fission: (a) a 40-keVee threshold and (b) a 100-keVee threshold.

of 100 keVee. The errors given here represent the change in the values at 77 and 180 deg based on the variance in the angle of correlation due to the size of the detectors.

The average neutron energy of pairs of detected neutrons also depends on the angle between the neutrons. An $\sim$10% higher average neutron energy was observed for angles of 180 deg versus 77 deg.

We compared the experimental data with simulations performed with MCNPX and MCNPX-PoliMi and found reasonably good agreement for all the MCNPX-PoliMi models.

ACKNOWLEDGMENTS

We would like to thank R. Vogt and J. Randrup for sharing the FREYA (fission reaction event yield algorithm) results used in the comparison of experimental to theoretical results and for stimulating discussions. This work was partly funded by the U.S. Department of Energy (DOE), National Nuclear Security Administration, NA-22 award DE-NA0001025 and by the DOE Nuclear Energy University Programs award DE-AC07-05ID14517-00120867.

REFERENCES

1. P. SANTI and M. MILLER, “Reevaluation of Prompt Neutron Emission Multiplicity Distributions for Spontaneous Fission,” *Nucl. Sci. Eng.*, **160**, 190 (2008); http://dx.doi.org/10.13182/NSE07-85.

2. R. VANDENBOSCH and J. R. HUIZENGA, *Nuclear Fission*, Academic Press, New York (1973).

3. R. VOGT and J. RANDRUP, “Event-by-Event Study of Neutron Observables in Spontaneous and Thermal Fission,” *Phys. Rev. C*, **84**, 044621 (2011); http://dx.doi.org/10.1103/PhysRevC.84.044621.

4. R. VOGT and J. RANDRUP, “Event-by-Event Study of Photon Observables in Spontaneous and Thermal Fission,” *Phys. Rev. C*, **87**, 044602 (2013); http://dx.doi.org/10.1103/PhysRevC.87.044602.

5. S. LEMAIRE et al., “Monte Carlo Approach to Sequential Neutron Emission from Fission Fragments,” *Phys. Rev. C.*, **72**, 024601 (2005); http://dx.doi.org/10.1103/PhysRevC.72.024601.

6. N. V. KORNILOV et al., “New Evidence of an Intense Scission Neutron Source in the ^{252}Cf Spontaneous Fission,” *Nucl. Phys. A*, **686**, 187 (2001); http://dx.doi.org/10.1016/S0375-9474(00)00561-3.

7. C. BUDTZ-JØRGENSEN and H.-H. KNITTER, “Simultaneous Investigation of Fission Fragments and Neutrons in ^{252}Cf (SF),” *Nucl. Phys. A*, **490**, 307 (1988); http://dx.doi.org/10.1016/0375-9474(88)90508-8.

8. G. A. PETROV et al., “Search for Scission Neutrons Emitted in Low Energy Fission of Heavy Nucleus,” *AIP Conf. Proc.*, **1175**, 289 (2009); http://dx.doi.org/10.1063/1.3258239.

9. S. DEBENEDETTI, T. W. BONNER, and J. E. FRANCIS, "Angular Correlations of Fission Neutrons," Atomic Energy Commission (1948).

10. M. S. ZUCKER and N. E. HOLDEN, "Evaluating Fission Neutron-Multiplicity Data," *Proc. Symp. Nuclear Data Evaluation Methodology*, Upton, New York, October 12–16, 1992.

11. S. A. POZZI et al., "Pulse-Height Distributions of Neutron and Gamma Rays from Plutonium-Oxide Samples," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **608**, *2*, 310 (2009); http://dx.doi.org/10.1016/j.nima.2009.07.007.

12. S. D. CLARKE et al., "Neutron and Gamma-Ray Cross-Correlation Measurements of Plutonium-Oxide Powder," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **604**, *3*, 618 (2009); http://dx.doi.org/10.1016/j.nima.2009.02.045.

13. H. O. MENLOVE, S. H. MENLOVE, and C. D. RAEL, "The Development of a New, Neutron, Time Correlated, Interrogation Method for Measurement of 235U Content in LWR Fuel Assemblies," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **701**, 72 (2013); http://dx.doi.org/10.1016/j.nima.2012.10.081.

14. J. J. HENKEL and J. T. MIHALCZO, "MCNP-PoliMi Calculations of Cf-252 Measurements for Subcritical HEU Annular Castings," *Trans. Am. Nucl. Soc.*, **101**, 414 (2009).

15. L. HOLEWA et al., "Using Neutron Angular Anisotropy Information to Dynamically Determine the Ratio of the (α,n) Rate to Spontaneous Fission Rate for Coincidence Counting Applications," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **701**, 249 (2013); http://dx.doi.org/10.1016/j.nima.2012.11.026.

16. J. CHAPMAN and S. CROFT, "The Use of 252Cf for Calibrating Safeguards Monitors"; http://www.canberra.com/literature/waste_special_systems/tech_papers/Cf-252Cali-paper.pdf (current as of Nov. 6, 2013).

17. S. CROFT, G. WAKEFIELD, and P. M. J. CHARD, "Topics in Neutron Multiplicity Counting at Harwell," *Proc. ESARDA Int. Workshop Passive Neutron Coincidence Counting*, p. 51, European Safeguards Research and Development Association (1993).

18. B. M. WIEGER et al., "Neutron Multiplicity Distribution Measurements of 235U Induced Fission," *Proc. 54th Annual Mtg. Institute of Nuclear Materials Management*, Palm Desert, California, July 14–18, 2013.

19. A. ENQVIST et al., "Energy-Angle Correlation of Neutron Emission from 235U Induced Fission," *Proc. 54th Annual Mtg. Institute of Nuclear Materials Management*, Palm Desert, California, July 14–18, 2013.

20. M. FLASKA and S. A. POZZI, "Identification of Shielded Neutron Sources with the Liquid Scintillator BC-501A Using a Digital Pulse Shape Discrimination Method," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **577**, *3*, 654 (2007); http://dx.doi.org/10.1016/j.nima.2007.04.141.

21. K. A. A. GAMAGE, M. J. JOYCE, and N. P. HAWKES, "A Comparison of Four Different Digital Algorithms for Pulse-Shape Discrimination in Fast Scintillators," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **642**, 78 (2011); http://dx.doi.org/10.1016/j.nima.2011.03.065.

22. "MCNPX Manual, Version 2.7.0." LA-CP-11-00438, D. B. PELOWITZ, Ed., Los Alamos National Laboratory (Apr. 2011).

23. S. A. POZZI et al., "MCNPX-PoliMi for Nuclear Nonproliferation Applications," *Nucl. Instrum. Methods Phys. Res., Sect. A*, **694**, 119 (2012); http://dx.doi.org/10.1016/j.nima.2012.07.040.

24. J. K. MATTINGLY, "High Order Statistical Signatures from Source-Driven Measurements of Subcritical Fissile Systems," PhD Thesis, University of Tennessee (1998).