This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

SAND2022-8608C

Neutron Capture Gamma-Ray Multiplicity Analysis for Characterization of Moderated Special Nuclear Material

Kyle Polack[a], Michael Hamel[a], Peter Marleau[a], Sean O'Brien[b], Elicia Tiano[a]

[a] *Sandia National Laboratories*[1]

[b] *Oak Ridge National Laboratories*

Prepared for the 63rd INMM Annual Meeting
July 24-28, 2022, Virtual Meeting

Abstract

Neutron multiplicity analysis is commonly used to estimate the mass and multiplication of special nuclear material (SNM) configurations. However, in heavily moderated configurations, neutron leakage is reduced due to thermalization and subsequent capture of neutrons. As neutron leakage is reduced, measurement efficiency decreases, resulting in increased measurement times and larger uncertainty in mass and multiplication estimates based on standard neutron multiplicity analysis. Measuring the characteristic gamma rays produced through thermal neutron capture provides a way to recapture the lost efficiency in heavily moderated systems to better estimate SNM mass and multiplication. Because these gamma rays are produced by the absorption of a neutron, they are representative of the neutron population and can therefore be used as neutron surrogates in standard multiplicity analysis. These gamma rays also effectively extend the range of the neutrons, as they are born at the point of neutron capture at relatively high characteristic energies (e.g., 2.2 MeV in the case of capture on hydrogen) with higher likelihoods of escaping the moderator. Recent experiments using the Beryllium-Reflected Plutonium (BeRP) ball (a 4.5 kg sphere of plutonium) shielded by high-density polyethylene (HDPE) indicate that time-tagged gamma-ray counts in the 2.2-MeV hydrogen capture peak can be used to show that the source is multiplying. Though estimated multiplications underpredict the expected values, the correct trend (as a function of HDPE thickness) was observed. In this paper, we present measurement results and show a preliminary multiplicity analysis to generate Feynman-Y plots. We also discuss the calculated source multiplicity values, the systematic error that appears in this calculation compared to the expected values, and ideas for future studies to improve source characterization using neutron capture gamma rays.

Introduction

We present measured results in this paper validating the concept of neutron capture gamma-ray multiplicity (NCGM) analysis. A neutron emitting source that is moderated with a hydrogen containing material will create gamma rays from the capture of neutrons on hydrogen in the moderator. Prior theoretical and simulation work has shown that these gamma rays are a direct surrogate for captured neutrons lost in the moderator [1]. For each neutron captured by hydrogen, a single 2.2 MeV gamma ray is emitted. At this energy, the probability of a hydrogen capture gamma ray leaking out of a low-Z moderator is high, which provides a signal to either replace or augment the neutron leakage signal.

[1] This work was supported by the Laboratory Directed Research and Development program at Sandia National Laboratories, a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. Approved for unlimited release.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Figure 1 depicts a cartoon of plutonium enclosed in high-density polyethylene (HDPE) to demonstrate the effect of moderation and production of capture gamma rays. Figure 2 compares the neutron leakage to the emission rate of unscattered 2.2-MeV gamma rays, created from neutron capture on hydrogen, in HDPE surrounding the BeRP ball. In this plot, the capture gamma-ray emission rate overtakes neutron leakage at an HDPE thickness of approximately 4” and subsequently falls off at a slower rate as the thickness of HDPE is increased.

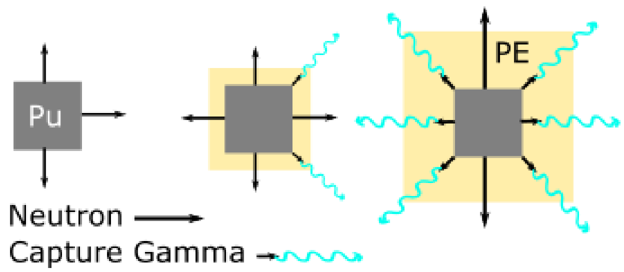

Figure 1. A cartoon depicting a plutonium source surrounded by an increasing amount of moderator. As the polyethylene (PE) surrounding the plutonium (Pu) grows, the neutron leakage increases due to rising neutron multiplication but eventually diminishes as neutrons are absorbed to emit characteristic gammas. These capture gamma rays can act as surrogates for captured neutrons.

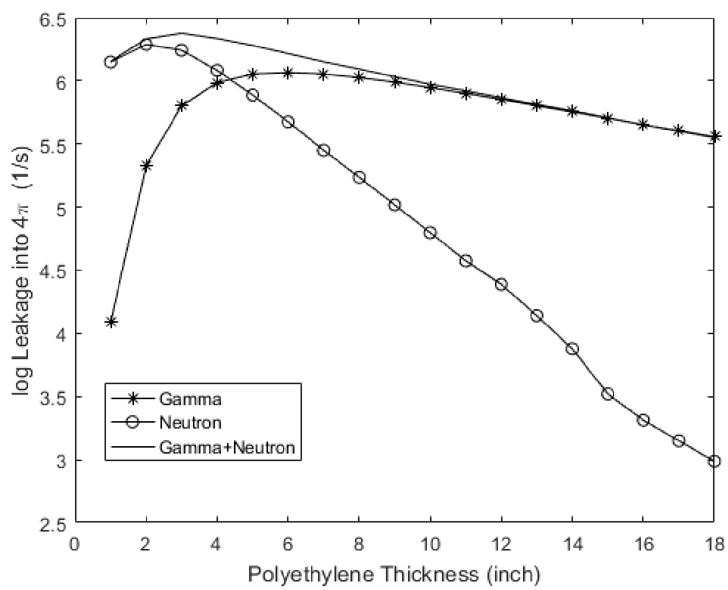

Figure 2. The total neutron, uncollided hydrogen capture gamma ray, and summed leakage into 4π of the BeRP ball, enveloped in HDPE shells as a function of shell thickness. As the amount of HDPE increases, the neutron leakage is originally the dominant leakage signal, but as the amount increases, the uncollided 2.2 MeV capture gamma rays become the dominate leakage signal.

In a multiplying source such as plutonium, the hydrogen capture gamma rays contain the timing information necessary to perform multiplicity analysis using the same techniques that are used for neutron multiplicity counting. Neutron multiplicity counting is a well-established technique that can be used to estimate both the mass and multiplication of special nuclear material (SNM). The NCGM method, used as a stand-alone analysis method or in conjunction with neutron multiplicity counting could offer advances in any application requiring the characterization of moderated SNM.

To validate the viability of the neutron capture gamma signature for multiplicity analysis, a series of measurements were performed using the Beryllium Reflected Plutonium (BeRP) ball, a 4.5 kg sphere of α-phase weapons-grade plutonium, enveloped in varying thicknesses of HDPE shells [2]. The measurement system used for the experiments included seven NaI(Tl) detectors, which recorded the energy and timing of detected events. Using this gamma-ray list-mode data, we selected events within a region of interest surrounding the 2.2 MeV hydrogen capture gamma peak and used the time tags associated with these events to perform neutron multiplicity analysis. Standard moments analysis was performed to assess the multiplication of the objects. The measurements showed that the 2.2 MeV gamma-ray signal was indicative of a multiplying source; however, estimates of object multiplication fell below the expected values. In this paper we will discuss the methods used to perform the measurements - including details of the measurements system, the source and polyethylene moderators used, the measured results and analysis, our conclusions, and our ideas for follow-on studies to improve characterization accuracy.

Experimental Methods

Experimental setup

Our measurement system consisted of eight NaI(Tl) detectors with 4×4×4 in.^{3} crystals coupled to Burle S83049 photomultiplier tubes. They were arranged in two banks of 2×2 arrays, which are depicted in Figure 3. Each bank of detectors was positioned so that the front of each array was located 50 cm radially from the center of the BeRP ball. In each array the detectors were spaced 17.8 cm center-to-center and with the center line of the array 17.8 cm above the surface of the cart they were set on. During the measurements, data was acquired using only seven of the NaI(Tl) detectors as the eighth channel on the digitizer was re-purposed to acquire time synchronized neutron data from an MC-15 multiplicity counter [3] for future analysis. To reduce the contribution of low energy counts, such as the 59.5 keV gamma ray from Am-241, each array was shielded with 0.48 cm of lead backed by 0.16 cm of tin.

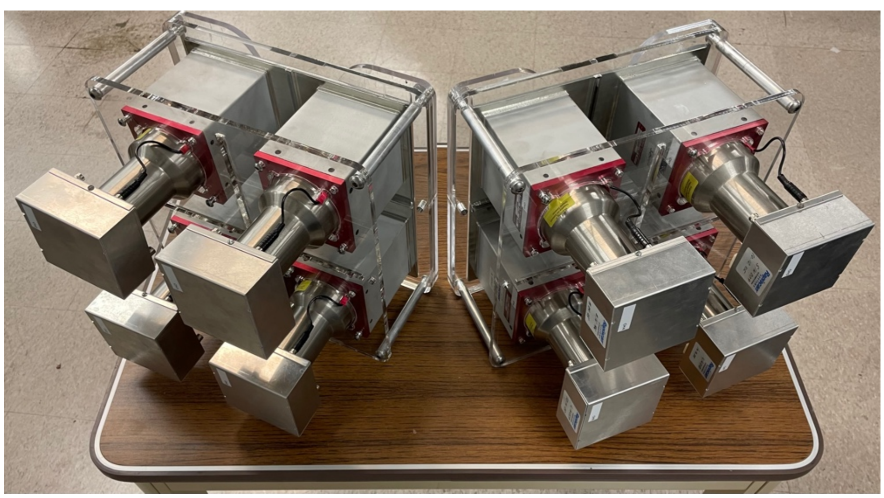

Figure 3. Photo of NaI(Tl) detector arrays used in this work. Each array consists of four 4×4×4 in.^{3} crystals coupled to Burle S83049 photomultiplier tubes. Detectors are shielded in front with 0.48 cm of lead backed by 0.16 cm of tin to reduce the contribution of low energy counts.

The BeRP ball was enclosed in polyethylene shells that were configured to create five different thicknesses of 2”, 3”, 4”, 5”, and 6”. These thicknesses were achieved through use of a set of nested shells each 0.5” in thickness. The source configurations were set on a separate cart (with the same height as the detector cart) adjacent to the detectors. In this measurement configuration the height of the source centerline would increase relative to the detector array as the thickness of the HDPE shells increased.

The detectors were readout by an eight-channel, 14-bit, 250 MHz, CAEN DT5725S digitizer. To improve data acquisition throughput rates, the CAEN Digital Pulse Processing – Pulse Height Analysis (DPP-PHA) firmware was used for on-board waveform analysis [4]. DPP-PHA reduces detected waveforms to a small set of list-mode parameters including timestamp and energy, which is calculated using a trapezoidal filter. Energy information was recorded across 4096 channels using a dynamic range of approximately 11.5 MeV.

Data Analysis

First, each detector was self-calibrated using the measured data. Calibration was performed with InterSpec software [5] using the 2.2 MeV peak from neutron hydrogen capture along with other high-energy lines present in the spectrum. These lines included the 4.4 MeV peak from Be(α, n) reactions originating from an Am-Be source located in the room or the 7.6 MeV lines from Fe(n,g) reactions. The resulting linear calibration coefficients were used to convert from the 4096-channel digitizer output to energy. The spectra for each detector were summed and the 2.2 MeV peak was fit in InterSpec to determine a region of interest (ROI) for event analysis. This window was approximately 2080 keV through 2390 keV, with small variations in ROI edges between HDPE thicknesses.

For all events located in the ROI, a Feynman-Y distribution was calculated to determine if the 2.2 MeV capture gamma signature was indicative of a multiplying source [6]. This distribution computes the value Y_m for increasingly long gate widths. The value Y_m characterizes the multiplication of a system by computing the deviation from a Poisson distribution (the expected distribution from a non-multiplying source) and is calculated as:

$$Y_m = \frac{\overline{C^2}}{\overline{C}} - \overline{C} - 1 \quad . \qquad \text{Eq. 1}$$

In Eq. 1, $\overline{C}$ and $\overline{C^2}$ are the first and second moments of the Feynman histogram, which records the number of gates in which *n* events are recorded for each time gate analyzed. In our analysis, sequential gating was used; the shortest time gate was 10 μs and increased with a step size of 10 μs to a maximum gate length of 5 ms.

To assess how well NCGM analysis can characterize a multiplying object, the list-mode data was also analyzed using Momentum [7], a software package developed by Los Alamos National Laboratory that is used for neutron multiplicity analysis with the MC-15. This analysis was facilitated by converting the list-mode data from the 2.2 MeV ROI into the file format generated by the MC-15, which allowed for the capture gamma data to be directly analyzed using Momentum. Using Momentum, it is possible to estimate the mass and multiplication of an unknown system if the measurement-specific efficiency of the detection system is known (e.g., through use of a well characterized detector). The detection system used for these measurements had not been sufficiently characterized, so we instead used the known mass of the BeRP ball to estimate both the system multiplication and the efficiency of the measurement. In this way we are able to determine how well NCGM analysis could be used to simultaneously estimate mass and multiplication using a well characterized detection system.

Results and Discussion

Gamma Ray Energy Spectra

The measured spectra for all HDPE thicknesses are shown in Figure 4. These spectra were measured between 30 and 60 minutes (depending on the configuration) but are scaled to 10 minutes in Figure 4, which corresponds to the total time used for the multiplicity analysis presented below. All five spectra show a clear peak at 2.2 MeV, caused by the gamma rays produced by hydrogen neutron capture. This peak resides on top of a large continuum consisting of fission gamma rays and Compton scattering events from higher energy fission and capture gamma rays. It is important to note that because multiplicity analysis is performed on an event-by-event basis, it is not possible to accept only the true 2.2 MeV photopeak counts. As such, all events in the ROI, including those below the continuum, are analyzed in this work though we acknowledge that future development may allow for improved event selection. The peak counts and total counts in the 2.2 MeV peak region are quantified in Table 1. As the thickness of the HDPE increases, the ratio of photopeak to total counts in the ROI also increases due to increased hydrogen capture.

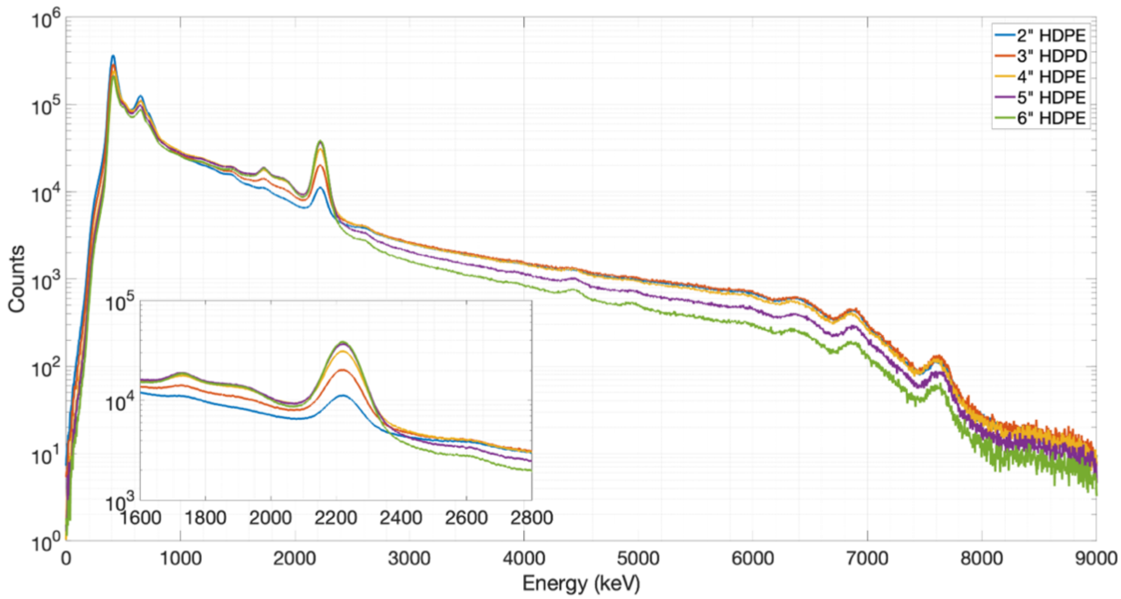

Figure 4. Gamma-ray energy spectra for various polyethylene thicknesses. Spectra are summed across all seven channels and are scaled for a 600 second measurement. Inset zooms on the 2.2 MeV photopeak originating from neutron capture on hydrogen.

Table 1. Count rates summed over seven NaI(Tl) detectors for the 2.2 MeV photopeak and the ROI defined by the photopeak bounds.

HDPE Thickness	2”	3”	4”	5”	6”
Photopeak Area (s^{-1})	362	881	1501	1874	2031
Total ROI Area (s^{-1})	1363	2042	2836	3131	3198
Peak to Total Ratio	0.27	0.43	0.53	0.60	0.64

The large contribution of counts beneath the 2.2 MeV region is notable for the possible effect it has on calculating source mass and multiplicity. The gamma rays emitted by the BeRP ball in this configuration will come from several sources. In addition to capture gamma rays, gamma rays created through fission will also be present. Like the neutron capture gamma rays, fission gamma rays contain timing information that can be used for multiplicity analysis. There are also x-rays and gamma-rays created through radioactive decay, both of which are uncorrelated to fission events and thus cannot be used in a multiplicity analysis.

In the 2.2 MeV ROI, nearly all counts will have derived from either fission gamma rays, capture gamma rays (including higher energy events that have not deposited full energy in the detector), or environmental background (which is a relatively insignificant contribution in the configurations presented here). The decay emission from the BeRP ball will factor very minimally into this ROI because the energy of almost all decay emissions are below 1 MeV. As such, almost all of the gamma-ray events in the 2.2 MeV energy region contain timing information that is correlated to fission chains. However, it is important to note that the fission gammas are not the focus of this effort and how their presence in the 2.2 MeV ROI impacts NCGM analysis is currently under investigation.

Feynman-Y Results

The Feynman-Y distributions for all source configurations are plotted in Figure 5. A 10-minute subset of the acquired data was used in all cases. These distributions all show clear indication of a multiplying source. As the gate size used for analysis is increased, the excess increase until it plateaus with long gate sizes. Based on simulated multiplicities using the GADRAS software [8], the multiplication of the source term should be the lowest for 2” of HPDE and increase with thickness. While the expected multiplication is largest for the 6” of HDPE case, the expected multiplication for 5” of HDPE is the same. The simulated multiplications expected for each source configuration are given in Table 2. The distributions in Figure 5, follow the same trend as the expected multiplication for each configuration. The Y_m value, at which the 2” case plateaus, is the lowest and this value increases for increasing amount of HDPE. As the amount of HDPE is increased, however, the expected multiplication and the Y_m plateau values increases by a decreasing amount. The measured Feynman-Y distributions for the 5” and 6” cases appear similar, which is consistent with the results predicted by GADRAS.

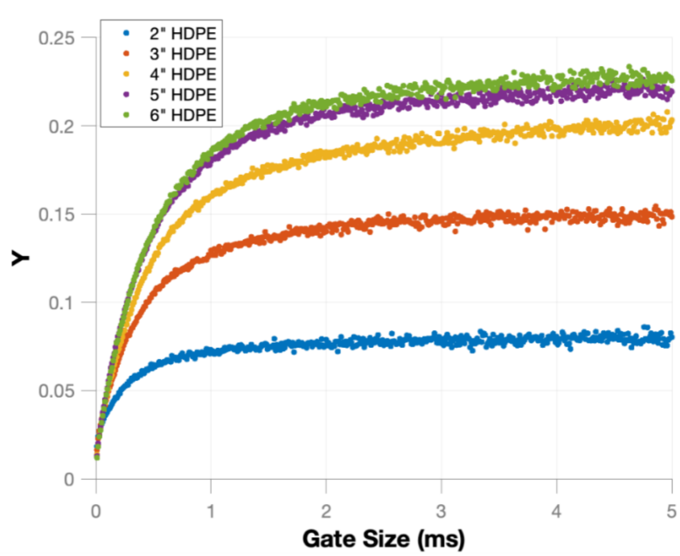

Figure 5. Feynman-Y distribution generated using gamma-rays recorded with energies between approximately 2080 keV and 2390 keV for the BeRP ball moderated by various thicknesses of HDPE.

Object Characterization

Table 2 summarizes the results of the object characterization performed using Momentum. As discussed in the Experimental Methods section, the detector efficiency parameter was adjusted until the known BeRP ball mass (4484 g) was achieved. The associated estimated multiplication and the efficiency used to obtain these values was recorded. To assess performance, the estimated multiplication is compared to that reported by GADRAS simulations of the five measured configurations, which are also shown in Table 2 along with the ratio between the NCGM estimate and the GADRAS estimate of multiplication.

For all configurations, the NCGM estimated multiplication underpredicts the multiplication reported by GADRAS. However, the estimated values follow the appropriate trend, with the NCGM estimates constantly low by a factor of approximately 0.53. The underprediction is inconsistent with results obtained from prior simulation studies, which demonstrated reasonable agreement with both the expected mass and multiplication of the object. The reason for this underprediction is currently unknown and is under ongoing investigation. An underprediction in multiplication is often indicative of an additional uncorrelated signal, which implies that an uncorrelated 2.2 MeV gamma signal is being included in the ROI.

Table 2. Estimated multiplication obtained through analysis of NCGM data in Momentum. Estimates were obtained by adjusting detector efficiency until the known mass of the BeRP ball was achieved. The estimated multiplication is compared against expected values obtained from GADRAS simulations.

HDPE Thickness	2”	3”	4”	5”	6”
Efficiency	9.06×10^{-4}	1.32×10^{-3}	1.58×10^{-3}	1.73×10^{-3}	1.75×10^{-3}
Mass (g)	4484	4484	4484	4484	4484
Calculated Total Multiplication (Measurement)	7.1	8.1	8.5	8.6	8.8
Expected Multiplication (GADRAS Simulation)	12.5	15.4	16.2	16.4	16.4
Ratio of Measured Multiplication to Expected Simulated Multiplication	0.57	0.53	0.53	0.53	0.54

Further analysis is required to understand if this is an impact of the fission gamma continuum or an external factor. One possible source of uncorrelated 2.2 MeV gamma rays is the presence of an AmBe source within an HDPE container that was used for occasional calibration runs. Though this source was moved away from the measurement system when not in use, the 4.4 MeV Be(α, n) peak is present in the spectra, which suggests some 2.2 MeV gammas from the AmBe container are also likely being detected. It is worth noting that the simulated multiplications may also be slight overpredictions due to details not fully modeled, such as deformations in the HDPE shells that cause small air gaps. These gaps would result in a smaller multiplication than the perfect HDPE shell used for simulation. The configurations were also measured using a pair of MC-15 detectors; however, this data has not yet been fully analyzed. Once available, the measured multiplication will be used to benchmark the performance of the NCGM method rather than the simulated data.

Conclusions and Future Work

This paper reported results demonstrating that capture gamma-rays generated through thermal neutron capture on hydrogen can be used for multiplicity analysis. The Feynman-Y distributions generated for all five source configurations were indicative of a multiplying configuration. However, the estimated multiplication using the 2.2 MeV capture peak was shown to underestimate the expected values by approximately a factor of two. Nonetheless, both the Feynman-Y distributions and the estimated multiplications followed the trends expected from simulated data. These results demonstrate that the capture gamma rays can be used as neutron surrogates to augment neutron multiplicity analysis or even replace it in heavily moderated configurations where neutron leakage is lacking.

Future work will focus on improvement in object characterization accuracy. Measured data will be assessed against simulations of the same configurations to determine any deviations that may lead to the observed underprediction of multiplication. Simulations will also allow us to better explore the impact of the non-capture gamma continuum in the ROI. It may be possible to improve event selection using interevent timing information between gammas and/or between gammas and neutrons. List mode data recorded from the MC-15 on the eighth channel of our digitizer will allow us to validate any future improvements reliant on gamma/neutron correlations.

Acknowledgements

These measurements were conducted at the National Critical Experiments Research Center. The National Critical Experiments Research Center is funded by the DOE Nuclear Criticality Safety Program, funded and managed by the National Nuclear Security Administration for the Department of Energy. The National Critical Experiments Research Center is operated by Los Alamos National Laboratory. Los Alamos National Laboratory is operated by Triad National Security, LLC, for the National Nuclear Security Administration of the US Department of Energy under Contract No. 89233218CNA000001.

References

[1] S. O'Brien and M. Hamel, "Characterizing Shielded Special Nuclear Material by Neutron Capture Gamma-Ray Multiplicity Counting," Sandia National Laboratories, SAND-2020-10610, 2020. doi:10.2172/1673454.

[2] J. Mattingly, "Polyethylene-Reflected Plutonium Metal Sphere: Subcritical Neutron and Gamma Measurements," Sandia National Laboratories, SAND2009-5804, 2009. doi:10.2172/974870.

[3] C. E. Moss, M. A. Nelson, R. B. Rothrock and E. B. Sorensen, "MC-15 Users Manual," Los Alamos National Laboratory, LA-UR-18-29563, 2018. doi:10.2172/1477606.

[4] CAEN, *User Manual UM5960 CoMPASS Rev. 18,* UM5960, 2022. [Online]. Available: https://www.caen.it/products/compass/

[5] W. Johnson, "InterSpec - Spectral Radiation Analysis Software," Sandia National Laboratories, [Online]. Available: https://github.com/sandialabs/InterSpec.

[6] R. P. Feynman, F. de Hoffman and R. Serber, "Dispersion of he Neutron Emission in U235 Fission," US Atomic Energy Commission, 1955. doi:10.1016/0891-3919(56)90042-0.

[7] C. E. Moss, J. D. Hutchinson and M. A. Nelson, "Momentum Users Guide," Los Alamos National Laboratory, LA-UR-18-31433, 2018. doi:10.2172/1485367.

[8] S. M. Horne, G. G. Thoreson, L. A. Theisen, D. J. Mitchell, L. Harding and S. O'Brien, "GADRAS Version 18 User's Manual," Sandia National Laboratories, SAND-2019-14655, 2018. doi:10.2172/1592910.