Time-Correlated-Pulse-Height Technique Measurements of Fissile Samples at the Device Assembly Facility

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Abstract

Detecting and quantifying special nuclear material via gamma spectroscopy techniques becomes exceedingly difficult in the presence of shielding and background radiation. Gamma particles are easily attenuated in high density materials, whereas the long half-lives of special nuclear material may preclude adequate signal to noise ratios. Neutrons exhibit higher penetrability over gamma rays, yet their presence does not guarantee the existence of special nuclear material. Many radioactive sources emit temporally correlated gammas and neutrons, but only for fissile and fissionable sources can these correlations extend over several reactions. Thus especially the detection of multiplying fission sources garners much attention. Whereas in non-multiplying fission sources, such as ^{252}Cf, correlated gamma-neutron-pairs arise from just one spontaneous fission event, multiplying fission sources would allow for a gamma to be temporally correlated with any neutron from a subsequent fission in a fission chain. Organic scintillation detectors exhibit the required properties for such measurements. These include their nanosecond-scale timing properties and their aptitude for pulse shape discrimination. A measurement system consisting of four 3 in by 3 in EJ-309 liquid scintillation detectors was used to measure time-correlated gamma-neutron-pairs from the BeRP ball, a 4.5 kg plutonium sphere at the Device Assembly Facility at the Nevada Test Site. This sphere was measured with polyethylene or tungsten reflectors of various thicknesses to attain a variety of multiplications. For a gamma-neutron-pair detected in separate detectors, the time difference between the triggering gamma and the subsequently detected neutron as well as its measured pulse height were recorded. A surface plot of this experimental data was superimposed on curves representing the neutron energy limit for a fixed time of flight assuming no multiplication. Both the experimental data and MCNPX-PoliMi simulations showed that the number of events beyond these curves were indicative of the source's multiplication, whereas a non-multiplying source would show zero events beyond these curves.

Introduction

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The sunset of 20th century threats has prompted the U.S. to scale down its nuclear weapons program which has been a chief supplier of ^{3}He. However, the emergence of new 21st century threats has only increased the demand for the rare isotope because of the deployment of neutron detectors at the U.S. border and ports following the attacks of September 11, 2001. For nearly a decade nuclear scientists have looked for viable alternative neutron detectors to ^{3}He tubes. Liquid organic scintillators, such as the EJ-309, are an attractive choice and do to their fast timing and neutron-gamma discrimination properties they lend themselves to measurement techniques not possible with ^{3}He tubes.

One of such techniques involves the analysis time correlated pulse heights (TCPH) between multiple detectors. Analysis of the TCPH surface plots can yield valuable information about the multiplication of the material of interest and this in turn can provide information on the relative threat of the source. This technique has previously been deployed to analyze low multiplication samples of MOX powder and PuGe disks [1] and simulation of Highly Enriched Uranium (HEU) and Pu spheres with polyethylene shells shielding [1][2]. In this work, this analysis was extended to measurements of BeRP Ball, an alpha-phase 4.48 kg Pu sphere, in bare and shielded configurations. The measurements were taken as part of a collaboration effort between Sandia National Laboratories and the University of Michigan. Simulations of the experimental set up with supplemental shielding configurations were performed using MCNPX-PoliMi [3] and MPPost [4]. Four 7.62 cm by 7.62 cm EJ-309 liquid scintillation detectors were used is this study located 50 cm from the center of the source to the detector face. The detectors were placed adjacent to each other, approximately 18 degrees apart. Three particular configurations were investigated: bare BeRP ball, shielded with 1 in of tungsten and shielded with 3 in of polyethylene.

Time Pulse-Height Correlations

With two or more liquid organic scintillators it is possible to determine the correlations between neutron-neutron (n-n), photon-neutron (p-n), and photon-photon (p-p) events if pulse shape discrimination (PSD) is employed to distinguish between the two types of radiation. If the p-n correlations are used to determine timing information by triggering on the photon interaction and pulse height from the neutron interaction then TCPH can be established for the source. A distribution of various pulse heights on the ordinate and corresponding arrival times on the abscissa can be represented on a surface plot and be analyzed for source's multiplication.

The theoretical time of arrival of the neutron can be determined from its energy:

$$t = \frac{d}{\sqrt{\frac{2E_n}{M_n}}} - \frac{d}{c} \tag{1}$$

where d, E_n, and M_n are the source to detector distance, energy of the neutron and its mass. The d/c term is used to compensate for the arrival time of the photon. For a non-multiplying source, such as ^{252}Cf, the measured time from the p-n correlation should not exceed the theoretical arrival time calculated from neutron pulse height. However, with a multiplying source it is possible to correlate photons from the original fission with neutrons born in later generations of the fission chain. This time delay would be represented on a TCPH plot since those neutrons would arrive at later times than the theoretical time calculated from their energy would indicate.

A theoretical line of the time of arrival can be drawn using Eq. (1) on a TCPH surface plot and for a non-multiplying source neutrons should fall below that line. This is due to the energy loss in interactions that the neutron experiences before interacting in the detector. For a multiplying source there should be a spread of neutrons that arrives over the theoretical line, and in theory the magnitude of the spread should indicate source's multiplication. The simplest analysis of taking a ratio between the neutrons counted above and below this theoretical line would result in a number that should correspond to source's multiplication. Multiplication of the source can be determined by a Monte Carlo k-calculation of the source configuration:

$$M = \frac{1}{(1 - k_{eff})} \ . \tag{2}$$

Simulated analysis of this ratio and source multiplication has been performed in the past and shown logarithmic behavior between the two variables [1][2].

Fission Chain Lengths

In principle source's multiplication is related to the length of fission chains measured by the generation number of the last neutron in the fission chain. As a preliminary study a bare, tungsten, and polyethylene shielded configurations were compared with respects to fission chain lengths. The configurations consisted of spherical shells of the shielding material that completely surrounded the BeRP ball. These configurations were simulated in MCNPX-PoliMi which provides a collision printout that among other things gives the particle type and its generation number [4].

The results of the simulations and subsequent post processing that tallied the last generation number of neutrons in a histogram are shown in Figure 1. As expected the bare case had the shortest fission chains and longest fission chains are present in the polyethylene shielded configurations due to its superior neutron reflection properties. However, with varying thicknesses the distribution of fission chain lengths between 2 inches of tungsten and 1 inch of polyethylene become closely aligned. This underscores the difficulty of pinpointing the shielding configuration if multiple materials are to be considered. In this case if a fissile or fissionable

source was suspected, the presence of tungsten or other high Z material could be deduced from the lower than expected level gamma radiation. This analysis would be possible due to the PSD employed with liquid organic scintillators.

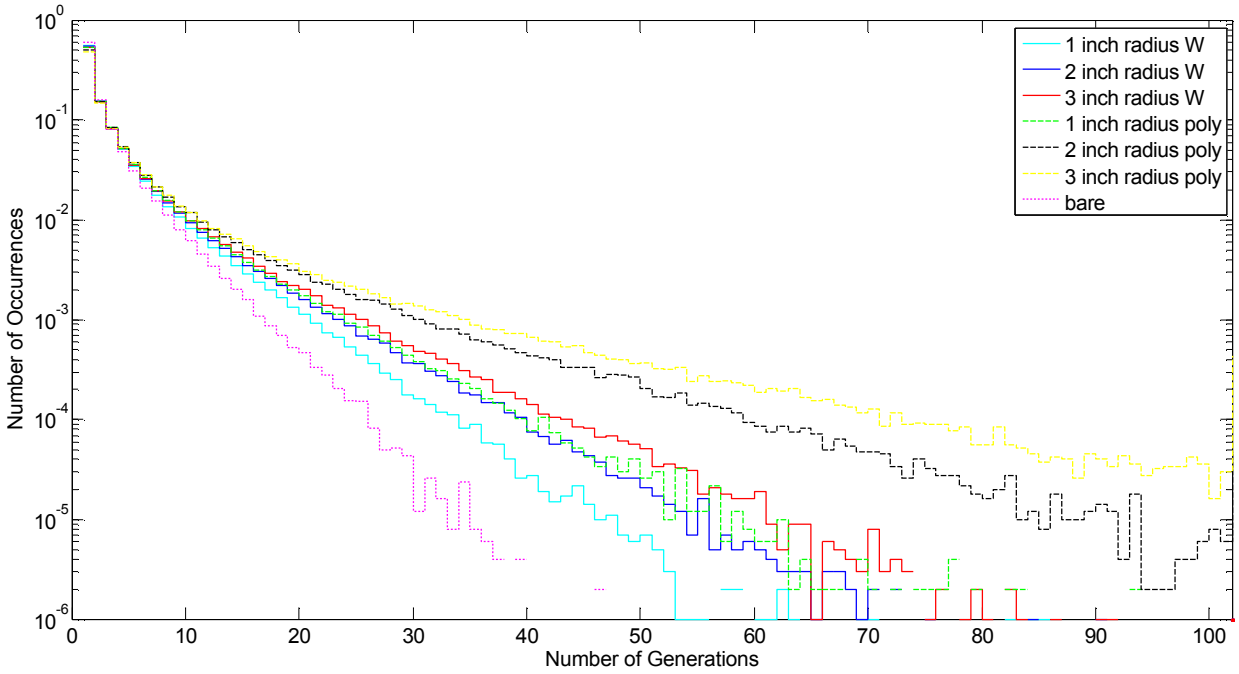

Figure 1. Fission chain lengths in different shielding configurations of the BeRP ball.

Energy Calibration

Two calibration runs were used to determine the conversion between output pulse height and energy in MeVee. The ^{137}Cs calibration source was placed 50 cm away from the four detectors and the total measurement time was 17 minutes. The results of the pulse height distributions from each of the four detectors are shown in Figure 2. Linear interpolation was performed on the Compton edge and it was assumed that its true position was located 20% from the top of the edge. Pulse heights of 0 volts were assumed to correspond to 0 MeVee. The voltage that corresponded to the 478 keVee Compton edge was average over the four detectors and calculated to 0.5147 V. This was used to convert between pulse height and energy for all four detectors. Ideally all detectors would be calibrated to have Compton edge located around the same point on the pulse height distribution. It is clear that for reasons unknown, detector four corresponding to channel 3 exhibited lower pulse heights for the same energy deposition as the other three detectors. Because a single conversion was used and it was determined from the average of the four detectors, the energy deposition in detector four could be underestimated creating an unintended shift in the TCPH plots.

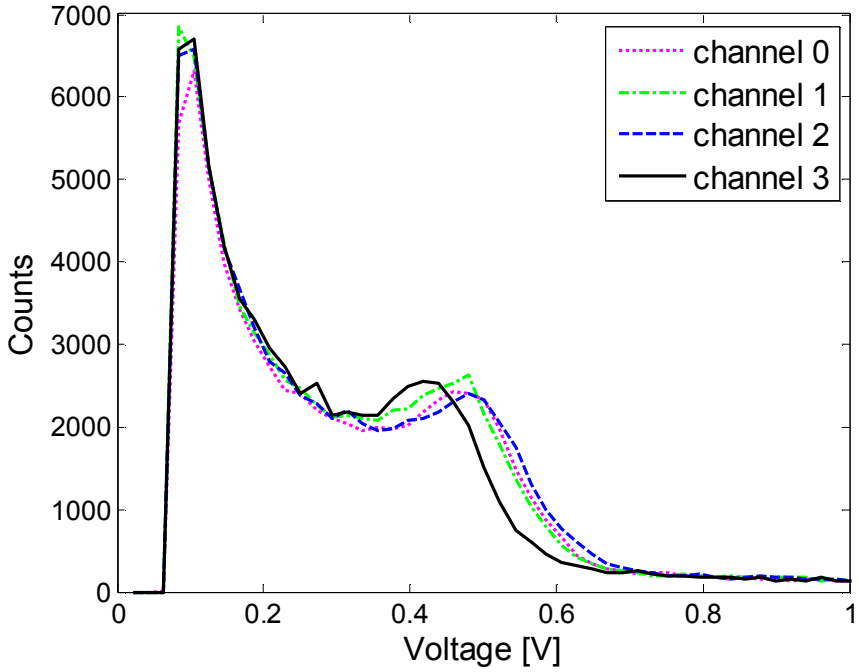

Figure 2. The pulse height distributions from ^{137}Cs calibration of the each of the 4 detectors.

Pulse Shape Discrimination

The digitized pulse data provided by Sandia National Laboratories was processed with U-M developed PSD algorithms. To determine the optimal PSD line for each of the four detectors the data from the first run of the 1 inch tungsten shielded configuration was used. The tungsten configuration was useful because it shielded the heavy flux of photons and provided data with an equitable distribution of photons and neutrons. The data was provided in a series of waveforms each 100 points in length representing 4 ns in time and the values of which represented the height of the waveform in digitizer units. The vertical resolution of the digitizer was 12 bits and the dynamic range was 2 V.

The pulses were cleaned by eliminating pulses that went beyond the dynamic range or were below the set threshold of 0.03 V. The tails were integrated starting from 20 ns after the pulse maximum up to 220 ns. A surface plot of tail and total integrals provides visual means of determining the cutoff line for photons or neutrons, as shown in Figure 3. This line was visually optimized to best discriminate between pulses at low energies and was fit to a quadratic function of the form:

$$y = Ax^2 + Bx + C \tag{3}$$

where y and x are the tail and total integral, respectively. If the calculated tail integral is smaller than the actual tail integral than the pulse is deemed to be a neutron, otherwise it is classified as a

photon. The coefficients A, B and C determined for each detector and used for processing the rest of the data are shown in Table 1.

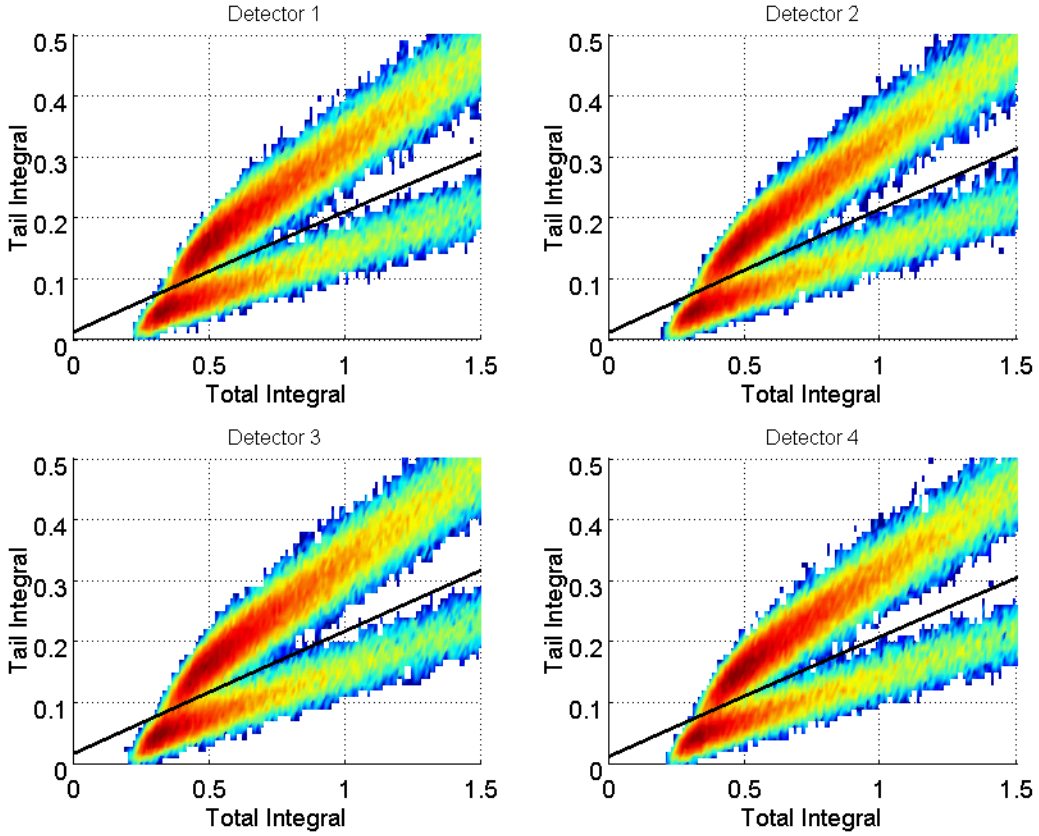

Figure 3. Surface plot of the tail and total integrals from the first run of the 1 inch tungsten shielded case.

Table 1. The PSD coefficients determined from the 1 inch tungsten case.

Coefficients	Detector 1	Detector 2	Detector 3	Detector 4
A	-0.0044066	-0.0043383	-0.0015272	-0.0031422
B	0.20141	0.2076	0.20171	0.19959
C	0.012242	0.011231	0.017326	0.012741

TCPH Results

Three cases of BeRP ball configuration were considered: bare, 1 in of tungsten shielding and 3 in of polyethylene shielding. The measurement times were 12 minutes, 150 minutes and 42 minutes, respectively. The threshold on the digitizer during measurement was set to approximately 0.08 MeVee, when converted to light output with the conversion ratio calculated from the energy calibration. Four 7.62 cm by 7.62 cm EJ-309 liquid scintillation detectors were

used is each measurement located 50 cm from the center of the source to the detector face. The detectors were placed adjacent to each other, approximately 18 degrees apart. All three setups were simulated with MCNPX-PoliMi and the results were post-processed with MPPost in order to generate the cross correlations between each detector pair necessary for the construction of TCPH distributions. The geometry of the experimental set up is shown in Figure 4. The 50 cm deep concrete disk located below this geometry is not shown. The purpose of the concrete was to simulate room scattered in a real measurement. A stainless steel shell that encased the Pu metal was also modeled and measured as part of the bare case.

The same post processing was performed for both the measured and simulated cases in order to preserve relevance in comparison. The background was subtracted by averaging the counts in the negative time domain between -80 and -20 ns. This procedure in effect eliminated most of the counts above 100 ns for both simulated and measured TCPH distributions. A new threshold of 0.2 MeVee was imposed in order to eliminate a low energy tail that persisted for longer arrival times and distorted the ratio above and below the discrimination line. The theoretical time of arrival also included the mean free path of the neutron in each of the detectors; this was done to further refine the analysis since most neutrons will not interact directly on the face of the detector. A relatively significant island of misclassified neutrons was present around 0 ns. In order to eliminate their effect the distribution of neutrons between 0 ns and 6 ns was not considered.

The resulting TCPH distributions for the bare case are shown in Figure 5. There appear to be more data points in the simulated result and that may be due to the longer simulation time compared with the measurement time. However, the ratios of the distributions above and below the discrimination line agree very well as shown in Table 2. Unfortunately, by raising the threshold the absolute value of this ratio also increased, as compared with previous work of similar bare configurations [1]. This is significant because as discussed before with additional shielding the relationship between multiplication and the ratio takes on a logarithmic behavior. Thus by raising the threshold some of the certainty in discriminating between various source multiplication levels may be lost. The tungsten TCPH results are shown in Figure 6. For this case there are more data points in the measured distribution and this effect could be attributed to the longer measurement time as compared with simulation time. The discrepancy between the measured and simulated ratios was 3% as shown in Table 2. As expected the absolute value of the ratio increased as more neutrons from later generations were correlated in time with photons of previous generations. The comparison between the measured and simulated 3 in polyethylene configuration is shown in Figure 7. The discrepancy between the ratios above and below the line is nearly 20%. The longer simulation time may be the culprit, however unlikely, since the data points apparent in the simulated and missing from the measured TCPH distribution above the discrimination line are a few orders of magnitude lower than the majority distributed neutrons.

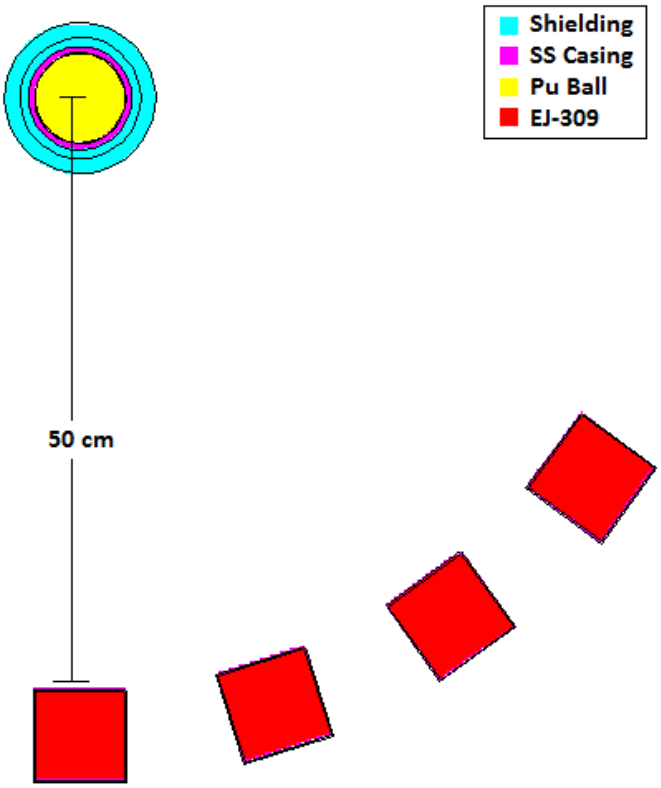

Figure 4. Geometry of the experimental set up in MCNPX-PoliMI,

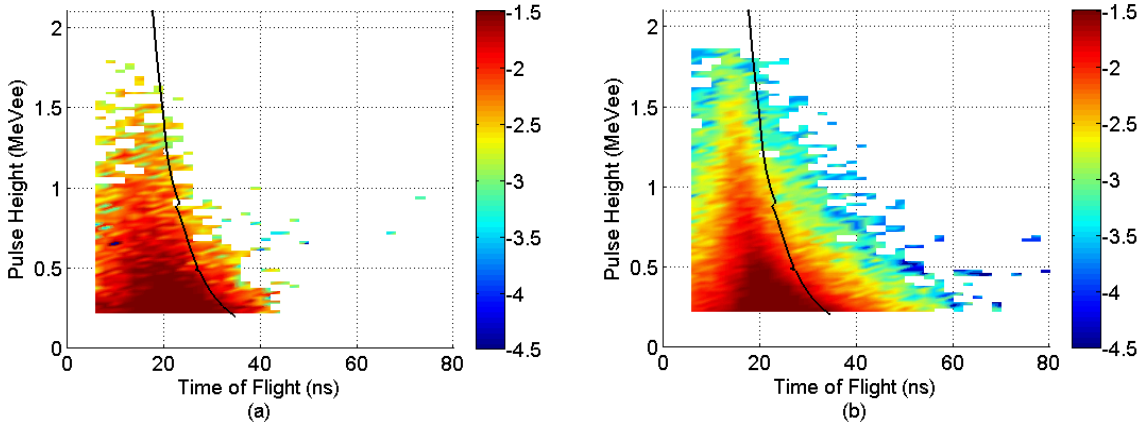

Figure 5. TCPH for bare BeRP ball: a) measured b) simulated.

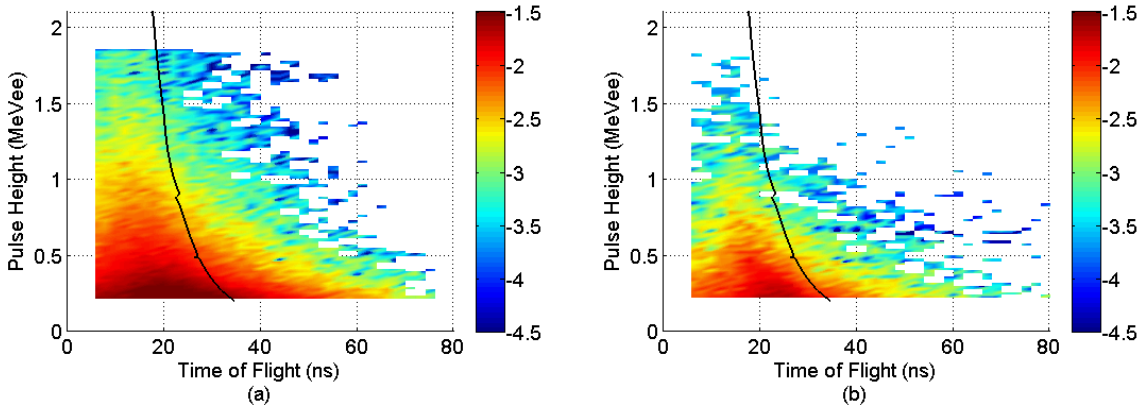

Figure 6. TCPH for 1 in tungsten shielded BeRP ball: a) measured b) simulated.

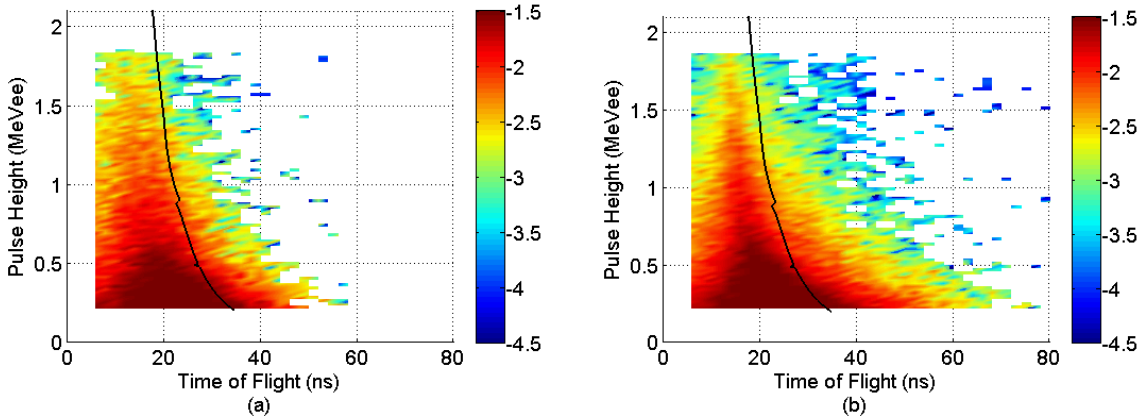

Figure 7. TCPH for 3 in polyethylene shielded BeRP ball: a) measured b) simulated.

Table 2. Comparison of the ratios calculated from TCPH neutrons above and below the theoretical line of arrival for measurement and simulation.

Configuration	Measurement	Simulation	Multiplication
Bare	0.468 +/- 0.01056	0.469 +/- 0.00478	4.44
1 in Tungsten	0.664 +/- 0.00489	0.644 +/- 0.01051	8.94
3 in Polyethylene	0.541 +/- 0.00612	0.648 +/- 0.00489	16.26

Conclusions

TCPH distributions for base, 1 in tungsten and 3 in polyethylene shielded configurations of the BeRP ball were successfully constructed from measured data and MCNPX-PoliMi simulated results. The ratio of neutron distributions above and below the theoretical time of arrival line was shown to increase with multiplication. However, the behavior was nonlinear between the two types of shielding used. The 3 in polyethylene case has multiplication of 16.26, nearly double of the 1 in tungsten case, but the simulated ratio is only 0.008 higher than the 1 in tungsten case. Raising the threshold was necessary to eliminate non-physical features in the measure TCPH but may have impacted the ability to distinguish between low and highly multiplying sources. Changing the threshold also impact the relative difference between simulated and measured

results. To improve the results a more robust way of performing PSD may be of use as well as further examination of the raw data provided by the digitizer. Some issues in the way the digitizer reset the time stamps when printing waveforms may have contributed to the larger differences shown in the 3 in polyethylene case. Finally, matching the simulation and measurement time may further clarify the discrepancies between the two results.

References

[1] E.C. Miller. *Characterization of Fissionable Material using a Time-Correlated Pulse-Height Technique for Liquid Scintillators.* (Doctoral dissertation), 2012.

[2] E.C. Miller, S.D. Clarke, A. Enqvist, S.A. Pozzi, P. Marleau, J.K. Mattingly, "Characterization of Special Nuclear Material using Time-Correlated Pulse-Height Analysis," *Journal of Nuclear Materials Management*, Volume XLI, Number 1, pp. 32-37 Fall 2012.

[3] E. Padovani, S. A. Pozzi, S. D. Clarke, E. C. Miller, "Introduction to MCNPX-PoliMi", Version 2.7, (2012).

[4] E. C. Miller, S. A. Pozzi, E. Padovani, S. D. Clarke, M. Flaska, J. Mattingly, "MPPost Manual," Version 2.7, (2012).