Multiplication and Presence of Shielding Material from Time-Correlated Pulse-Height Measurements of Subcritical Plutonium Assemblies

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Abstract

We present the results from the first measurements of the Time-Correlated Pulse-Height (TCPH) distributions from 4.5 kg sphere of α -phase weapons-grade plutonium metal in five configurations: bare, reflected by 1.27 cm and 2.54 cm of tungsten, and 2.54 cm and 7.62 cm of polyethylene. A new method for characterizing source multiplication and shielding configuration is also demonstrated. The method relies on solving for the underlying fission chain timing distribution that drives the spreading of the measured TCPH distribution. We found that a gamma distribution fits the fission chain timing distribution well and that the fit parameters correlate with both multiplication (rate parameter) and shielding material types (shape parameter). The source-to-detector distance was another free parameter that we were able to optimize, and proved to be the most well constrained parameter. MCNPX-PoliMi simulations were used to complement the measurements and help illustrate trends in these parameters and their relation to multiplication and the amount and type of material coupled to the subcritical assembly.

Keywords: Fission chain, Liquid scintillator, Subcritical assembly, Non-linear Inverse Problem

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1. Introduction

1.1. Motivation and the new approach

The ability to distinguish between multiplying and non-multiplying sources is of great interest to the safeguards and non-proliferation community. Neutron coincidence counting is one of the primary techniques in characterizing special nuclear material (SNM) properties such as presence of oxides, fissile mass, and by extension multiplication [1]. We have focused on cross-correlation measurements with liquid organic scintillators as a possible method for characterizing SNM [2, 3]. The end-goal is a system that is smaller and more portable than traditional thermal neutron capture based multiplicity counters. In addition, the method we propose would make accurate knowledge of system efficiency superfluous for the purposes of characterizing SNM. In this paper, we present a new technique for using inter-event timing data that could be collected by such a system.

The Time-Correlated Pulse-Height (TCPH) distribution from measurements of the Beryllium Reflected Plutonium (BeRP) [4] ball under various reflector configurations is presented in this work. We also present a method for inversely solving parameters that describe the fission chain timing distribution. Our underlying assumption is that for a multiplying source, such as the BeRP ball, the TCPH distribution is smeared by the distribution of times between fission events in a chain. This fission chain timing distribution is shown to be sensitive to reflector composition, which complicates an absolute measurement of multiplication.

A physical model of the fission chain distribution is beyond the scope of this paper. Instead, we used a gamma distribution as a proxy for the time smearing caused by the development of fission chains. This approach has allowed us to draw empirical conclusions between the gamma distribution fit parameters and physical variations in fissile material assemblies, such as multiplication and coupling with a neutron moderating or reflecting material.

We define the difference between moderating and reflecting materials by the relative energy loss of elastically scattering neutrons. Neutrons can scatter off

31 of a high-Z reflector material at large angles without significant energy loss,
 32 and therefore quickly, on the order of nanoseconds, return to the multiplying
 33 medium. By contrast, in low-Z moderators scattering neutrons slow down sig-
 34 nificantly and may return to multiplying medium several microseconds later.

35 The utility of the TCPH distribution has been extensively studied with
 36 non-multiplying ^{252}Cf [5], low-multiplying mixed oxide powder and plutonium-
 37 gallium disks [6], and highly enriched uranium [7] measurements and simulations
 38 [8]. BeRP ball experiments measuring the TCPH as a signature have only thus
 39 far been simulated, and these results demonstrated a correlation between mul-
 40 tiplication and spreading in the TCPH distribution [9]. These earlier efforts
 41 to characterize multiplication relied on counting the correlated events that fell
 42 above the maximum theoretical arrival time of neutrons as calculated by their
 43 deposited energy. In this work, we advance beyond this methodology by re-
 44 constructing the time distribution between fission events and using this new
 45 signature for characterizing multiple aspects of a subcritical assembly.

46 1.2. TCPH distribution and relation to fission chain timing

47 Cross-correlation measurements combined with the pulse shape discrimina-
 48 tion (PSD) methods allow for the classification of neutron-neutron, gamma-
 49 neutron, neutron-gamma, or gamma-gamma events [10, 11]. The TCPH distri-
 50 bution is a bivariate histogram of the time difference between correlated neu-
 51 trons and gamma-rays, and deposited neutron energy measured by the light
 52 output in the scintillator. In a non-multiplying source, such as ^{252}Cf , most
 53 correlated neutrons and gamma-rays detected within a short time window (10s
 54 of nanoseconds) by our fast system are generated from the same fission event.
 55 Therefore, the arrival time of a neutron, which depends on neutron incident
 56 energy, sets an upper limit on the light output achievable for that event.

57 Figure 1(a) shows an example of a TCPH distribution from a hypotheti-
 58 cal non-multiplying mono-energetic source of 1 MeV neutrons and correlated
 59 gamma-rays. In this example, we assume the detector response is linear and all
 60 scattering angles are equally likely. In this case, all neutrons arrive 35 nanosec-

61 onds after the correlated gamma-ray due to the travel time of a 1 MeV neutron
62 across a source-detector distance of 50 cm. In reality the arrival time would be
63 slightly longer because of the extra collision distance within the detector. For
64 the purposes of this example we assume that all interactions take place at the
65 face of this hypothetical detector.

66 For multiplying sources, the presence of fission chains makes it possible for
67 neutrons and gamma-rays to be correlated in different events along the fission
68 chain. The time of arrival is therefore either shorter or longer depending on
69 the order of generation along the fission chain of the correlated gamma-neutron
70 pair. In a multiplying assembly of fissile material this effect manifests itself as
71 a spreading or smearing of the TCPH distribution. An example of the effect of
72 this time smearing caused by source multiplication is shown in Figure 1(b). The
73 spreading in the negative and positive time direction is caused by the possibility
74 of correlating gamma-rays with neutrons from past and future generations of
75 fission events, respectively.

76 Gamma-gamma and neutron-neutron pairs could also be used to create
77 TCPH-like distributions. However, these types of correlated events suffer from
78 significantly more detector cross-talk as the same particle scatters twice in the
79 detection system. Additionally, in neutron-neutron pairs, both particles have
80 a time-of-flight that depends on their respective energies and thus the time
81 between neutrons is spread even for pairs originating from the same fission.
82 This decreases the sensitivity of the TCPH approach to time spread due to the
83 presence of fission chains.

84 By contrast, correlated gamma-rays from the same fission should arrive at
85 the detector simultaneously. Therefore, in a manner similar to that illustrated in
86 Figure 1, any time spread can be attributed to fission chain smearing. However
87 correlated gamma-gamma pairs have many sources of uncorrelated background
88 that degrade the signal-to-noise of this signature. In addition to correlated fis-
89 sion gamma-rays, fissile material typically produces many more uncorrelated
90 decay gamma-rays. Depending on the amount of material present, these uncor-
91 related gamma-rays can create an overwhelming rate of accidental coincidences

Figure 1: Calculated TCPH distributions for (a) non-multiplying and (b) multiplying sources assuming a mono-energetic 1 MeV neutron source at a distance of 50 cm with linear detector response and equal probability of scatter in all angles. Each distribution is normalized to unity and displayed on a logarithmic scale.

92 within a TCPH time window.

93 Gamma-rays are also highly attenuated by the emitting fissile material itself,
 94 so they are primarily sensitive to the outer layers of an assembly. It is therefore
 95 desirable to include at least one neutron in the correlated pair to gain sensitivity

96 to a larger fraction of the total volume of material.

97 We have therefore determined that correlated gamma-neutron and neutron-
98 gamma pairs offer the best sensitivity and signal to noise. The gamma-ray
99 provides a clean indication of the time of a fission, while the neutron is both
100 more penetrating and a clear indication of fission, reducing the rate of accidental
101 coincidences.

102 2. Experiment

103 The BeRP ball is a 4.5 kg sphere of α -phase weapons-grade plutonium metal,
104 originally manufactured in October 1980 by Los Alamos National Laboratory
105 [4]. This sphere has a mean radius of 3.7938 cm, and is encased in a 304 stainless
106 steel shell that is 0.0305 cm thick. In our configurations, two identical hemishells
107 of either polyethylene or tungsten were used that completely covered the BeRP
108 ball. A photograph of the BeRP ball with polyethylene hemishells is shown in
109 Figure 2.

Figure 2: Photograph of the BeRP ball nested in polyethylene hemishells, courtesy of reference [4].

110 A series of measurements of the BeRP ball were conducted at the Nevada
111 National Security Site (NNSS) to acquire data to assess TCPH distribution

analysis for highly multiplying assemblies of fissile material. To explore a range of multiplications with different levels of moderation and reflection, we measured five configurations: bare, 1.27 cm and 2.54 cm thick close fitting shells of tungsten, and 2.54 cm and 7.62 cm thick close fitting shells of high density polyethylene (HDPE). The measurement times for each configuration were 5515, 3600, 8999, 3600, and 2509 seconds, respectively.

Data were collected using four 7.62×7.62 cm cylindrical EJ-309 liquid scintillation detectors. The source-to-detector distances were measured from the center of the BeRP ball. Most configurations were measured at 50 cm distances with the exception of 2.54 cm tungsten and polyethylene measured at 48 and 60 cm, respectively. A diagram of the experimental setup, taken from an MCNPX-PoliMi [12] model, is shown in Figure 3. Anode outputs from 7.62 cm Electron Tubes photomultiplier tubes (PMTs) coupled to each cell were digitized using CAEN DT5720 digitizer, capable of 12-bit (nominal) vertical resolution and 250 MHz sampling rate. All data processing was performed off-line after the measurements, and a 0.2 MeVee (MeV electron-equivalent), equivalent to neutron energy of 1.2 MeV, threshold was applied in post-processing.

3. TCPH model construction

In order to access the fission chain timing distribution, we constructed a model of TCPH distributions for multiplying sources. Our model consists of a linear combination of the expected distribution for gamma-neutron pairs that are correlated by the same fission and those correlated by different fissions within the same chain.

The like-fission distribution is determined by a combination of the detector response to fission spectrum neutrons (Watt) and the time delay expected for the source-to-detector distance. The different-fission distribution includes additional time smearing due to the time difference between any two fissions within a chain. For this “smeared” different-fission distribution the neutron has a detector response and travel time characteristic of its energy, but now a

Figure 3: Diagram of the experimental setup of the BeRP ball surrounded by four EJ-309 detectors. The detectors were spaced approximately 8.5° apart.

141 gamma-ray from a different fission starts the clock for the inter-event time.

142 3.1. Like-fission TCPH distribution

143 The TCPH distribution is a bivariate histogram of an estimate of neutron
 144 energy (by pulse height) and time between correlated neutron and gamma-ray.
 145 This time is dependent on the incident neutron energy and source-to-detector
 146 distance. Therefore, for like-fission events the TCPH distribution should depend
 147 on three factors: source-to-detector distance, source neutron energy spectrum,
 148 and the energy-dependent detector response to neutrons. Under controlled lab
 149 conditions, the source-to-detector distance can be readily measured, but it is
 150 possible to include this as a free parameter as we will later demonstrate.

151 We begin with neutrons with energies sampled from the ^{239}Pu fission Watt
152 spectrum [13]. The pulse height response of the detector is stored in a response
153 matrix that maps an incident neutron energy to a probability density function
154 over pulse heights. The matrix was constructed using an MCNPX-PoliMi sim-
155 ulation to determine the expected true energy deposition. Subsequently, a light
156 output function was used to convert that energy into a pulse height distribution.
157 We used a light output function of the following form:

$$L = aE_p - b(1 - \exp(cE_p)) \quad (1)$$

158 where a , b and c are detector specific constants and E_p is the proton recoil
159 energy in MeV. We used parameter values for EJ-309, specified in reference
160 [14], of 0.817 MeVee/MeV, 2.63 MeVee and $-0.297 \text{ MeVee}^{-1}$ respectively. We
161 found that the small variations in light output functions published in references
162 [14] and [15] had no discernible impact on the resulting TCPH distributions and
163 our fitted parameters. The simulated neutron response matrix and the resulting
164 TCPH distribution are shown in Figure 4.

165 3.2. Fission chain timing distribution

166 In order to include the effect on the TCPH distribution caused by gamma-
167 rays and neutrons being correlated from different fissions in the same chain,
168 we posit that the time spread is driven by the distribution of time difference
169 between all possible pairs of events in a fission chain, as illustrated in Figure
170 5. This distribution will be governed by multiple physical attributes, including
171 sub-critical multiplication which is defined as

$$M = \frac{1}{(1 - k)} \quad (2)$$

172 where k is the ratio of the number of neutrons in any generation to the previous
173 one. Multiplication directly corresponds to the average length of a fission chain.
174 The presence of neutron moderating materials, such as polyethylene, will stretch

Figure 4: Simulated EJ-309 neutron response matrix and the resulting TCPH distribution with assumed ^{239}Pu Watt neutron energy spectrum and source-to-detector distance of 50 cm. Each distribution is normalized to unity and displayed on a logarithmic scale.

175 the time between fission events as the neutrons inducing fission are moderated
 176 and reflected.

177 As stated previously, a detailed physical model of the fission chain process
 178 and how it responds to these factors is beyond the scope of this paper. There

179 has been a lack of demand to efficiently model the actual timing between fission
 180 events in a chain. For reactor analysis the principal aspect of the fission chain
 181 is its average length and time-dependent analysis (point reactor kinetics) is
 182 typically limited to the rise and fall of total neutron population in a system.
 183 As a result an off-the-shelf tool that could efficiently model fission chain timing
 184 dynamics is not readily available. Instead, we introduce a gamma distribution
 185 as an empirical substitute for a physical model.

Figure 5: Time differences between n^{th} -nearest-neighboring fission events in a chain starting with the spontaneous fission (sf), followed by subsequent induced fissions. The correlation of all possible event pairs from a fission chain contribute to the spread in the TCPH distribution from multiplying sources. Since it is possible to correlate a gamma-ray with a neutron from either a previous or future fission event in a chain, the TCPH distribution is smeared in both time directions.

186 The probability density function of a gamma distributed random variable is

$$f(x) = \frac{1}{\Gamma(\alpha)\theta^\alpha} x^{\alpha-1} e^{-x/\theta} \quad (3)$$

187 where α is the shape parameter and θ is the rate parameter. Two properties of
 188 the gamma distribution make it a plausible representation for the distribution of
 189 times between fission events. First, the gamma distribution describes the wait-
 190 ing times until α^{th} Poisson distributed event. Second, the sum of independent
 191 gamma distributions follows a gamma distribution. The independence condition
 192 is not true for events in a fission chain; however, we can empirically demonstrate
 193 the time distribution between fission events roughly follows a gamma distribu-

194 tion by comparing MCNPX-PoliMi simulations. The simulated distribution of
 195 time differences between fission events in a bare BeRP sphere and corresponding
 196 gamma distribution fits are shown in Figure 6. Although the time distribution
 197 between exact number of fission events follow a gamma distribution, with co-
 198 efficient of determination (R^2) between 0.930 and 0.949, the fit for all possible
 199 fission combinations has R^2 of 0.997.

Figure 6: Distribution of time differences for various n^{th} -nearest-neighbor fission events is represented by each colored line, with the black line showing the distribution between all fission events in a chain. The distributions were obtained from an MCNPX-PoliMi simulation of bare BeRP ball (stairs) and then normalized to unity and fit by gamma distributions (solid lines).

200 The results from the bare BeRP case are encouraging, but there are sev-
 201 eral important caveats to consider. Unsurprisingly, the goodness of fit of the
 202 gamma distribution deteriorates for configurations with increasing amount of
 203 moderator. There is nothing in the gamma distribution that captures the effect
 204 that reflected neutrons can have on the fission chain distribution. Most impor-
 205 tantly, however, the simulation in Figure 6 includes the time between fissions
 206 throughout the fissile material, but neglects the effects of attenuation within
 207 the material on particles that escape and are available for detection.

208 3.3. TCPH distribution for multiplying sources

209 The TCPH distribution for a multiplying source is constructed using a linear
 210 combination of the non-multiplying TCPH distribution, as shown in Figure 4(b),
 211 and a TCPH distribution smeared by the gamma distribution. The smearing is
 212 accomplished by setting each bin on the TCPH distribution equal to the sum of
 213 neighboring bins along the time axis, with a weight given by the gamma distri-
 214 bution for the time delay between bins. Included in the sum is the contribution
 215 of the original same fission bin weighted by factor n .

The factor n represents the fraction of correlated pairs from the same fissions
 over all fission and in theory should be proportional to the length of fission chain
 L :

$$n \propto \frac{L}{\binom{L}{2}}. \quad (4)$$

216 By definition the average length of the fission chain is related to the sub-critical
 217 multiplication from Eq. 2 such that $M = (\bar{L} - 1)$.

To account for n in our model we add a conditional statement to the gamma
 distribution:

$$F(x|\alpha, \theta, n) = \begin{cases} f(x|\alpha, \theta) & \text{if } x > 0 \\ n & \text{otherwise.} \end{cases}$$

218 where $f(x)$ is defined in Eq. 3. The smeared TCPH distribution is calculated
 219 by

$$\mathbf{M} = \mathbf{N} \times F(\mathbf{A}) \quad (5)$$

220 where $\mathbf{N}$ is a pulse height by time matrix of the like-fission TCPH distribution.
 221 The matrix $\mathbf{A}$ is a symmetric Toeplitz matrix with the first row defined as the
 222 cumulative difference between the times in the TCPH distribution $\mathbf{N}$. In this
 223 case, the function F performs a point-wise operation on values in matrix $\mathbf{A}$.

224 The resulting TCPH distribution $\mathbf{M}$ depends on α , θ and n which define
 225 how the distribution is smeared, and the distance from the detector which shifts

the non-multiplying TCPH distribution $\mathbf{N}$. If the distance is known it can be fixed, otherwise it can be included as a free parameter with the other three parameters. To solve for the parameters we define the loss function as the root-mean-square error (RMSE) between the modeled TCPH distribution $\mathbf{M}$ and a measured distribution. We then minimize this function by employing an unconstrained non-linear optimization method based upon the Nelder-Mead simplex algorithm [16, 17].

We solve for the parameters of interest in three steps in order to ensure the best-fit parameters. In the first and the final third step we optimize all four parameters, and in the intermediate second step we fix the source-to-detector distance. In between the steps we change the initial guesses to the previous solutions. The Nelder-Mead simplex algorithm works best with fewer parameters, therefore by reducing their number in the middle step we hone in on the best initial guess for the gamma distribution parameters that minimize the RMSE. We decided to fix the distance parameter because it proved to be the most well constrained with the smallest relative covariance with respect to the other parameters.

4. Results and Discussion

4.1. Measurements

4.1.1. Best-fit parameters and their physical interpretations

A subset of the measured TCPH distributions and corresponding optimized models built from best-fit parameters are shown in Figure 7. The like-fission TCPH distribution, shown in Figure 4(b), is smeared to produce TCPH distributions shown in subfigures (b), (d) and (f) of Figure 7. The smearing parameters are optimized in order to match the measured TCPH distribution shown in subfigures (a), (c) and (e) of Figure 7.

The optimized gamma distribution parameters and factors of n are shown in Table 1. We expect that the parameter n would decrease with increasing sub-critical multiplication given its relationship to fission chain length in Eq. 4.

255 This trend is apparent within each shielding material configuration, but is not
 256 present between them. Furthermore, the relative uncertainty in this parameter
 257 is over five times greater than the others, which limits its predictive capability.

258 The shape parameter, α , clearly demarcates the differences between the
 259 polyethylene moderator and tungsten reflector. Although α has no obvious
 260 physical interpretation when its a non-integer less than 1, it is valuable that it
 261 can distinguish between intervening material type independently of multiplica-
 262 tion.

263 We would expect that the rate parameter, θ , would increase with multi-
 264 plication because it is proportional to the degree of smearing of the TCPH
 265 distribution. The corollary to this is the measure of the gradient of the TCPH
 266 distribution found in [6]. This positive correlation is only apparent for the tung-
 267 sten case, and is actually negative for the polyethylene. However, in Section 4.2
 268 we show that for simulated polyethylene cases the rate parameter is positively
 269 correlated with multiplication, as expected.

Table 1: Optimized gamma distribution parameters for the five measured configurations of the BeRP sphere with standard errors of 1 standard deviation shown.

case	α	θ	n	Multiplication
bare	0.57 ± 0.04	12.41 ± 0.62	0.08 ± 0.03	4.429 ± 0.002
1.27 cm W	0.87 ± 0.04	14.91 ± 0.63	0.11 ± 0.03	6.447 ± 0.004
2.54 cm W	0.90 ± 0.02	19.67 ± 0.53	0.06 ± 0.01	8.752 ± 0.006
2.54 cm HDPE	0.48 ± 0.03	26.74 ± 1.38	0.08 ± 0.03	7.743 ± 0.005
7.62 cm HDPE	0.53 ± 0.03	22.54 ± 0.91	0.07 ± 0.02	20.3 ± 0.2

270 The optimized source-to detector distances are shown in Table 2. Multipli-
 271 cation values were calculated from MCNP5 simulations of each configuration
 272 [13]. In all cases the optimized distance was smaller than the measured dis-
 273 tance, with the greatest discrepancy present in the thicker tungsten case. This
 274 suggests that this effect may be due to self-shielding; correlated particles from
 275 fission events closer to the surface of the BeRP ball on the side of the detectors

are more likely to be detected. Therefore, any additional high-Z material would preferentially select for gamma-rays born at the surface of the BeRP ball, which would decrease the optimized source-to-detector distance. We see this effect in simulation, where the source-to-detector distance decreases significantly for increasing tungsten reflector thickness but remains only slightly altered for low-Z polyethylene moderator.

Table 2: Optimized distances for the five measured configurations of the BeRP sphere with standard errors of 1 standard deviation. The actual distance was measured from the center of the BeRP ball to the face of the detectors.

case	optimized distance (cm)	actual distance (cm)
bare	49.02 ± 0.06	50 ± 0.5
1.27 cm W	49.04 ± 0.12	50 ± 0.5
2.54 cm W	44.75 ± 0.11	48 ± 0.5
2.54 cm HDPE	59.72 ± 0.09	60 ± 0.5
7.62 cm HDPE	49.25 ± 0.09	50 ± 0.5

Figure 7: Measured TCPH distributions (left) and corresponding models with parameters from the minimization algorithm (right) for various BeRP ball configurations. Each distribution is normalized to unity and displayed on a logarithmic scale.

282 4.1.2. *Discrepancies in the polyethylene cases*

283 Experimental setup differences and the physics of moderating neutrons offer
284 a couple of explanations for the unexpected lower rate parameter of the 7.62 cm
285 polyethylene case compared with the thinner 2.54 cm polyethylene case. First,
286 the moderator has the effect of slowing the average time between fissions to
287 a microsecond time-scale, which makes true correlations difficult to distinguish
288 from uncorrelated background. The effect that additional polyethylene has in in-
289 creasing multiplication is therefore not captured within the 120 ns measurement
290 correlation time window. The diminishing returns on sensitivity of the TCPH
291 distribution to multiplication with increasing layers of polyethylene moderator
292 has previously been demonstrated in simulation [5].

293 Furthermore, the thinner polyethylene case was measured 10 cm further
294 away, and on a different experimental campaign than the other polyethylene
295 case. A farther source-to-detector distance has the effect of broadening the
296 TCPH distribution, which should be captured by our model. However, it could
297 be that there are nuisance parameters due to the differences between the two
298 experiments that are inexplicably contributing to this discrepancy. Ideally those
299 would be teased out with additional experiments, but with limited access to the
300 NNSA and the BeRP ball we chose to supplement our findings with simulation
301 results.

302 4.2. *Simulations*

303 MCNPX-PoliMi simulations with shielding configurations of 1.27-7.62 cm for
304 polyethylene and 1.27-7.62 cm for tungsten in 1.27 cm intervals were simulated
305 to better illustrate the trends in the TCPH distribution. The optimization
306 results of the shape and rate parameter for both simulations and measurement
307 are shown in Figure 8, with multiplication proportional to the area of each
308 marker.

309 The standard errors were taken from a covariance matrix of the best-fit
310 parameters derived from a numerical estimation of the Jacobian. There are
311 considerable absolute differences between measurement and simulation, but the

general trends remain the same with the notable exception of the aforementioned measured polyethylene cases. The simulations reveal that it would be possible to measure the relative change in multiplication, independently of the change of surrounding moderator for a reflector or vice versa. The main driver in the size of the relative standard errors was the number of total correlated gamma-neutron pairs. This is most apparent for the thick tungsten cases which effectively shield the vast majority of fission gamma-rays.

Figure 8: Optimized shape and rate parameters for both simulation (blue) and measurement (red) cases. The shielding configurations varied in thickness from 1.27-7.62 cm with 1.27 cm intervals. The increase in symbol size corresponded to an increase in multiplication, which is proportional to the area of each marker. Note that the bare configurations are furthest to the left for both measurements and simulations.

319 **5. Conclusions**

We introduced a new approach for the characterization of SNM based on a signature of temporally correlated gamma-rays and neutrons. In our approach we fit the empirical approximation of the timing distribution between fissions in a fission chain to the measured TCPH distribution. We have shown that for the BeRP ball these empirical parameters correlate with both multiplication and the type of material (e.g. low-Z moderator or high-Z reflector) coupled to the fissile

assembly. This property makes this method a candidate for treaty verification applications, where confidence in warhead dismantlement is the objective. In this case warhead dismantlement would involve the removal of high explosives, which is a form of moderating material, from the fissile material of a warhead. Our reliance on a signature that is unique to SNM makes it more difficult to spoof dismantlement of fissile material. Furthermore, the signature we used can be captured with a portable set of fast organic scintillators which could be carried by an inspector.

Though the gamma distribution was shown to reproduce the measured TCPH distributions in bare fissile assemblies, as increasing amounts of reflector were added it became a less relevant proxy for the complete effect of the fission chain dynamics. Despite this we saw that the rate parameter of the optimized gamma distribution correlated positively with multiplication. Simulation results revealed this trend, but it was not shown to be the case for the measured polyethylene cases. This multiplication-rate parameter relationship followed a different trend-line for moderated and reflected systems; which were easily identified by the shape parameter.

For this work, an empirical model was utilized to identify general trends in the fissile assemblies. However, if this were replaced by a physical model of the timing distribution of fissions within fission chains, then it may be possible to further improve the available information present in this signature. In future work, it may be found that potential candidate models such as multi-region point-kinetics, 1D deterministic transport solvers, or even simple fission chain Monte Carlo simulations can be used to relate this signature more directly to the physical distribution of fissile material.

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References

- [1] N. Ensslin, Principles of Neutron Coincidence Counting, Los Alamos National Laboratory, LA-13422-M (November 1998).
- [2] A. Enqvist, M. Flaska, S. Pozzi, Measurement and simulation of neutron/gamma-ray cross-correlation functions from spontaneous fission, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 595 (2) (2008) 426 – 430. doi:10.1016/j.nima.2008.07.007.
- [3] E. Miller, J. Dolan, S. Clarke, M. Flaska, S. Pozzi, E. Padovani, P. Peerani, P. Schillebeeck, Experiments and simulation of cross-correlations on MOX fuel, in: Proceedings of the Institute of Nuclear Materials Management 52nd Annual Meeting, Palm Desert, California, 2011.
- [4] J. Mattingly, Polyethylene-reflected plutonium metal sphere: Subcritical neutron and gamma measurements, Tech. Rep. SAND2009-5804, Sandia National Laboratories (2009).
- [5] E. Miller, S. Clarke, A. Enqvist, S. A. Pozzi, P. Marleau, J. K. Mattingly, Characterization of special nuclear material using a time-correlated pulse-height analysis, Journal of Nuclear Materials Management XLI (1) (2012) 32–37.
- [6] E. Miller, J. Dolan, S. Clarke, S. Pozzi, A. Tomanin, P. Peerani, P. Marleau, J. Mattingly, Time-correlated pulse-height measurements of low-multiplying nuclear materials, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 729 (2013) 108 – 116. doi:10.1016/j.nima.2013.06.062.
- [7] P. Marleau, A. Nowack, M. Paff, M. Monterial, S. Clarke, S. Pozzi, Gamma/neutron time-correlation for special nuclear material detection active stimulation of highly enriched uranium, Tech. Rep. SAND2013-7442, Sandia National Laboratories (2013).

- 393 [8] M. G. Paff, M. Monterial, P. Marleau, S. Kiff, A. Nowack, S. D. Clarke,
394 S. A. Pozzi, Gamma/neutron time-correlation for special nuclear material
395 detection active stimulation of highly enriched uranium, *Annals of Nuclear*
396 *Energy* 72 (2014) 358 – 366. doi:doi.org/10.1016/j.anucene.2014.06.
397 004.
- 398 [9] M. Monterial, M. Paff, S. Clarke, E. Miller, S. A. Pozzi, P. Marleau, A. N.
399 S. Kiff, J. K. Mattingly, Time-correlated-pulse-height technique measure-
400 ments of fissile samples at the device assembly facility., in: *Proceedings*
401 *of the Institute of Nuclear Materials Management 54th Annual Meeting*,
402 *Palm Desert, California, 2013*.
- 403 [10] G. F. Knoll, *Radiation Detection and Measurement*, 3rd Edition, John
404 Wiley & Sons, Inc., 2000.
- 405 [11] J. Adams, G. White, A versatile pulse shape discriminator for charged
406 particle separation and its application to fast neutron time-of-flight spec-
407 troscopy, *Nuclear Instruments and Methods* 156 (3) (1978) 459 – 476.
408 doi:10.1016/0029-554X(78)90746-2.
- 409 [12] S. A. Pozzi, E. Padovani, M. Marseguerra, MCNP-PoliMi: a Monte-Carlo
410 code for correlation measurements, *Nuclear Instruments and Methods in*
411 *Physics Research Section A: Accelerators, Spectrometers, Detectors and*
412 *Associated Equipment* 513 (3) (2003) 550 – 558. doi:doi.org/10.1016/
413 j.nima.2003.06.012.
- 414 [13] X-5 Monte Carlo Team, *MCNP - A General Monte Carlo N-Particle Trans-*
415 *port Code, Version 5, Volume I: Overview and Theory*, Los Alamos National
416 *Laboratory, Appendix H - Fission Spectra Constants and Flux-To-Dose*
417 *Factors Constant for Fission Spectra (April 2003)*.
- 418 [14] A. Enqvist, C. C. Lawrence, B. M. Wieger, S. A. Pozzi, T. N. Massey,
419 Neutron light output response and resolution functions in EJ-309 liquid

- 420 scintillation detectors, Nuclear Instruments and Methods in Physics Re-
 421 search Section A: Accelerators, Spectrometers, Detectors and Associated
 422 Equipment 715 (2013) 79 – 86. doi:10.1016/j.nima.2013.03.032.
- 423 [15] C. C. Lawrence, M. Febbraro, T. N. Massey, M. Flaska, F. Becchetti, S. A.
 424 Pozzi, Neutron response characterization for an EJ299-33 plastic scintilla-
 425 tion detector, Nuclear Instruments and Methods in Physics Research Sec-
 426 tion A: Accelerators, Spectrometers, Detectors and Associated Equipment
 427 759 (2014) 16 – 22. doi:10.1016/j.nima.2014.04.062.
- 428 [16] J. A. Nelder, R. Mead, A simplex method for function minimization, The
 429 Computer Journal 7 (4) (1965) 308–313. doi:10.1093/comjnl/7.4.308.
- 430 [17] J. C. Lagarias, J. A. Reeds, M. H. Wright, P. E. Wright, Convergence
 431 properties of the Nelder-Mead simplex method in low dimensions, SIAM
 432 Journal of Optimization 9 (1998) 112–147.