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Gamma/Neutron Time-Correlation for Special Nuclear Material Detection – Active Stimulation of Highly Enriched Uranium

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Gamma/Neutron Time-Correlation for Special Nuclear Material Characterization – Active Stimulation of Highly Enriched Uranium

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Abstract

A series of simulations and experiments were undertaken to explore and evaluate the potential for a novel new technique for fissile material detection and characterization, the time-correlated pulse-height (TCPH) method, to be used concurrent with active stimulation of potential nuclear materials. In previous work TCPH has been established as a highly sensitive method for the detection and characterization of configurations of fissile material containing Plutonium in passive measurements. By actively stimulating fission with the introduction of an external radiation source, we have shown that TCPH is also an effective method of detecting and characterizing configurations of fissile material containing Highly Enriched Uranium (HEU). The TCPH method is shown to be robust in the presence of the proper choice of external radiation source. An evaluation of potential interrogation sources is presented.

ACKNOWLEDGMENTS

I would like to thank our University of Michigan collaborators for the Monte Carlo simulations support and the support staff at the Device Assembly Facility at the Nevada National Security Site for all of their help in support of the measurements made there.

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NOMENCLATURE

Am-Li	Americium-Lithium (alpha,n) neutron source
BeRP	Beryllium-Reflected Plutonium
DAF	Device Assembly Facility
D-D	Deuterium-Deuterium neutron generator
DOE	Department of Energy
D-T	Deuterium-Tritium neutron generator
DU	Depleted Uranium
HDPE	High Density Polyethylene
HEU	Highly Enriched Uranium
LANL	Los Alamos National Laboratory
MCNP	Monte Carlo N-Particle
MLEM	Maximum Likelihood Expectation Maximization
NNSS	Nevada National Security Site
PMT	Photomultiplier Tube
RGD	Radiation Generating Device
SF	Spontaneous Fission
SNL	Sandia National Laboratories
SNM	Special Nuclear Material
TCPH	Time-Correlated Pulse Height
TOF	Time of flight
TTAL	Theoretic Time of Arrival
WGPu	Weapons Grade Plutonium

1.0. INTRODUCTION

In a previous report, "Gamma/Neutron Time-Correlation and Spectrometry for Special Nuclear Material Characterization – BeRP Ball Measurements", we have established a new method to detect and characterize fissionable material, the Time Correlated Pulse Height (TCPH) method. This method has been validated in a series of subcritical experiments using an α-phase, weapons-grade plutonium (WGPu) metal sphere, the beryllium-reflected plutonium (BeRP) ball (1), in the Nevada Nuclear Security Site (NNSS) Device Assembly Facility (DAF). In past work Monte Carlo simulations of the experimental configurations at DAF using the Monte Carlo N-Particle (MCNP) package were validated and used to establish detector response matrices. We demonstrated that Maximum Likelihood Expectation Maximization (MLEM) algorithms can be employed for both the efficient detection of fissile material and the characterization of the type and thickness of moderating materials in the vicinity of multiplying nuclear material. In Section 1.1 we briefly describe the TCPH method, however a more complete description of the technique is offered in the aforementioned report.

In all experimental configurations to date, only passive measurements have been made, meaning no external source of radiation was introduced to the interrogated material. The purpose of this work is to explore and evaluate the use of the TCPH technique during active stimulation of the nuclear material.

In Section 2 we motivate the need for active stimulation of materials to gain sensitivity to the presence of Highly Enriched Uranium (HEU). Due to the low spontaneous fission rate of Uranium, passive detection is exceedingly difficult, particularly when gamma-ray signatures may be shielded by the presence of high-Z materials.

In Section 3 we report on Monte Carlo simulations of active measurements of HEU. In Section 3.1 we evaluate possible radiation sources that can be used as to introduce the stimulating radiation. In Section 3.3 it is shown that with the introduction of active neutron stimulation, significant quantities of HEU can be detected and characterized in reasonable dwell times (10s of minutes to hours).

Finally in Section 4 we validate some of the predictions made in a series of laboratory experiments. Measurements were made with bare Californium-252 (^{252}Cf) and Americium-Lithium (Am-Li) radiological sources as well as a deuterium-deuterium radiation generating device (D-D generator). It was found that the ^{252}Cf source emits too many high energy correlated gammas and neutrons to be useful as an interrogating source. Because of the low energy of the correlated gamma-rays and neutrons emitted by the Am-Li source, nearly no gamma-neutron correlations are detected making Am-Li an ideal radiological active stimulation source. However, because it may be desirable to switch the interrogating radiation on and off and because they have no intrinsic gamma-neutron correlations, a D-D neutron source is also nearly ideal.

1.1. Motivation and Background

The presence of shielding and background radiation makes detecting special nuclear material (SNM) with gamma spectroscopy exceedingly difficult. Gamma-ray emission rates are often too low for timely detection of SNM and small amounts of shielding even further reduce any measurable signal. An intelligent detection system recognizes the fact that SNM emits both gamma rays and neutrons. Many nuclear materials emit temporally correlated gamma rays and neutrons, but SNM is unique in that these temporal correlations can extend over longer times. In a fissile source with multiplication greater than one, such as WGPu or highly enriched uranium (HEU), neutrons from a single fission induce further fission events, thus creating correlated fission chains.

The time-correlated-pulse-height (TCPH) technique measures temporally correlated photon-neutron pairs (2) (3). For such a measurement to be possible, detectors with nanosecond-scale timing and reliable photon-neutron pulse shape discrimination capability are required. Organic liquid scintillation detectors fulfill both of these requirements (4) and are thus well suited for this measurement technique. In this work, an array of four organic liquid scintillator detectors is used. A photon interaction in one detector followed by a neutron interaction in another detector, within a time window of 100 nanoseconds, is considered to be a photon-neutron correlated pair.

For each measured photon-neutron correlated pair, two pieces of information are of interest: the difference in time-of-flight between the arrival of the photon and neutron, and the measured pulse height energy (in Mega-electron Volts electon equivalent energy (MeVee)) due to the neutron interaction in the detectors. A surface plot is created from these data and information regarding the correlated fission chain dynamics is extracted (5).

Assuming that the photon and neutron are emitted from the same fission, there is a maximum time-of-flight difference for a photon (2nd term) and neutron (1st term) given a fixed source-to-detector distance:

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

where d, E_n, and M_n are the source-to-detector distance, energy of the neutron and its mass, respectively. Calculating this maximum time of arrival for a large range of neutron energies thus provides a discrimination line that is superimposed on the TCPH plot. We will hereafter refer to this line as the theoretical time of arrival line (TTAL). Any correlated photon-neutron pair arising from the same fission thus must fall beneath this line. However, correlated photon-neutron pairs from different fissions within a fission chain, as would be seen in a multiplying source, can appear above and beyond this discrimination line. It should be noted that because the neutron can also be emitted before the detected photon, fission chain dynamics will also smear the distribution to shorter times to the left of the TTAL.

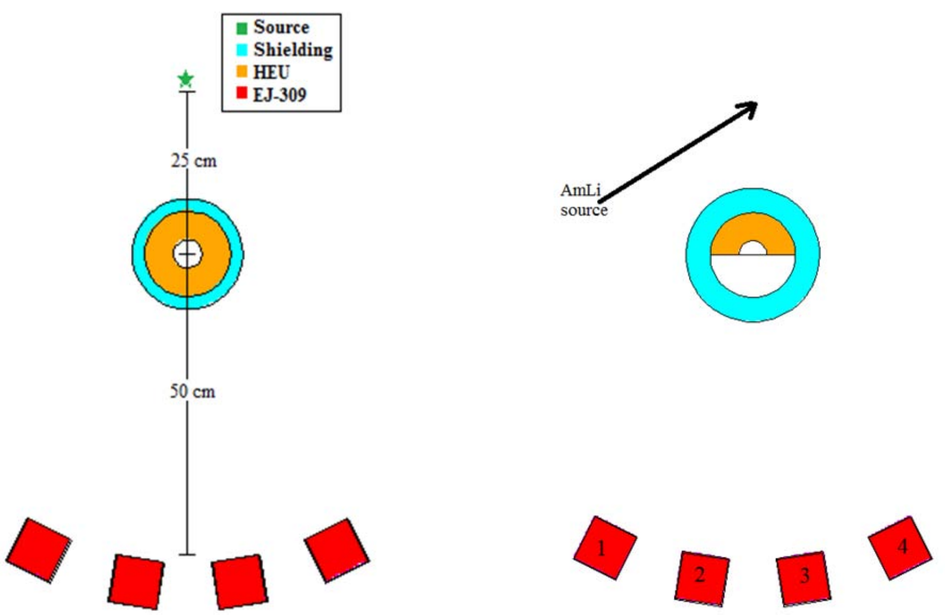
Thus, TCPH measurements can distinguish non-multiplying correlated photon-neutron sources, like industrial (α,n) sources and ^{252}Cf spontaneous fission sources, from multiplying correlated photon-neutron sources, i.e. SNM. Additionally, for multiplying sources, the ratio of events falling above and below the discrimination line in the TCPH plot scales with the SNM's self-multiplication. Thus, TCPH allows for a clear distinction between the fissile threat material, and any other nuclear material that would otherwise contribute to a false positive.

The TCPH technique can be used both passively and actively, i.e. with the presence of an interrogating source to induce more fission in the SNM. Both options exhibit their inherent advantages and disadvantages. The passive measurement technique is simpler and foregoes the handling of an active source by personnel and thus the associated dose. However, active techniques with their capacity to induce fission within a material can considerably shorten the required measurement time, albeit at the cost of adding further complications to the measurement.

For the measurement of HEU in particular, the passive technique is impractical for real world applications. The primary contribution of spontaneous fission in HEU comes from ^{238}U and even so the probability of spontaneous fission is low, as is its specific activity. As a result, the time required to get meaningful counts on a TCPH distribution is on the order of years. For this reason it is imperative that an active source is used to facilitate the detection HEU.

1.2. Geometry of Interest

The HEU source chosen for this study are the Rocky Flats HEU shells (6). This configuration of HEU is unclassified and available at the DAF at the NNSS for possible future measurements. For all simulated cases a subset of the Rocky Flats shells were placed 50 cm away from the faces of four simulated 7.62 by 7.62 cm EJ-309 detectors with the source 25 cm away from the center of the HEU on the side directly opposite the detectors. Each detector is spaced 18^{o} apart in a symmetrical configuration oriented toward the center of the HEU along an arc. An illustration of the MCNPX model used is shown in Figure 1. The source is shown as a star for illustration purposes only, in reality the source was modeled as a point-source. The HEU target has a mass around 25.4 kg for the full sphere, shown in Figure 1(a), and 12.7 kg for the hemisphere, shown in Figure 1(b). For both cases bare configurations, in which no moderator is present, were also used but are not shown. For the 12.7 kg hemisphere a moderator configuration in which only the half of the HEU facing the source was moderated was used but is not shown. This configuration was chosen to explore the effects of moderator on neutrons emitted by the HEU while allowing neutrons from the interrogating source to be moderated. These two configurations of moderator for the 12.7 kg hemisphere will be referred to as "full moderated" and "half moderated" throughout the report.



(a) (b)

Figure 1: MCNPX geometry used in simulations,(a) the sphere configuration with 2 cm of polyethylene (b) hemisphere with 4 cm of polyethylene are shown.

2. PASSIVE MEASUREMENTS OF HEU

In passive TCPH measurements there are no natural correlated photon-neutron events that may "contaminate" the TCPH plot other than potential accidental uncorrelated coincidences. On the other hand, as will be shown in the simulation results, the specific activity of some SNM, such as HEU, may be so low, that extraordinarily long measurement times are required to obtain a statistically meaningful TCPH plot. This noticeably conflicts with the general desire for timely detection.

For simulations of passive TCPH measurements of HEU, the built-in ^{238}U spontaneous fission source was used in MCNPX-PoliMi particle transport code (7). Its neutron and photon emission energy spectra are shown in Figure 2. The total pulse height distributions (total, neutrons, photons) measured with all four detectors are shown in Figure 3 for the bare (Figure 3a) and two moderated HEU simulations (Figure 3b and Figure 3c). One can clearly see how the addition of polyethylene moderator gradually increases the total pulse height distribution, but in the case of the neutron pulse height distribution, more polyethylene, as seen in Figure 3c, also eventually shields some neutrons from reaching the detectors.

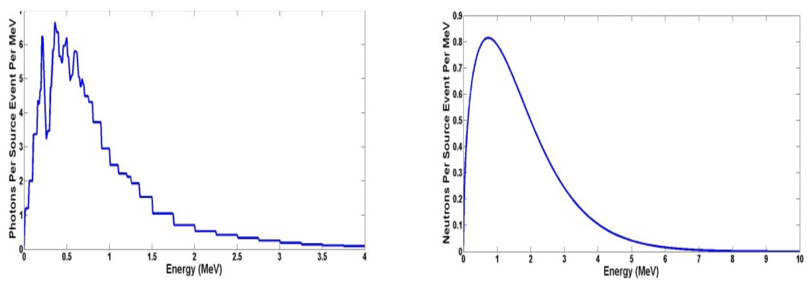
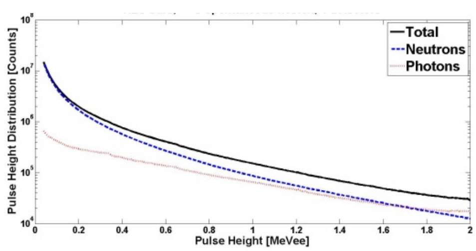
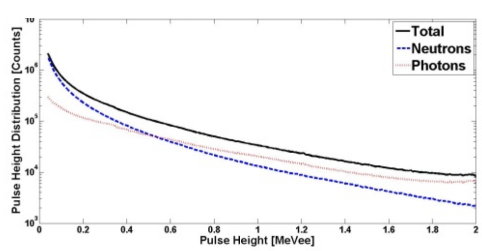


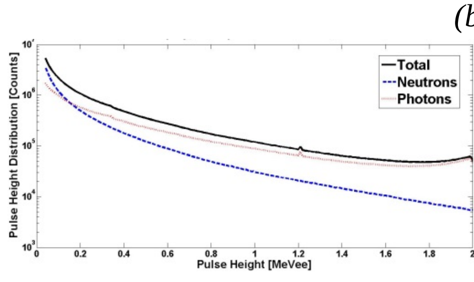
Figure 2: MCNPX-PoliMi built-in ^{238}U spontaneous (left) fission photon and (right) neutron spectra



(a)



(b)



(c)

Figure 3: Pulse height distributions (total, neutron, photon) for all 4 detectors;(a)passive bare HEU ^{238}U SF source; (b) passive HEU ^{238}U SF source + 2 cm poly; (c) passive HEU ^{238}U SF source + 4 cm poly

To demonstrate the need of adding an external source of neutrons to induce fissions in the HEU, a series of passive cases were simulated in which a ^{238}U spontaneous fission (SF) source was distributed throughout the HEU. The four configurations consisted of one 12.7 kg half-sphere of bare HEU and three configurations of 25.4 kg sphere of HEU, bare and moderated with 2 cm and 4 cm of polyethylene.

The TCPH distributions for ^{238}U SF source cases are shown in Figure 4. For the bare 25.4 kg configuration 18 billion fissions were simulated, and 1.8 billion fissions were simulated for the moderated 25.4 kg configurations as well as the bare 12.7 kg configuration. In all cases it is assumed that the HEU contains 5.3% ^{238}U. Assuming 12,400 Bq/g specific activity of ^{238}U with a SF probability of 0.00005% (8) the equivalent measurement times represent 68 and 6.8 years for the 25.4 kg configurations and 13.7 years for the 12.7 kg configuration. The color scale on these plots is a log-scale. Since the TCPH distributions were scaled to count rates (counts/sec), these distributions show about 1 count for every 3 months. This is not practical for real life scenarios and thus an active source is needed.

The TCPH plots also show how the TCPH ratios increase with increasing source multiplication which is achieved by the addition of moderator, as seen for the bare and two moderated configurations of the 25.4 kg HEU sphere in Figure 4. The multiplication values were computed using equation (2) with the k_{eff} values shown in Table 3 and Table 4 from Monte Carlo k-calculations:

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

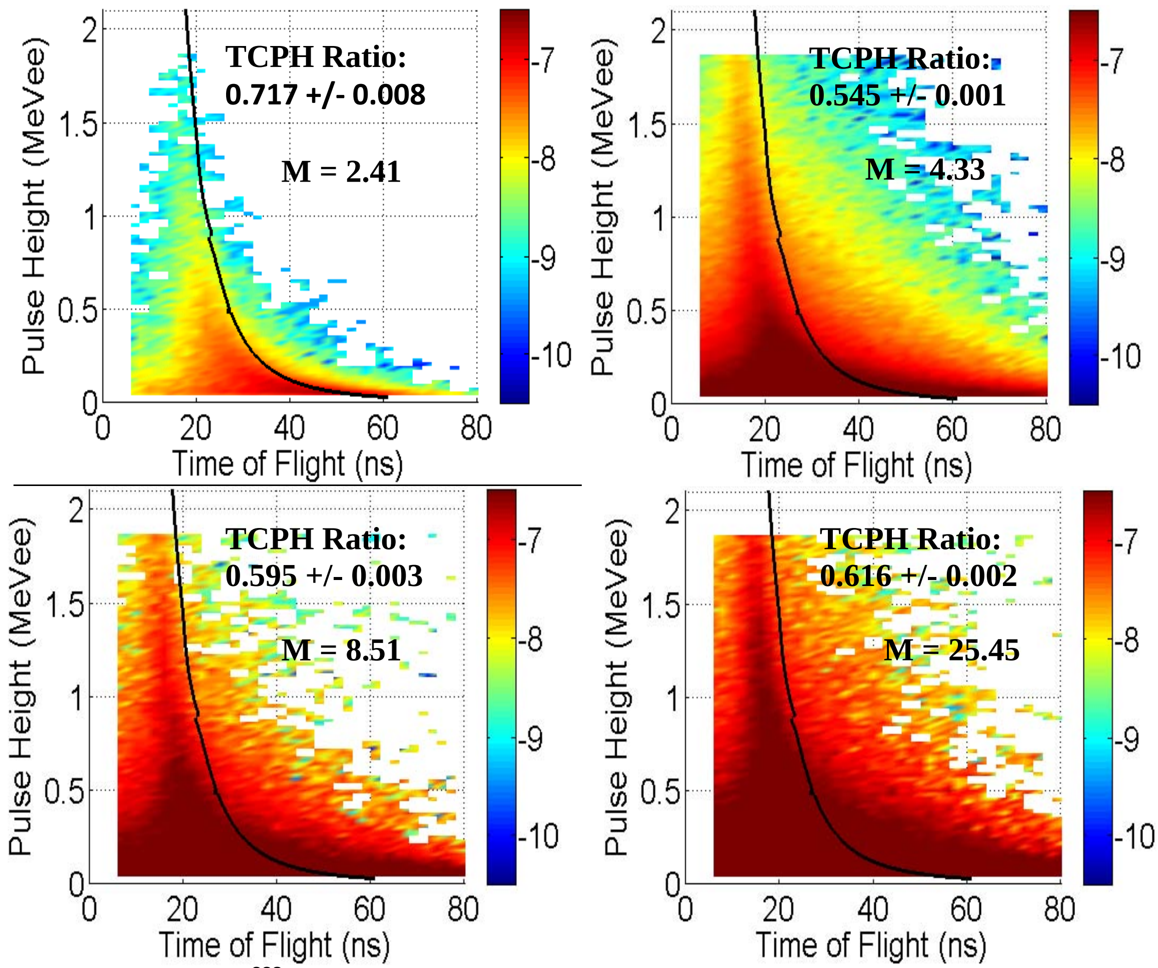


Figure 4: TCPH for ^{238}U passive source in clockwise direction starting from top left: 12.7 kg bare, 25.4 kg bare, 2 cm and 4 cm configurations.

3. SIMULATIONS OF ACTIVE STIMULATION OF HEU

In order to increase the count rate of correlated photon-neutron pairs, one could naturally resort to an external interrogating source of neutrons to induce many more fissions in the SNM.

3.1. Interrogation Source Selection

Ideally the interrogating source should not emit any correlated photon-neutron pairs; these could contribute to the TCPH plot and must be accounted for to detect the photon-neutron pairs from the SNM. In practice, any commonly available external neutron source will also emit gamma rays; however, if these gamma rays are of sufficiently low energy to fall below the detector threshold, they may not contribute to the TCPH plot. Similarly, it is important to consider the neutron spectrum of the external source. Thermal neutrons are ideal for inducing fission in ^{235}U while higher energy neutrons may lead to undesirable induced fissions in ^{238}U and registration in the neutron detectors.

The MCNPX-PoliMi particle transport code in conjunction with its post-processor MPPost was used for all simulated TCPH plots (9). The photon and neutron spectra emitted by the built-in PoliMi sources for the active interrogation TCPH simulations are shown in the following figures.

Figure 5 shows the simulated source spectra for ^{252}Cf, a spontaneous fission source. Californium's specific activity is such that even a small quantity is sufficient to interrogate SNM; however, on average each spontaneous fission emits a large number of photons and neutrons with energies above typical detector thresholds. Thus, as seen in Figure 6(a), the interrogating source may produce its own TCPH distribution. If the HEU is not present, but one uses the TTAL for theoretical TCPH events from the SNM, then one would obtain a TCPH plot as show in Figure 6(b). In that figure, TCPH events from ^{252}Cf are occurring above the TTAL drawn for the theoretically present SNM. When the HEU is added in the simulation as shown in Figure 6(c), one then obtains TCPH events both from the HEU and the ^{252}Cf. Because the ^{252}Cf source-to-detector distance is different from the SNM to detector distance, this can lead to a TCPH plot with two overlapping distributions. If the ^{252}Cf source is further away than the SNM from the detectors, it would lead to contamination in the TCPH plot above TTAL which gives a higher TCPH ratio than predicted by multiplication of the target. In theory, the contribution of ^{252}Cf source to overall TCPH distribution can be subtracted away, but at the expense of adding statistical uncertainty. ^{252}Cf 's 2.645-year half-life also means the source would have to be replaced periodically.

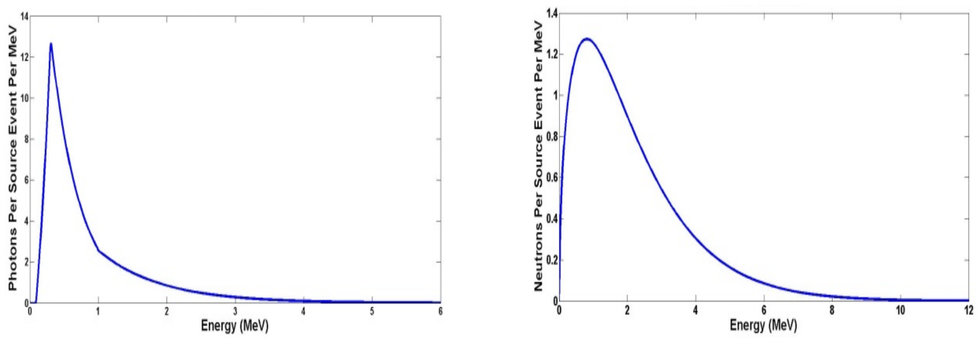
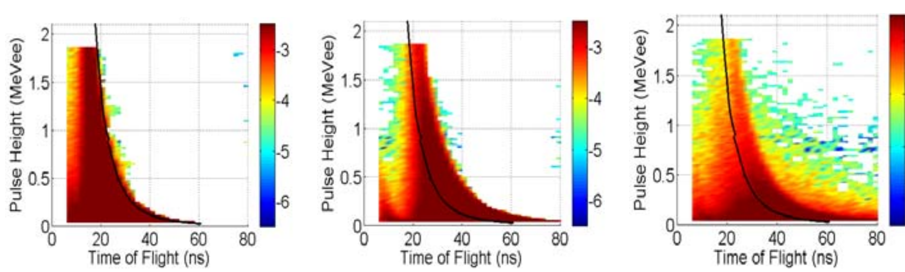


Figure 5: MCNPX-PoliMi built-in ^{252}Cf spontaneous fission (left) photon and (right) neutron spectra



(a) *(b)* *(c)*

Figure 6: TCPH plots of (a) bare ^{252}Cf source with 50 cm TTAL, (b) bare ^{252}Cf source with 75 cm TTAL, (c) bare ^{252}Cf source with HEU and with 50 cm TTAL

Another external neutron interrogation source option is a common deuterium-deuterium radiation generating device (D-D neutron generator). In our simulations, this is modeled as a mono-energetic 2.45 MeV neutron source (10). Photons are not simulated, although some photons are created in D-D generators that could lead to additional accidentals in the TCPH plot. As will be shown later, similar fission rates as those obtained with Americium-Lithium (Am-Li) can be obtained with D-D neutrons. The advantages of D-D generators are their compactness, portability and the ability to shut them off when not in use.

Am-Li, an (α,n) source, shows much promise. Its photon spectrum (see Figure 7(a)) reveals that nearly all emitted photons are well below commonly used detector thresholds for organic liquid scintillation detectors. This means that no correlated photon-neutron pairs from the Am-Li source should appear in TCPH plots, in contrast to what is seen with ^{252}Cf. However, in practice Am-Li sources may contain contaminants, such as beryllium and oxygen at the few percent level, that could lead to the production of higher energy correlated gamma rays (11). The maximum and average neutron emission energies from an Am-Li source (see Figure 7(b)) are much lower than those of ^{252}Cf, thus significantly reducing the contribution of the interrogating source to the TCPH plots.

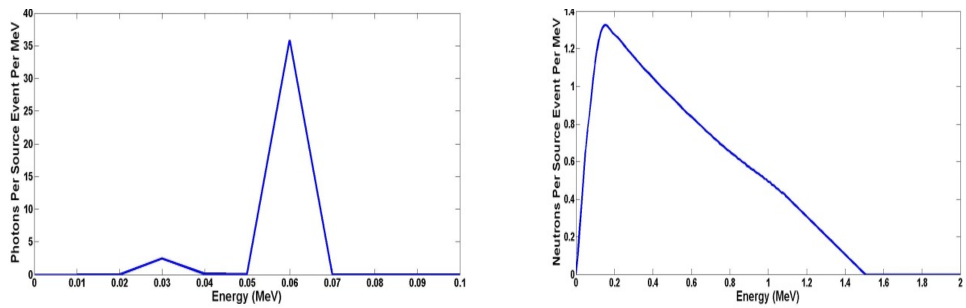


Figure 7: MCNPX-PoliMi built-in Am-Li (α,n) (left) photon and (right) neutron spectra

3.2. Moderator Effects

As shown in a series of simulation results in Figure 8, the TCPH ratio is strongly correlated to both the amount of moderator and the total fission rate (both being dependent on the multiplication). One would expect fission rates to initially increase with increasing moderator thickness as the probability of thermalizing incident neutrons increases, thus increasing the population of neutrons with high probability of inducing fission within the SNM. However, as moderator thickness increases, neutrons from the active interrogation source as well as the fission neutrons released by the SNM are shielded. Therefore, an optimum moderator thickness exists.

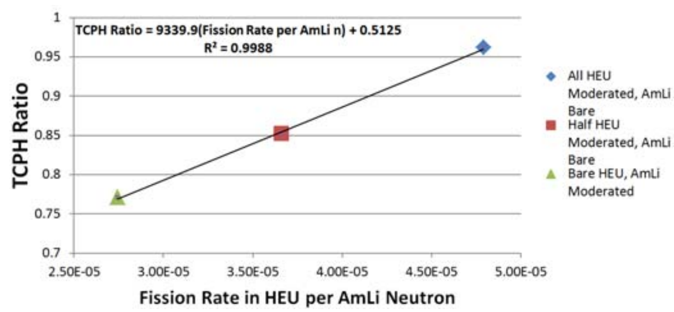


Figure 8*: TCPH ratio as a function of fission rate per source neutron for three Am-Li active interrogation of HEU simulations*

Using the same simulation geometry as shown in Figure 1, polyethylene moderator optimization results were obtained for active interrogation using Am-Li (Table 1and Figure 9), as well as D-D (Table 2 and Figure 10). The optimum polyethylene moderator radii for the Am-Li and D-D cases were thus found to be 4 cm and 5 cm, respectively. This is not surprising, since the average neutron energy emitted by D-D is higher than that emitted by Am-Li, thus requiring a slightly thicker moderator for the D-D case. While surrounding the SNM with moderator may not be a realistic option for actual measurements, these simulations still give a reasonable representation of the expected effects of moderator presence in a source configuration.

Table 1*:* Polyethylene moderator optimization simulation analysis results for active interrogation of HEU with Am-Li

Radius (cm)	Fission Rate in HEU (fissions per cm³ per Am-Li neutron)	Flux in HEU (neutrons per cm² per Am-Li neutron)	Integrated Current Exiting Poly (neutron per Am-Li neutron)
10	1.1972E-05	1.0302E-04	3.05175E-01
9	1.5155E-05	1.3310E-04	3.97657E-01
8	1.8669E-05	1.6846E-04	5.05791E-01
7	2.2243E-05	2.0687E-04	6.24874E-01
6	2.5247E-05	2.4631E-04	7.45647E-01
5	2.6995E-05	2.7959E-04	8.54478E-01
4	2.7036E-05	3.0558E-04	9.36263E-01
3	2.5253E-05	3.1961E-04	9.82302E-01
2	2.2279E-05	3.2304E-04	9.97901E-01
1	1.9435E-05	3.1892E-04	9.99949E-01

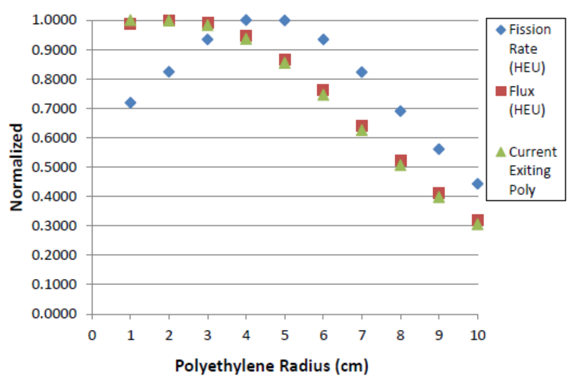


Figure 9: Polyethylene moderator optimization simulation analysis results for active interrogation of HEU with Am-Li

Table 2: Polyethylene moderator optimization simulation analysis results for active interrogation of HEU with D-D

Radius (cm)	Fission Rate in HEU (fissions per cm³ per DD neutron)	Flux in HEU (neutrons per cm² per DD neutron)	Integrated Current Exiting Poly (neutron per DD neutron)
10	1.78797E-05	3.21380E-02	6.68979E-01
9	1.93551E-05	3.48845E-02	7.36289E-01
8	2.06688E-05	3.76490E-02	8.02360E-01
7	2.17457E-05	3.99517E-02	8.63811E-01
6	2.22618E-05	4.17247E-02	9.16607E-01
5	2.24189E-05	4.29553E-02	9.57045E-01
4	2.21025E-05	4.35137E-02	9.82922E-01
3	2.13173E-05	4.33450E-02	9.95262E-01
2	2.06630E-05	4.31757E-02	9.98930E-01
1	2.03846E-05	4.34025E-02	9.99593E-01

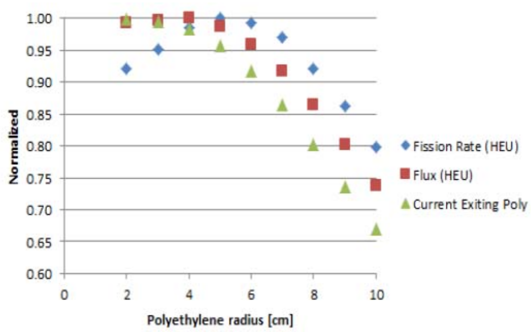


Figure 10: Polyethylene moderator optimization simulation analysis results for active interrogation of HEU with D-D

3.3. Active Measurement Simulations

Three active sources were modeled: Am-Li, D-D generator, and ^{252}Cf. These were chosen to demonstrate the importance of choosing the proper source when performing active interrogation and seeking TCPH distributions. The source must induce fission in the target, while also minimally impacting the TCPH distribution measured.

The pulse height distributions are shown in Figure 11 and the TCPH plots are shown in Figure 12 for the 12.7 kg hemispheres of HEU configurations with an external Am-Li source. In Figure 11(a) one can see the pulse height distribution obtained from Am-Li with no HEU present. It can be seen that compared to ^{252}Cf, the obtained photon pulse height distribution is several orders of magnitude lower, thus Am-Li exhibits no measurable photon-neutron correlated pairs. Integrating the photon pulse height distributions for ^{252}Cf (Fig. 14(a)) and Am-Li (Figure 11(a)) shows that for the same measurement time and detector threshold of 30 keVee, Am-Li provides over a 99% reduction in photons detected than ^{252}Cf.

Recalling the Am-Li photon source spectrum from Figure 7(a), one might expect no photon pulse height distribution to be present, but 2.2 MeV gamma-rays from the capture of neutron on the hydrogen in the detectors are detected. However, these capture gammas are not correlated to the emission of a detectable neutron (it having been "captured") and thus they will not contribute to the TCPH distribution.

The pulse height distributions for the two moderated configurations shown in Figure 11(c) and (d) indicate that the addition of polyethylene moderator increases the pulse height distributions slightly. The two moderator configurations are the full and half configurations described shown in Figure 1(b). These configurations also show increasing neutron distribution over TTAL with increasing amount of moderator, as is also reflected in the shown TCPH ratios on the TCPH plots in Figure 12.

The TCPH distributions for the Am-Li source with 25.4 kg sphere of HEU configurations are shown in Figure 13. For each configuration 18 billion source neutrons were simulated. The TCPH distributions were scaled to absolute count rate (1/s) by assuming an interrogating neutron source rate of 1 million n/s. These three configurations represent 5 hours of measurement time each. Because of multiplication within the source, the distribution of neutrons increases beyond the theoretical time of arrival line with the addition of moderator. This trend is seen in the TCPH ratios included in Figure 13 for each configuration. These ratios also increase with the shown multiplication values computed with equation (2).

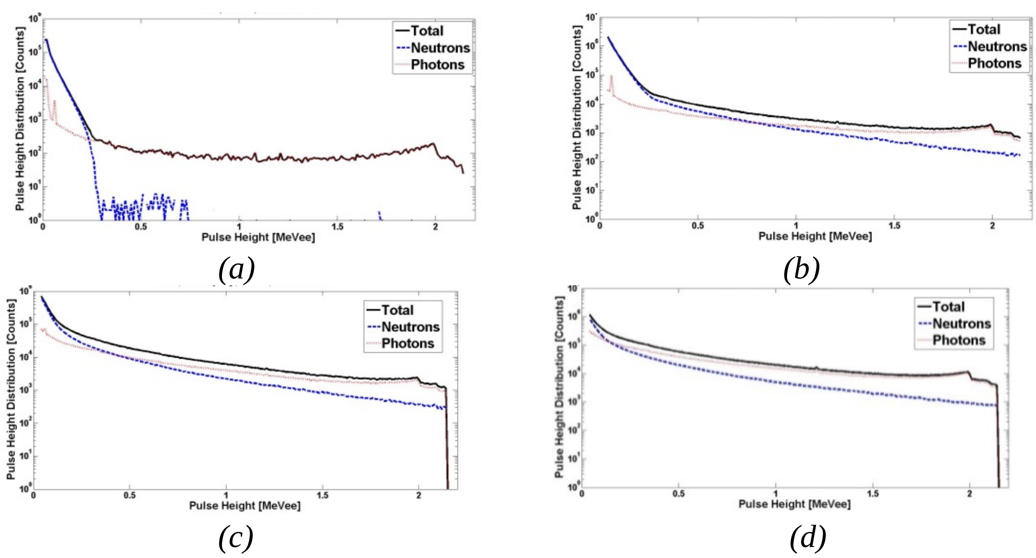


Figure 11*: Pulse height distributions (total, neutron, photon) for all 4 detectors;(a)active, HEU voided, Am-Li source; (b) active, HEU bare, Am-Li source ; (c) active, HEU + 2cm poly, Am-Li source; (d) active, HEU + 4cm poly, Am-Li source*

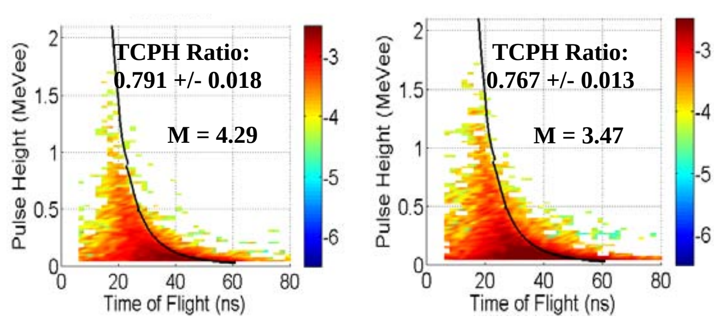


Figure 12: TCPH for Am-Li source with 12.7 kg hemisphere of HEU with full moderation (left) and half moderation (right).

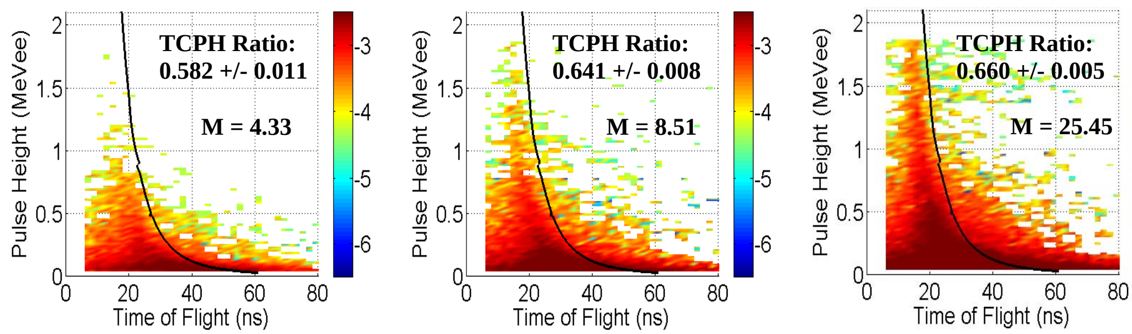
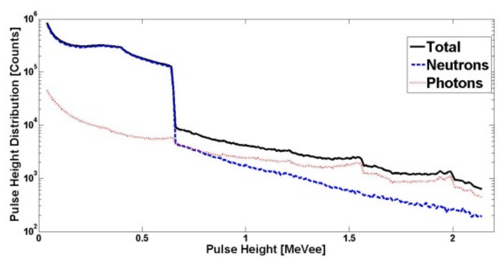
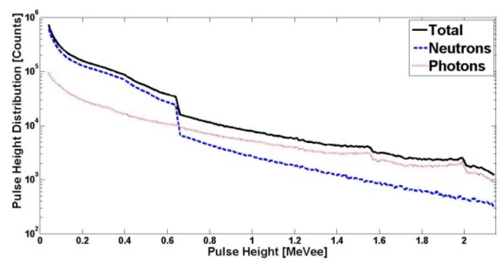


Figure 13: TCPH for Am-Li source with 25.4 kg sphere of HEU from left to right: bare, 2 cm and 4 cm configurations.

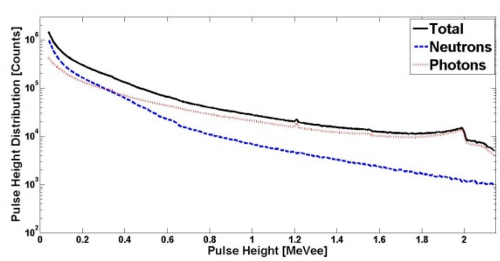
The pulse height distributions and TCPH distributions for D-D generator configurations are shown in Figure 14 and Figure 15, respectively. For each case, 18 billion neutrons were simulated. The TCPH distributions were scaled assuming D-D neutron generation rate of 100 million neutrons per second. This represents around 3 minutes of measurement time, significantly shorter than Am-Li source, an advantage gained through D-D generators greater neutron source strength. The increase of photon-neutron correlations above TTAL is evident as moderator is added to the configuration. The TCPH ratio does not change between 2 and 4 cm configurations, within statistical uncertainty. This could be attributed to the increased thermalization from the moderator pushing the photon-neutron correlations beyond the time window of the TCPH distribution.



(a)



(b)



(c)

Figure 14: Pulse height distributions (total, neutron, photon) for all 4 detectors; (a) active, HEU bare, D-D generator ; (b) active, HEU +2cm poly, D-D generator; (c) active, HEU +4cm poly, D-D generator.

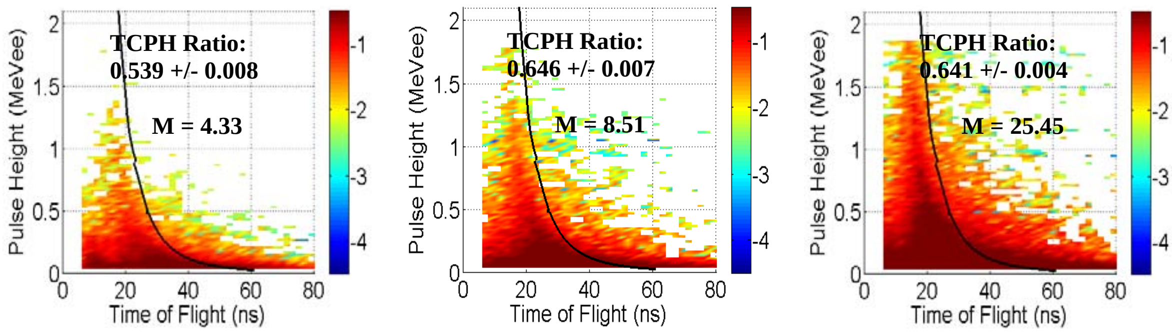


Figure 15: TCPH for D-D generator from left to right: bare, 2 cm and 4 cm configurations.

The pulse height distributions and TCPH distributions for ^{252}Cf source cases are shown in Figure 16 and Figure 17, respectively. For each case, 18 billion fissions were simulated. Again, the TCPH distributions were scaled by the assumption of an interrogating neutron source rate of 1 million n/s such that these three configurations represent 18.75 hours of measurement time each. Unlike the Am-Li cases, the increase in neutron distribution of the TTAL is unclear from these TCPH distributions. In fact the ratio of neutrons above and below TTAL appears to be decreasing with increasing moderators.

As discussed in previous sections, this effect is due to the contribution of the ^{252}Cf source to the TCPH plot. Since the ^{252}Cf source is farther away from the detectors than the HEU, the ^{252}Cf contribution to the TCPH distribution lies beyond the TTAL. As a result the bare configuration has a disproportionally high number of neutrons above the TTAL. These neutrons diminish, however, as polyethylene is added which acts effectively as a shield for the ^{252}Cf source neutrons. As more moderator is added, induced fission in the HEU begins to dominate the TCPH distribution.

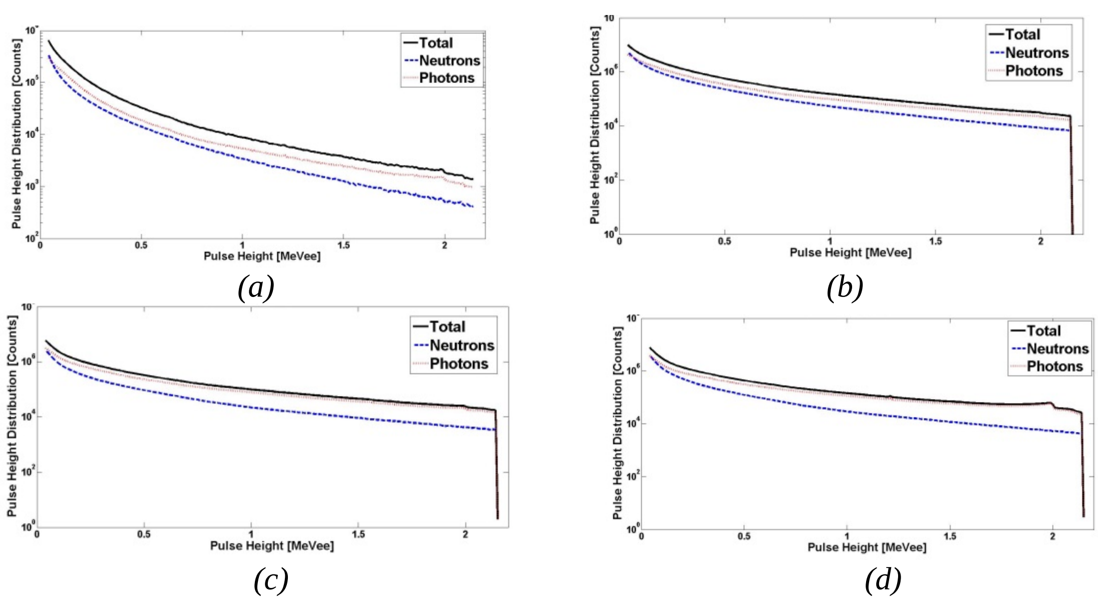


Figure 16*:* Pulse height distributions (total, neutron, photon) for all 4 detectors;(a)active, HEU voided, ^{252}Cf source; (b) active, HEU bare, ^{252}Cf source ; (c) active, HEU + 2cm poly, ^{252}Cf source; (d) active, HEU + 4cm poly, ^{252}Cf source

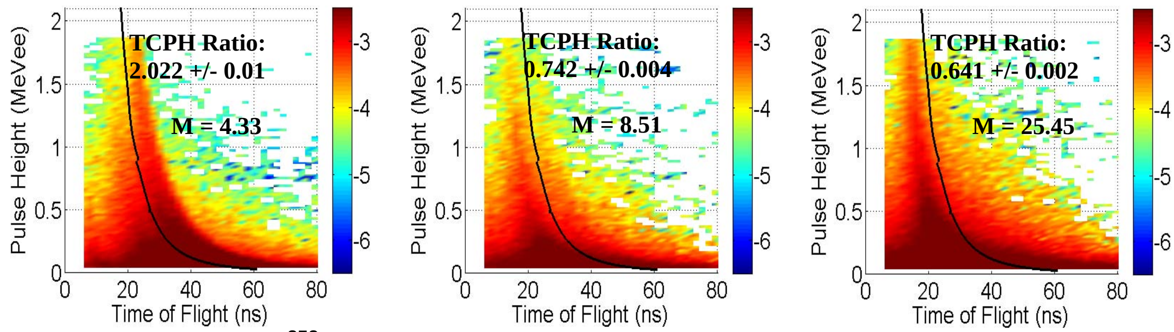


Figure 17: TCPH for ^{252}Cf source from left to right: bare, 2 cm and 4 cm configurations.

The reason for the differences in the ^{252}Cf and Am-Li cases can be best demonstrated by simulating the impact that the two sources have of TCPH distributions without the target HEU present. The resulting TCPH distributions are shown in Figure 18. The ^{252}Cf source produces many correlated photons and neutrons from spontaneous fission, while the Am-Li source gives only one correlated gamma-neutron pair per neutron emitted. Furthermore, Am-Li photons are of very low energy, which are below the usual operating detector threshold in actual measurements. With a threshold of 0.03 MeVee, as in all other cases shown, no correlated photon-neutron pairs would be present in the TCPH distribution. It is worth noting that the TTAL shown is computed for the 50 cm HEU detector distance, whereas the ^{252}Cf detector distance is 75 cm. Therefore much of the ^{252}Cf contribution to the TCPH plot occurs beyond the TTAL.

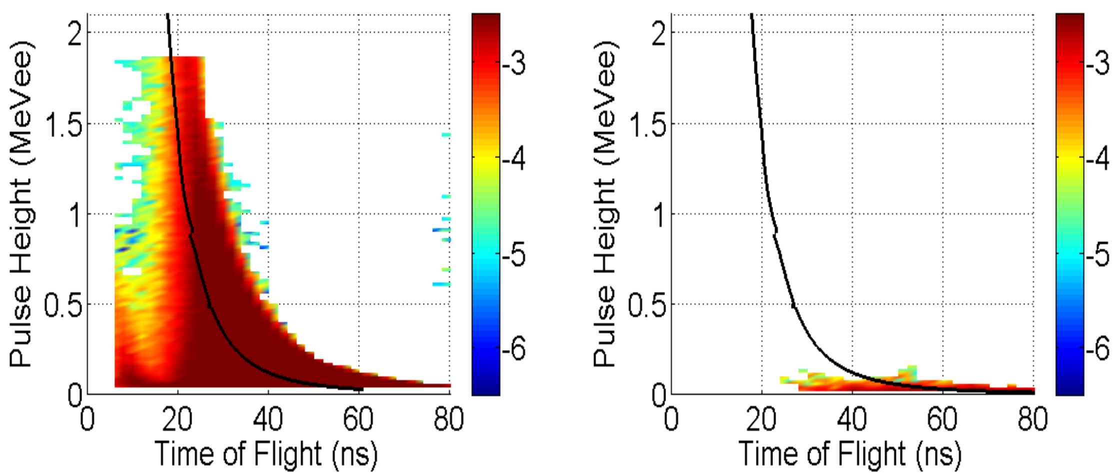


Figure 18: TCPH distributions for Cf-252 (left) and Am-Li (right) with no HEU present.

The correlation between TCPH ratio and multiplication for the Am-Li, D-D, and ^{238}U SF source cases with 25.4 kg sphere of HEU are shown in Figure 19. The ^{252}Cf case is not shown because ^{252}Cf dominates the plot. However, if the TCPH background from ^{252}Cf were known ahead of time, one could envisage subtracting this as a background. The correlation between TCPH ratio and multiplication for the 12.7 kg hemisphere of HEU configurations is shown in Figure 20. A positive relationship is observed between multiplication and TCPH ratio as with the sphere configurations.

The data for all configurations are summarized in Table 3 and Table 4. All TCPH ratios for the hemisphere configurations were higher than the bare full sphere configuration, despite the latter having a higher multiplication. This discrepancy between the full and half sphere configurations is likely related to the difference in geometry. The way in which the polyethylene acts as a shield and neutron reflector is not constant between these two configurations. This most likely contributes to the difference in the absolute TCPH ratio; however the important trend of increasing ratio with multiplication is present in both configurations. While it has been established that the TCPH technique is well suited for detecting the presence of a multiplying source, like SNM, being able to identify the mass of the SNM would be complicated by the myriad imaginable moderator plus SNM configurations without an assumption on the symmetry of the source (i.e. spherical).

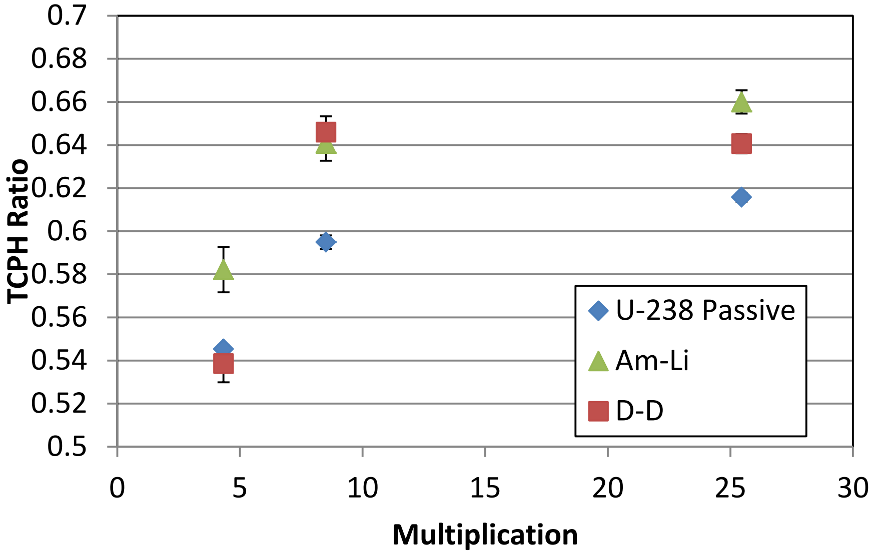


Figure 19*:* TCPH ratio and multiplication correlations for two source configurations for the 25.4 kg sphere of HEU.

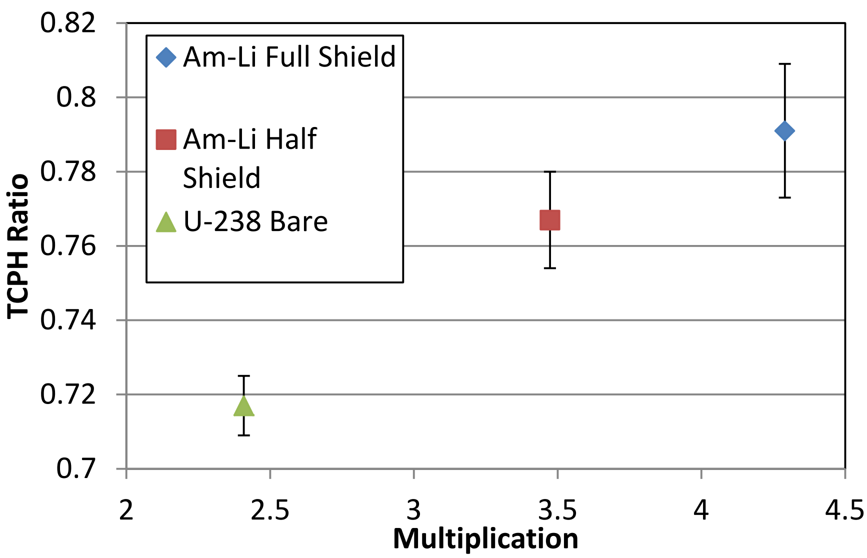


Figure 20: TCPH ratio and multiplication correlations for the three moderator configurations for the 12.7 kg hemisphere of HEU.

Table 3: TCPH ratio and k_{eff} for the 25.4 kg of HEU sphere configurations.

Source	Moderator Configuration		
	Bare	2 cm	4 cm
U-238 SF	0.545 +/- 0.001	0.595 +/- 0.003	0.616 +/- 0.002
Cf-252	2.022 +/- 0.010	0.742 +/- 0.004	0.641 +/- 0.002
Am-Li	0.582 +/- 0.011	0.641 +/- 0.008	0.660 +/- 0.005
D-D	0.539 +/- 0.009	0.646 +/- 0.007	0.641 +/- 0.005
k_{eff}	0.7689	0.8825	0.9607

Table 4: TCPH ratio and k_{eff} for the 12.7 kg of HEU hemisphere configurations.

Source	Moderator Configuration		
	Bare	Half-Moderated (4cm)	Fully-Moderated(4cm)
Am-Li	N/A	N/A	0.791 +/- 0.018
Am-Li	N/A	0.767 +/- 0.013	N/A
U-238 SF	0.717 +/- 0.008	N/A	N/A
k_eff	0.5848	0.7121	0.7669

4. ACTIVE MEASUREMENTS

A series of laboratory measurements were conducted to validate some of the general trends found in the Monte Carlo simulations as described in the previous section. Because no HEU was available, these measurements were primarily conducted using the bare interrogating source with the exception of the D-T interrogation of DU. In all cases, no multiplication is expected to be present.

4.1. ^{252}Cf Interrogation Source

As indicated in Sections 3.1 and 3.3 and shown in Figure 6, Figure 17, and Figure 18 we should expect that ^{252}Cf, being a fission source with high gamma-neutron correlation, will be strongly imprinted on the TCPH distribution. Indeed, ^{252}Cf measurements were the first validating tests of the TCPH method as a null (non-multiplying) case. Figure 21 shows the TCPH distribution obtained in a laboratory measurement with the source at 50 cm stand-off. Many gamma-neutron correlations are seen in the region below the TTAL as expected.

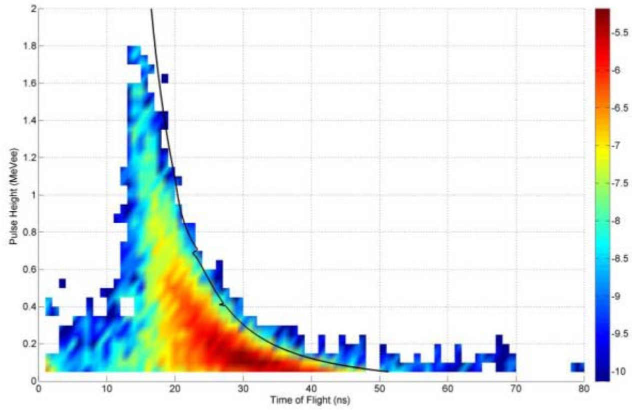


Figure 21: TCPH measurement of a ^{252}Cf fission source at 50 cm stand-off.

4.2. Am-Li Interrogation Source

Measurements were conducted using an Am-Li neutron source (~1e6 n/s) in a configuration identical to that described in Section 3.3. Four liquid scintillator detectors were oriented in an arc with 50 cm radius with the Am-Li at a distance of 75 cm. The resulting TCPH distribution is shown in Figure 22 and just like the predictions seen in Figure 18 (right), very few gamma-neutron correlations are detected. A dwell time of 5 hours was chosen to match the simulation time and the few events that are registered in the TCPH plot are consistent with accidental correlations from the high single uncorrelated event rates measured in the detectors.

This result corroborates the conclusion that Am-Li is an ideal candidate source for the active interrogation of SNM.

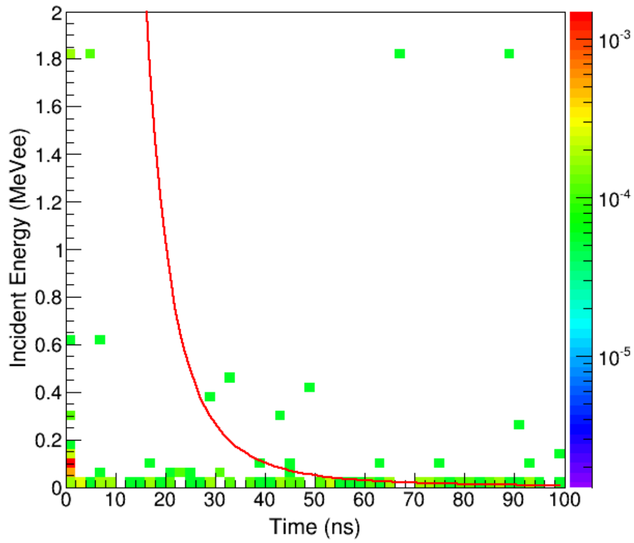


Figure 22: TCPH measurement of a bare AmLi source. The dwell time is the equivalent of the simulation shown in Figure 18 (right).

4.3. D-T Neutron Generator Interrogation Source

Though the Monte Carlos described in Section 3.3 simulated a D-D (2.45 MeV) neutron generator, for these measurements a D-T (14.1 MeV) neutron generator was employed. This was done in order to demonstrate a positive case of active induction of fission in a material. HEU was not available in our laboratory, but we were able to acquire ~5 kg of depleted Uranium (DU) which, being primarily ^{238}U requires an incident neutron energy greater than a few MeV for efficient fission induction rather than the primarily ^{235}U in HEU which more efficiently fissions with the introduction of low energy neutrons.

Our measurement of the interrogation of DU with a D-T source is therefore a surrogate for the interrogation of HEU with a D-D source. In many ways this is a more challenging environment to detect induced fission in that the D-T source was driven at a neutron emission rate two orders of magnitude higher than in the Monte Carlo simulations of the D-D source causing a high rate of accidental (uncorrelated) coincidences. In addition, the higher energy neutrons are much more efficiently detected in the liquid scintillator cells, thus the interrogation source is more likely to register on the TCPH plot.

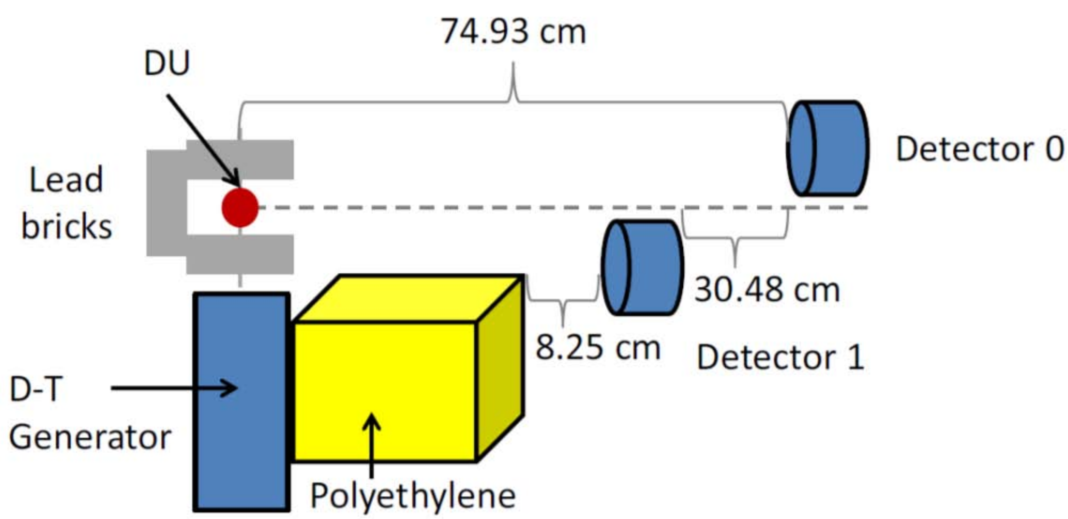


Figure 23: Diagram of the active interrogation configuration used to measure the stimulation of DU.

Figure 23 is an illustration of the experimental configuration used in these measurements. Two 12.7 cm diameter by 5.04 cm thick liquid scintillator detectors were shielded from the direct line of site of the D-T generator by a 30.5 cm thick block of high density polyethylene. Five kilograms of DU was held in close proximity to the D-T generator and measurements were made with and without a two inch thick box of lead surrounding the DU.

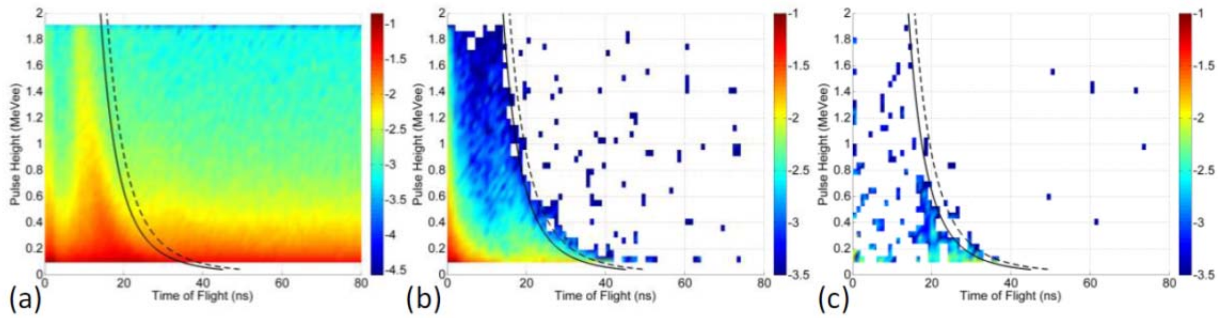


Figure 24: TCPH measurements during the active stimulation of DU, a) the raw time correlated spectrum, b) the spectrum with the accidental background subtracted, c) the spectrum with correlations caused by the D-T generator removed, showing the remaining induced fission events.

Figure 24 shows the measured TCPH distributions a) including accidental events and the D-T generator, b) with the accidental contribution removed, but with the D-T contribution remaining, and c) with both accidentals and D-T removed. What is left is a clear excess of correlated fission events indicating the detection of fissionable material. It should be noted that these measurements represent only several hours of dwell time whereas it would require years to register the same number of correlated events from DU without interrogation.

Finally, Figure 25 shows the same background and D-T subtracted interrogation of DU with the presence of 2 inches of lead shielding on all sides. A factor of three higher correlated gamma-neutron rate is indicated due to the reflection of high energy neutrons back into the DU. It should be noted that two inches of lead would make the detection of this quantity of DU extremely difficult using passive gamma spectroscopy and/or neutron counting.

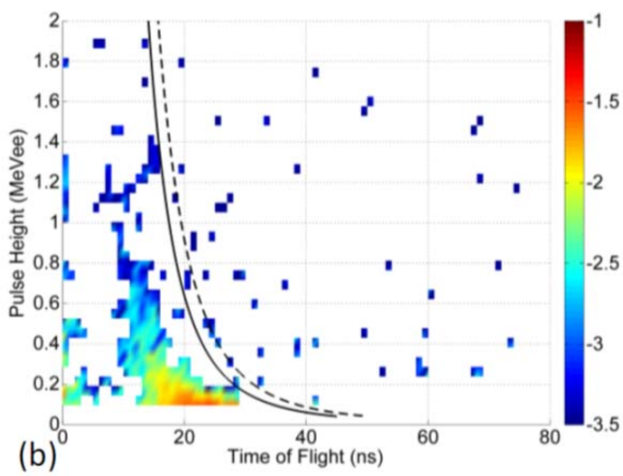


Figure 25: TCPH measurement of DU with a 2 inch thick lead shield indicating an enhancement of induced fission.

5. CONCLUSIONS

The capability of detecting and characterizing a multiplying source with TCPH distributions has been demonstrated. A positive trend in the TCPH ratio with increasing multiplication has been predicted and validated in past passive measurements of configurations of WGPu. The TCPH method, however, is shown to have limitations when applied toward passive measurements of HEU. Simulations of passive measurements indicate that the measurement times required are impractical for real world applications. For this reason active stimulation of fission using an external neutron source was investigated.

Three active sources were explored: ^{252}Cf, D-D and Am-Li. The comparisons proved the importance of using a source that does not produce measureable temporally correlated photons and neutrons that will contribute to a TCPH distribution. It was shown both in simulations a laboratory experiments that a ^{252}Cf fission source is a poor choice of interrogating neutrons. The large number of high energy correlated gammas and neutrons can cause an overwhelming signature in a TCPH distribution.

However, both simulations and laboratory measurements indicate that an Am-Li neutron source is an ideal candidate as a radiological interrogation source. Its advantages include: the emission of low energy neutrons that fall below the threshold for induced fission for 238U while still inducing 235U and the low energy of correlated gamma-rays allowing it to remain all but invisible in TCPH plots. Thus the use of Am-Li as an external stimulating source greatly reduces measurement times, while leaving TCPH distribution populated with photon-neutron correlations only from any HEU that may be present.

The use of a D-D generator improves the required measurement time even further, theoretically shrinking it to an order of several minutes. Because any photons produced by the D-D generator are uncorrelated with the neutrons, the photon-neutron correlations that would appear on a TCPH distribution will be accidental only and can be removed as was demonstrated with the much stronger D-T laboratory measurements.

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