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Detection and Characterization of Shielded Highly Enriched Uranium Under Active Interrogation Through Time Correlated Fission Events

Mateusz Monterial, Peter Marleau, and Sara A. Pozzi

***Abstract*—The time-correlated pulse-height (TCPH) distribution can be used to differentiate between multiplying (e.g ^{235}U, ^{239}Pu) and non-multiplying (e.g Am-Li, ^{252}Cf) sources. In the past, this approach proved effective at characterizing the multiplication of alpha phase plutonium metal through a passive measurement. Recently, Sandia National Laboratories has completed a measurement campaign with its new Correlated Radiation Signature (CoRS) system involving active interrogation of highly enriched uranium (HEU) with an Am-Li source. An additional obstacle was introduced to the measurement configuration by shielding the HEU with depleted uranium (DU). Simulation results have proven Am-Li source to be a suitable interrogating source because of its relatively low-energy neutron spectrum. The TCPH distribution was successfully used to determine the presence of a multiplying medium inside DU shells. The correlation between multiplication and an empirical parameters broke down for externally driven configurations, but in all cases the presence of a multiplying source was detected.**

I. Introduction

THE traditional nondestructive assay of bulk highly enriched uranium (HEU) has relied on neutron coincidence counting with ^{3}He based detectors. For analysis presented in this work we swapped the increasingly scarce ^{3}He tubes for fast, pulse shape discrimination (PSD) capable, organic scintillators. This has a few important implications that allows us to measure signatures unavailable to traditional neutron detectors. First, we take advantage of the PSD capability to correlate both gammas and neutrons emitted from our source. Second, the nanosecond time resolution allows us to focus on relatively narrow (100-200 ns) time correlated windows. Third, the light yield resulting from a recoiling proton can be used to estimate the energy deposited by the incident neutron.

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Combined, these capabilities allow us to measure the Time Correlated Pulse-Height (TCPH) distribution. It is a bivariate histogram of the times between correlated neutrons and gammas and the incident neutron energy. TCPH was used in the past to characterize plutonium with different moderator/reflector configurations under a passive measurement [1]. TCPH measurements using active interrogation of HEU were demonstrated in simulations [2]. In this work we present the results from a measurement campaign, carried out by a team from Sandia National Laboratories, of various configurations of actively interrogated HEU. For added complexity some configurations included depleted uranium (DU) shielding, which present a challenge for passive gamma spectroscopic methods. Specifically we sought to determine if it was possible to detect the presence of a multiplying material and furthermore to characterize its pertinent aspects (i.e. multiplication, moderator/reflector configuration).

II. Setup

A. Equipment and Target Materials

The detection system consists of eight 2" stilbene crystals arranged cylindrical pattern much like the rotating barrel of a revolver. The pulses were digitized by an 8 channel, 14-bit, 500 MS/s CAEN DT5730 desktop digitizer operated in asynchronous acquisition mode. The materials of interest were the Training Assembly Criticality Shells (TACS), a collection of four pairs of hemispherical shells that together make up 23 kg of HEU (93.15%). Four identical pairs of DU hemishells were also available as a stand in for the HEU shells, for some configurations we replaced the outer most HEU shell with DU. In addition, an internal shell of Lucite was always present, and an external reflector of the same material was used in some configurations. Out of the total of five configurations, illustrated in Fig. 1, three were externally driven and the remainder had the interrogating source placed in the inner cavity. For the externally driven cases the detector was placed on the opposite side of the TACS with respect to the interrogating source.

Am-Li was chosen as the interrogating source because of its soft neutron spectrum which in principle should be new the operating threshold of the detector. The coincident gamma is also low energy, making Am-Li invisible on a TCPH distribution. Furthermore, the soft spectrum preferentially stimulated fission in the HEU as opposed to the DU, due to the exceedingly small fission cross-section of ^{238}U below 1 MeV. In practice, some Am-Li sources are contaminated with Be which in turn

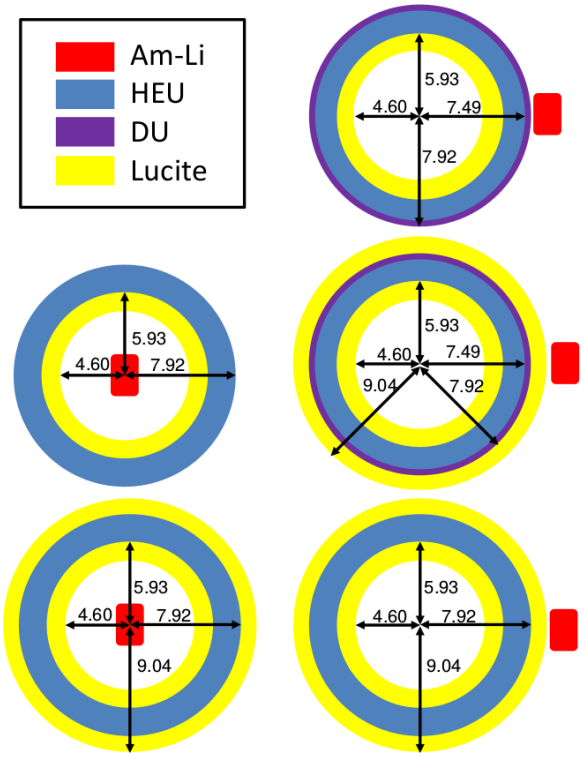

Fig. 1: The two internally (left) and externally (right) drive configurations. All dimensions are expressed in centimeters.

make the effective neutron spectrum much harder and make it possible to pick up uncorrelated accidental neutrons from the interrogating source.

B. Calibration

Gamma spectra from organic scintillators lack photopeaks, making exact conversion between digital values into electron recoil energy a dubious proposition at best. In addition the energy resolution, which in itself is difficult to measure, is typically quite poor for this class of low-density hydrogenous detectors. Therefore, the ability to precisely determine the location of a substantially blurred Compton edge, more of an art than exact science, is a venture fraught with peril and often given only tertiary consideration and effort. Nevertheless, this information is needed to back out incident neutron energy, therefore it was necessary to give calibration more than just a glancing look.

Instead of estimating the location of the Compton edges, we derived the parameters of interest by matching simulation and measured spectra. A Na-22 source, which provides two distinct gamma-ray energies, was measured for calibration. In order to match simulated and measured spectra the former had to be smeared by an energy dependent Gaussian function, and the latter converted from pulse integrals (V·ns) to energy (MeV).

MCNP6 was used to gather the necessary simulated spectrum with a pulse height distribution (F8) tally. The simulated spectra, s_i, a histogram of i number of 10 keV energy bins, E_i, was smeared by energy dependent Gaussians, f_i. The width of each Gaussian is dependent on the energy bin by

$$\sigma = a\sqrt{E_i} + b \tag{1}$$

The relation in (1) comes from the fact that the resolution is proportional to the number of charge carriers the production of which is a Poisson process dependent on the initial deposited energy. The actual "smearing" operation is a multiplication of Gaussians with the simulated spectrum

$$\begin{bmatrix} h_1 \\ h_2 \\ \vdots \\ h_i \end{bmatrix} = \begin{bmatrix} f_1 & f_2 & f_3 \dots f_i \\ f_1 & f_2 & f_3 \dots f_i \\ \vdots & \vdots & \vdots \end{bmatrix} \begin{bmatrix} s_1 \\ s_2 \\ \vdots \\ s_i \end{bmatrix} \tag{2}$$

where h_i is the resulting simulated spectra with effects of energy resolution.

The measured spectra, on the other hand, was converted into energy space by a quadratic relation:

$$E = cI^2 + dI \tag{3}$$

where E is the electron recoil energy and I (V·ns) the pulse integral. The constant term was omitted since its accounted for by proper baseline subtraction. We expect the detector to have a linear response at this energy range, however a quadratic term was necessary to make the Compton edges align. We suspect that the electronics are introducing some level on non-linear behavior at higher energies. The fifth and final parameters involved measured background, multiplied by a constant factor, which was subtracted from the measured calibration spectrum.

Together with the constants in (1) and (3) this problem made for a system with 5 unknown variables which we optimized using the Nelder-Mead method. The loss function was cast as the difference between the measured and simulated spectra, weighted by a vector that increased the relative importance of regions around the Compton edges. An exampled of optimized measured and simulated spectra for one of the channels in shown in Fig. 2.

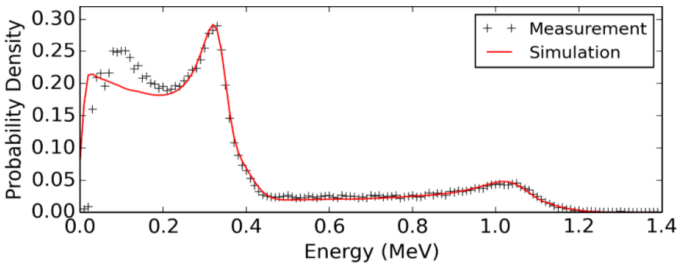

Fig. 2: Best fit match of the measured and simulated spectra for each detector. The simulation omitted surrounding materials which results in a lack of a backscatter peak present in the simulation.

III. Methods

A. Fission Chains and TCPH

Before laying out how we use TCPH to gather relevant information, we will set out the underlying physics that make it

useful. As stated previously, we are measuring the distribution of times between correlated gammas and neutrons, taking note of the light yield from the neutron interaction. Gammas, thanks to Einstein, having the same speed in a particular medium provide a clean clock start for the event that gave birth to it. The neutron on the other hand is betrayed by its energy, which gives an indication of the travel time from the originating event. Any discrepancy between this time and the correlated time between gamma and neutron indicates that the two did not originate from the same event. The distinction between originating events of correlated neutrons and gammas is central to differentiating non-multiplying and multiplying sources.

For non-multiplying sources, the correlated neutron and gamma should originate from the same event. In ^{252}Cf the event is spontaneous fission, and for (α,n) sources with a beryllium target the gamma is born from the deexcitation of ^{12}C. However, for fissile materials it is possible to correlate neutrons with gammas from different fission events along a fission chain. Therefore, it is possible for neutrons to appear later in time than predicted by the light output. On a TCPH distribution we can demarcate the expected neutron energy given the time difference from the gamma, and call this the Theoretical Time of Arrival Line (TTAL). For a majority of interaction an incident neutron will deposit a fraction of its energy onto the detector, therefore for non-multiplying sources the TCPH distribution should fill below the aforementioned TTAL. Correlating neutrons and gammas from different fission events in a chain will fill the space above that line and give a clear indication of a multiplying source.

B. Neutron Light Output and Theoretical Time of Arrival Line

To construct a TCPH distribution we require the time between correlated gammas and neutrons, possible through PSD, and energy deposited by the incident neutron. The latter calculation necessitates converting the light output from a neutron interaction into deposited energy. To this end we invert a commonly used exponential relation of deposited energy, E_p, and light output, L:

$$E_p = \frac{L}{(a-b)} + \frac{1}{c}\,\mathrm{LambertW}\left(-\frac{bc}{a-b}e^{-\frac{cL}{a-b}}\right) \quad (4)$$

where constants a, b and c are 0.601, 1.861 and 0.288, respectively [3].

The TTAL is calculated from the deposited energy, E_p, the distance to the measured object, d, and the gamma travel time:

$$t = d\left(\sqrt{\frac{939.57}{2E_pc^2}} + \frac{1}{c}\right). \quad (5)$$

An example of a TCPH distribution with a TTAL overlayed on top is shown in Fig. 3. We expect that the distribution will be smeared over the TTAL for multiplying sources, therefore the ratio of the total counts above and below the line should be related to the overall level of multiplication:

$$TTAL\ Ratio = \frac{Integrated\ Counts\ Above}{Integrated\ Counts\ Below} \quad (6)$$

Ideally this ratio would correlate with multiplication, but in reality there is a dependence on interrogating source geometry and the reflector/moderator material.

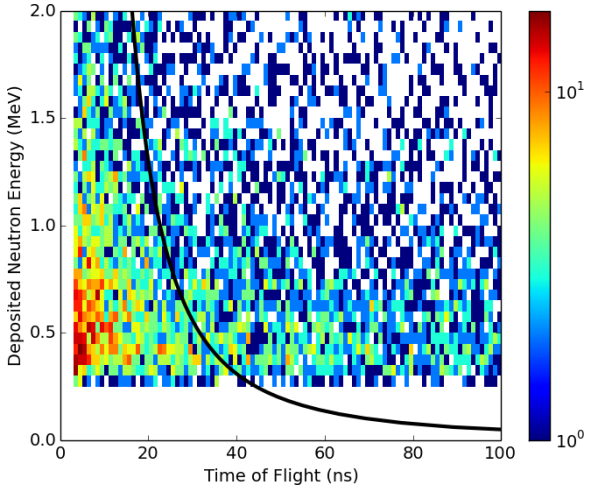

Fig. 3: TCPH distribution gathered from the internally interrogated configuration of HEU with 1 in of Lucite around. The black line represent the theoretical time of arrival of the neutron based on the deposited energy.

IV. Results and Discussion

Although TCPH distribution as shown in Fig. 3 is adequate for demonstrating the principles of the technique, it is difficult to discern subtle differences between different configurations. This is made especially difficult by the relatively low counts achieved for the externally driven configurations. To better illustrate the differences among configurations we first shift all the counts by the difference between time 0 and TTAL. Essentially this amounts to shifting all counts by the time calculated from 5. In addition we project all the counts onto the time axis, transforming the bivariate TCPH distribution into a more familiar time correlation distribution. The resulting plots for all configurations are shown in Fig. 4.

The first thing that jumps out is that the non-multiplying Am-Li source is quite different from the multiplying configurations. The seemingly random amplitude in counts is the result of background subtraction of an underlying flat distribution indicative of chance correlations. The amplitude of this flat distribution is a function of the overall source strength. The configurations with the stimulated HEU show a gentle decreasing slope that extends from time 0 to later times. The amplitude difference between the externally and internally driven cases is proportional to overall count rate which varies because of the greater number of induced fissions in the HEU from the internally driven configurations. The slope on the other hand is indicative of the average time between fissions and the length of fission chains. In particular the cases with additional Lucite moderator have shallower slopes, which is the expected effect from a hydrogenous reflector.

The multiplication among configurations is also different, and here we attempted to characterize it through the TTAL Ratio from (6). This is calculation equivalent to taking the ratio of counts before and after time 0 from the distributions shown in Fig. 4. The resulting ratios as a function of multiplication are shown in Fig. 5. With the exception of the DU shielded cases a positive correlation exists between TTAL ratio and

multiplication. However, this correlation is not equivalent between the externally and internally driven cases, which underscores the difficulty of having an absolute measurement of multiplication with this method. A more robust determination of this parameter may be possible if the source geometry and reflector configurations were better constrained, perhaps through entirely different method and detector system.

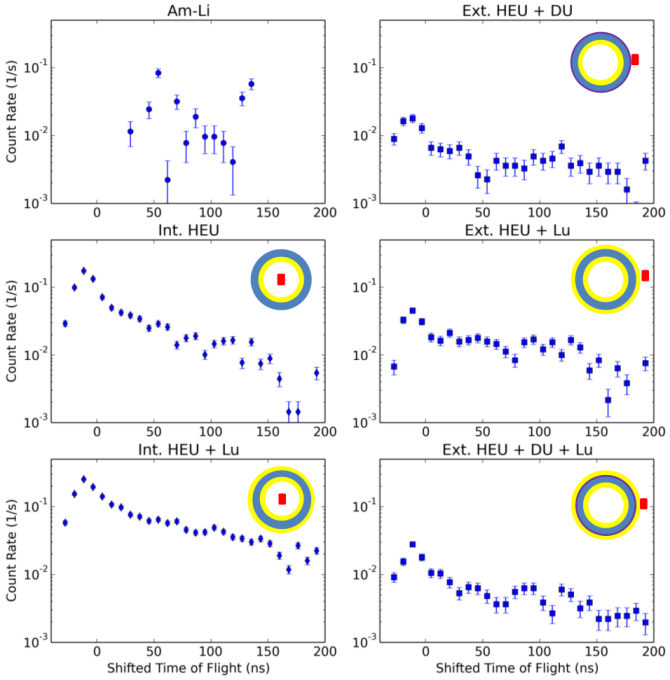

Fig. 4: TCPH distributions projected on the time axis after shifting each gamma-neutron correlation time the TTAL from (5).

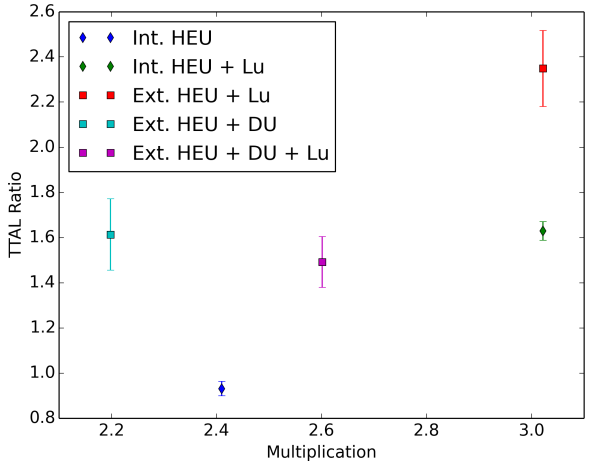

Fig. 5: Ratio of the correlations above and below TTAL as a function of multiplication.

V. Conclusions

We demonstrated the viability of the TCPH distribution in characterizing fissile material under active interrogation. Determination of a multiplying source was made readily apparent, but characterization of relevant parameters was limited due to low statistics and different interrogating source positions. Interrogation of a target from the opposite side of the detection system is the most difficult scenario for TCPH analysis, since fissions will be induced on the far side of the target from the detectors point of view. However, it was still enough to detect the presence of a multiplying material, even with DU shielding.

Furthermore, TCPH reliance on neutrons made it possible to determine presence of HEU under DU shielding, a configuration which presents a challenge for traditional gamma spec methods. When the count rate and dwell time allowed for significant statistics, as with the internally driven cases, then the relative level of multiplication could be characterized. But even in the externally driven cases the presence of a multiplying source was determined.

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