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Stilbene Scintillator Detector Array and Data-Acquisition System for Active Interrogation

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Stilbene Scintillator Detector Array and Data-Acquisition System for Active Interrogation

An NA-22 Project Report

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August 2018

Abstract

A large area Stilbene crystal detector array has been assembled and calibrated for use in active interrogation measurements. The array consists of 10 high-efficiency 4"x2" stilbene cells. The cells were grown by Inrad Optics using LLNL developed technology. The array can be used to detect multiple signatures of SNM items, both actively and passively. Prompt and delayed gamma rays as well as prompt and delay neutrons can be detected and counted separately. The array is coupled to the Mesytec MDPP-16 VME-based digitizer module for data acquisition. This module performs on-board pulse-shape discrimination of signals from the Stilbene crystal detectors. This data-acquisition module allows separation of neutron and gamma-ray events with a low demand on data transfer rates. In addition, the high-resolution timing (<100 ps) is suitable for correlated gamma-ray and neutron multiplicity counting. This report describes the construction and calibration of the system. We also discuss applications of the system as well as how the system is currently being utilized by end users.

Introduction

Stilbene crystal detectors offer a promising approach for nuclear material assay applications because of several advantages compared to He-3 detectors and Xylene- or mineral-oil based liquid-scintillator detectors. Stilbene is an optimal scintillator detector because it has the highest organic scintillation efficiency with much higher light yield than plastics or liquids. It has excellent pulse-shape discrimination (PSD) which allows for excellent neutron and gamma-ray discrimination figure-of-merit (FOM), which in turn implies high neutron-counting detection efficiency. In addition, unlike liquid scintillators, stilbene crystals are not a flammable or combustible hazard, simplifying detector shipment and facility acceptance.

We have constructed a large array of Stilbene detectors for active and passive nuclear material assay. The array can be used to detect multiple signatures of SNM items, both actively and passively. Prompt and delayed gamma rays as well as prompt and delay neutrons can be detected and counted separately. The array consists of 10 4”x2” Inrad Optics Stilbene crystals.

He-3 based detectors reveal neutron behavior in the 10’s to 100’s of micro-s due to the need of moderating the neutrons to increase detection efficiency. Fast-neutron scintillators such as Stilbene or liquid scintillators reveal neutron behavior in the sub nano-s to 10’s or 100’s of micro-s[1,2]. This is the time scale of individual fission chains in the material and allows the least obstructed view of what is happening in the material. It has also been shown that fast neutron scintillators can show in time an effect called neutron restart where leaving a chain can be reflected back into the system and create new fissions[3].

Figure 1 shows a conceptualized picture of a fission chain. The neutrons can either initiate new fissions, be absorbed, or exit the system and be detected or not detected by a neutron detector. Figure 2 shows a graphical representation of the occurrence of events in time.

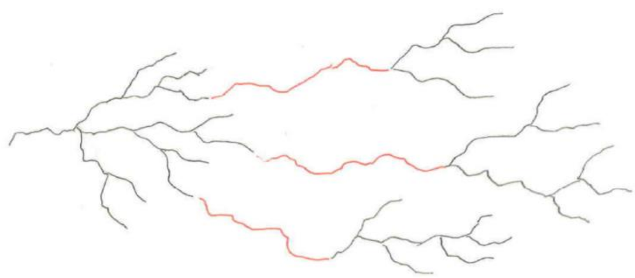

Figure 1 Representation of a Fission Chain with three neutrons restarting new chains after one has ended

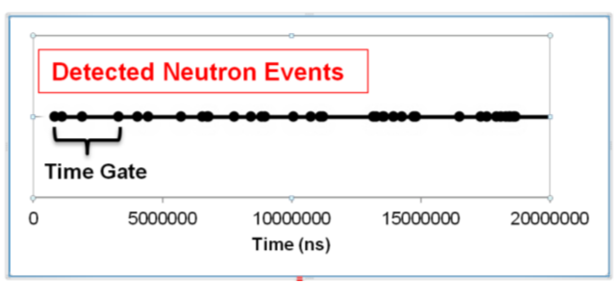

Figure 2 Example distribution of detected events as a function of time.

Figure 3 shows scintillator data from a multiplying HEU system. The top plot is the cumulative counts versus time, i.e. the time-evolution of total counts. The lower plot is the time between

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[1] M. K. Prasad And N. J. Snyderman, “Statistical Theory Of Fission Chains And Generalized Poisson Neutron Counting Distributions,” Nucl. Sci. Eng., 172, 300 (2012);

[2] K. S. Kim, L. F. Nakae, M. K. Prasad, N. J. Snyderman, and J. M. Verbeke, “Time Evolving Fission Chain Theory and Fast Neutron and Gamma-Ray Counting Distributions,” Nucl. Sci. Eng., 181, 1 (2015).

[3] K. Kim, L. Nakae, M. Prasad, N. Snyderman, and J. Verbeke, “Fission Chain Restart Theory”,

events (time-till-next) versus time., i.e. the time-evolution of delta-time. This is called the fission waterfall plot because individual fission chains are visible as vertical streaks. Figure 4 shows a waterfall plot for another HEU system (top) as well as the projection of the waterfall plot on to the delta-time axis (bottom). This lower plot is another visual representation of the fission timing information unique to scintillator detectors. The events in the distribution on the right side are the singles, doubles, triples, etc., that would also be seen by He-3 based detectors. The events in the distribution on the left side are the nano-s time scale fission-chain events not visible when using He-3 detectors.

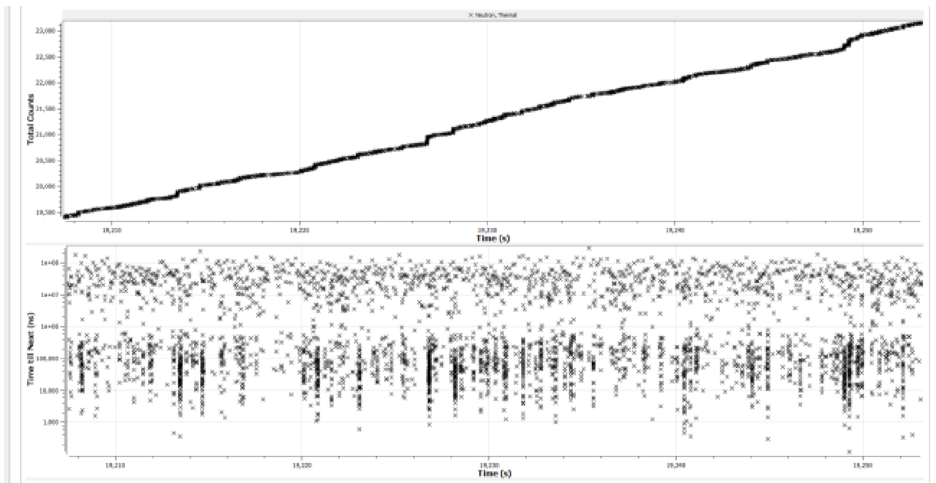


Figure 3 Time-evolution plots of multiplying HEU system. The upper plot is cumulative counts vs. measurement time. The lower plot is adjacent events delta-time versus measurement time and is called the waterfall plot. The downward streaks in the waterfall plot represent neutron fission chains with nano-s time resolution

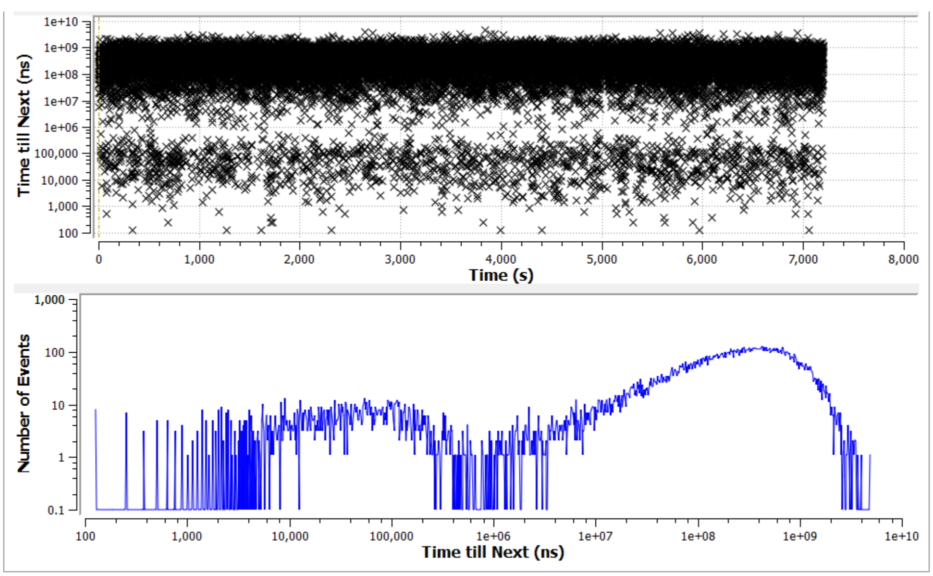


Figure 4 Waterfall plot of Time-until-next vs. Time (top) and projection (bottom) for HEU in moderator

The data acquisition system then analyzes the time distribution of events using time bins of varying width to determine the probability of obtaining 0, 1, 2, 3…etc. neutrons in each bin. This allows construction of the neutron count distributions. An example scintillator count distribution shown in Figure 5. This count distribution is for the LANL BeRP ball is which is an unclassified 4.5kg α-phase sphere of plutonium. It is 93.7 wt % ^{239}Pu, 3.0" dia., machined in 1980, and clad in 304 stainless steel. Count distributions are then used to compute the Feynman doubles, triples, quads, etc. as shown in Figure 6. The asymptotic values of these plots are used in the neutron multiplicity counting formalism to compute system efficiency, multiplication, alpha ratio, and source mass.

This window on fissioning materials is made possible by combining scintillator detectors such as Stilbene with new modular electronics that can time-stamp events with sub-nano-s timing precision. This is a relatively new use of this technology and the applications range from nuclear material accountability, arms control verification and nuclear non-proliferation to emergency response diagnostics.

Below we describe the construction and calibration of the 10-cell stilbene detector array using gamma-ray check sources ^{22}Na, ^{137}Cs, ^{60}Co, ^{232}U, and D-D (2.45 MeV) and D-T (14.1 MeV) neutron generators. We also discuss current and potential application of the Stilbene array.

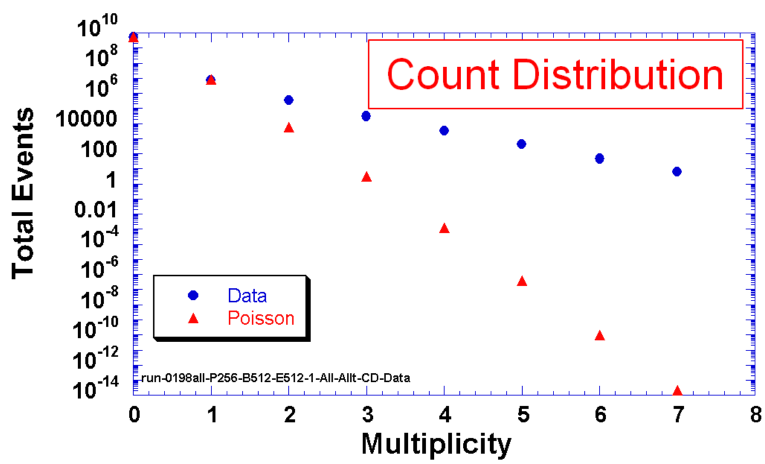


Figure 5 Example neutron count distribution for the 512ns time bin from a liquid scintillator measurement of the bare BeRP ball.

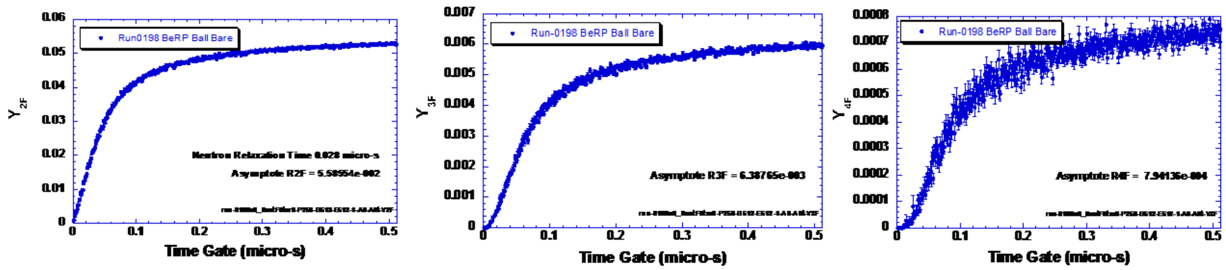
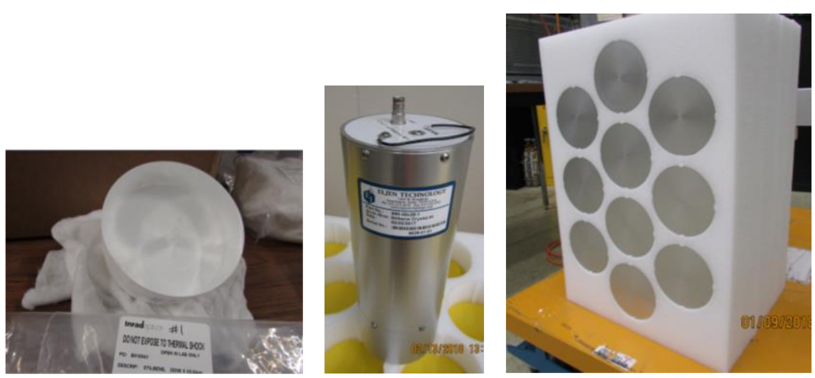


Figure 6 Plot of doubles, triples, and quads for BeRP ball

Hardware Description

The Stilbene detector array consists of 10 high-efficiency 4” x 2” stilbene crystal cells. The cells were grown by Inrad Optics, Northvale, NJ, using LLNL developed technology[4]. The crystals were then assembled with a photomultiplier tube and housing by Eljen Technology, Sweetwater, TX. Procurement of the Stilbene detectors was through the NA-22 project End-to-End Warhead Monitoring. Figure 7 shows photographs of an individual Stilbene crystal, an assembled detector, and the full 10-cell array.

The ideal response of a liquid scintillator detector to monoenergetic neutrons is therefore a step function. Figure 8 shows the ideal scintillator spectrum from 10 MeV neutrons. In a real detector the actual response spectrum has the shape shown in Figure 9. In addition to a neutron rate, this spectrum contains neutron energy information that can be used in analysis. The shape from a gamma-ray interaction is similar. Note that this type of detector does not produce a photopeak from gamma rays like a NaI or HPGe detector would. The peak area visible with a gamma-ray source is actually the Compton edge from the Compton scattering of gamma rays by electrons.



[4] Natalia Zaitseva, Andrew Glenn, Leslie Carman, H. Paul Martinez, Robert Hatarik, Helmut Klapper, Stephen Payne, "Scintillation properties of solution-grown trans-stilbene single crystals", Nuclear Instruments and Methods in Physics Research Section A, Volume 789, 21 July 2015, Pages 8-15

Figure 7 Photographs individual Stilbene crystal (left), assembled detector (center) and 10-cell array in rigid foam support (right)

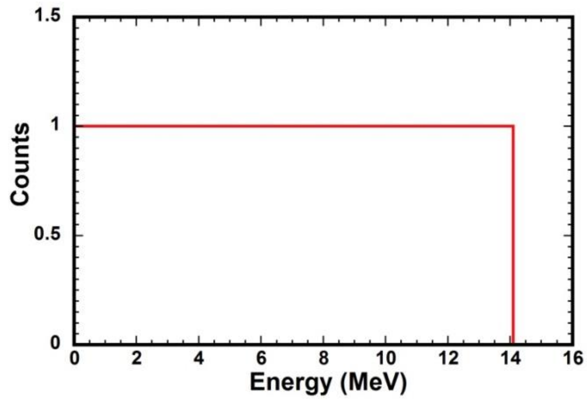


Figure 8 Idealized response of scintillator detector to 14.1 MeV monoenergetic neutrons

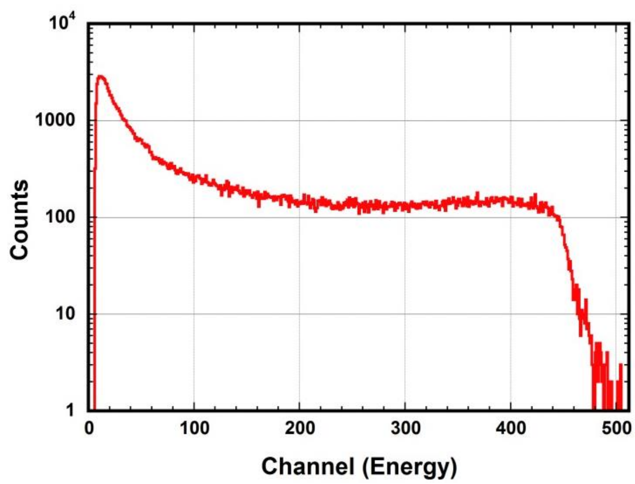


Figure 9 Example 14.1 MeV D-T spectrum from an EJ-301 liquid scintillator detector

Signals from the detector array are sent to a Mesytec MDPP-16 VME-based digitizer module for data acquisition. This is a newly developed module and was selected for use after testing indicated high-quality data was possible along with ease of use of the included data-acquisition software MVME. The module performs on-board pulse-shape discrimination (PSD) of signals from the scintillator detectors to provide separation of neutron and gamma-ray events[5]. This data-acquisition module has a low demand on data transfer rates since the PSD is performed on the board. In addition, the <100 ps high-resolution timing is suitable for correlated gamma-ray and neutron multiplicity counting. A photograph of the module and block diagram are shown in

[5] Andreas Ruben, Peter C. Bender, Partha Chowdhury, Phillip L. Kerr, Florian Lüke, Gregor Montermann, Andrew M. Rogers, Robert Schneider, "A New, Versatile, High-performance Digital Pulse Processor with Application to Neutron/Gamma-Ray Pulse-Shape Discrimination in Scintillator Detectors", NIMA_SORMA XVII, July, 2018.

Figure 10. The instrument also has excellent energy resolution enabling energy spectroscopy measurements with a wide range of gamma-ray detectors including High-Purity Ge detectors.

All measurements were performed using a ready-to run, platform independent and open-source DAQ software package MVME. This software includes hardware configuration and run control as well as online data monitoring. A screen of the MVME interface and an operational block diagram are shown in Figure 11. Most of the hardware settings programmed into the matching VME registers are pre-configured, which allows a short learning curve. To achieve high data rates, the MVME software takes advantage of the list sequencer mode of the VM-USB and SIS3153 controller.

Online data monitoring and visualization includes a three-level analysis which allows calibration and basic calculations such as sums, ratios, etc. and 1D/2D histogramming. A built in script language allows creation of plug-in processes, which can do complex data manipulation.

Data rates of up to 50 Mbytes per second can be stored. For the MDPP-16 this means: with 5 channels responding simultaneously in one event, a rate of 1 MHz can be registered while measuring amplitude and timing.

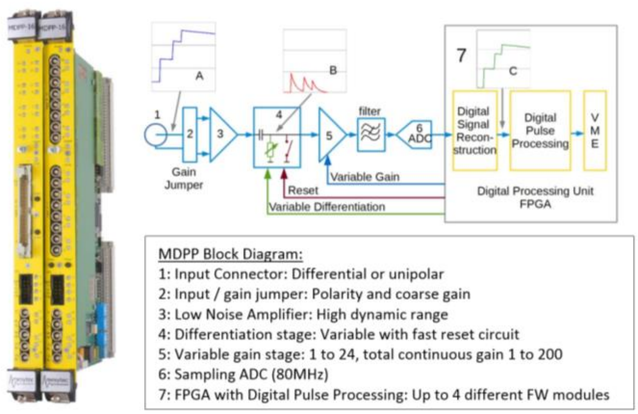


Figure 10 MDPP-16 with two front- input configuration (left) and MDPP Block Diagram (right)

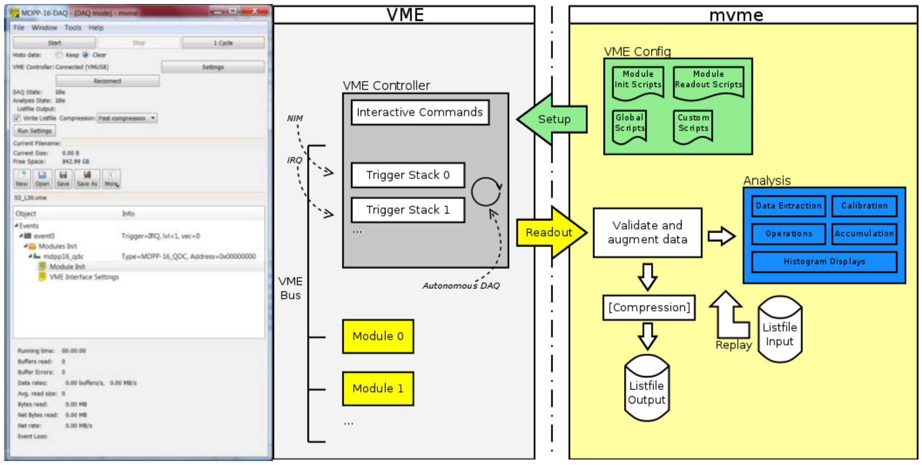


Figure 11 MVME Software GUI Window and Block Diagram

The ability to discriminate neutrons and gamma-ray signals in scintillator detectors such as Stilbene relies on the different decay times of states in the scintillator excited by different particles. Figure 12 shows the decay times of gamma rays, neutrons, and alpha particles in a scintillator detector[6]. When gamma-ray and neutron pulses are processed with pulse-shape discrimination electronics, two distinct bands appear in a plot of decay time vs. energy deposited in the scintillator. This is shown in Figure 13 with the gamma rays in the lower band and neutrons in the upper band.

The projection of the 2D plot on to a 1D plot of decay time vs. counts produces the well-known PSD plot shown in Figure 14. This plot is used to obtain a Figure-of-Merit (FOM) for the ability of a detector and associated electronics to separate neutrons and gamma rays. The higher the FOM the better is the separation. The FOM is depends on the lower and upper values of energy region used in the projection. In general the lower the starting point for the energy region, the lower the FOM. The data above lowest starting energy able to maintain FOM > 1 is considered the usable data. The best scintillators typically have a lower energy threshold of ~500 keV neutron energy.

The calibration of scintillator detectors typically use gamma-ray check sources such as ^{22}Na, ^{137}Cs, ^{60}Co, ^{232}U. Calibration is referenced to the Compton edge, so the energy scale in given in keV electron equivalent (keVee). Monoenergetic neutron sources such as D-D (2.45 MeV) and D-T (14.1 MeV) neutron generators are then used to calibrate the neutron scale.

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[6] L. M. Bollinger and G. E. Thomas, Reviews of Scientific Instruments 32, 1044 (1961)

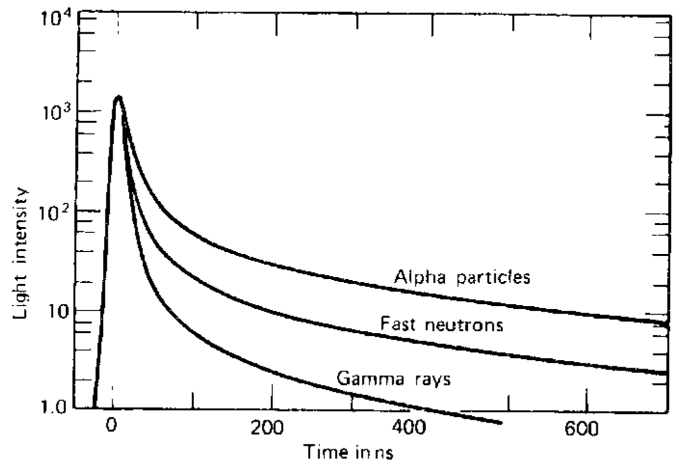

Figure 12 Time dependence of scintillation pulses in scintillator when excited by different incident particle types

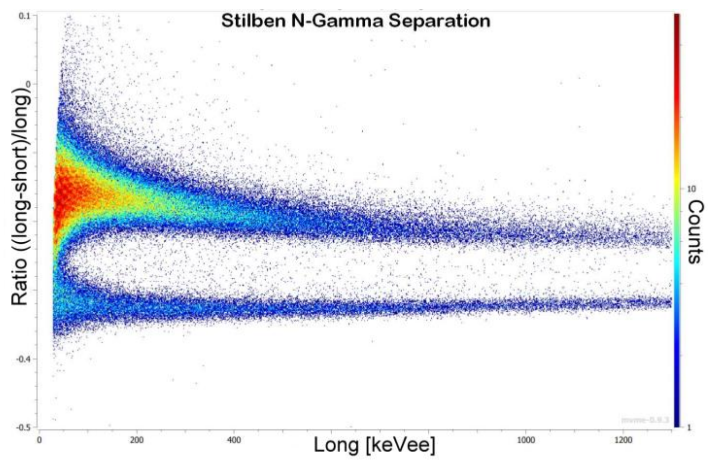

Figure 13 2D Pulse-shape discrimination plot for Stilbene crystal detector using the MDPP-16 PSD module

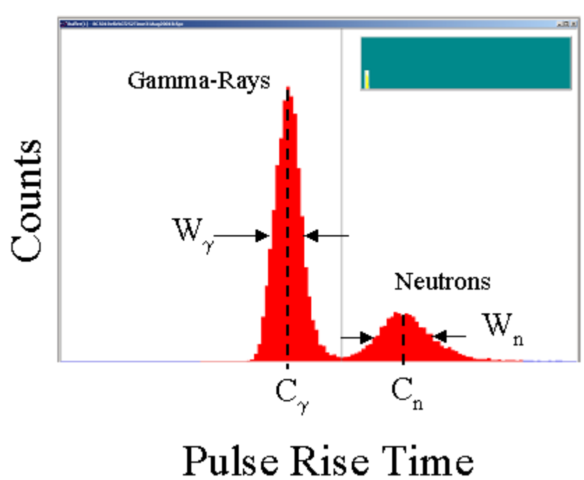

Figure 14 PSD figure-of-merit variable definitions.

The FOM is calculated using:

$$FOM = \frac{(C_n - C_\gamma)}{W_n + W_\gamma},$$

Equation 1

where C_n is the centroid of neutron events, C_γ is the centroid of gamma-ray events, W_n is the width of the neutron peak, and W_γ is the width of the gamma-ray peak.

Measurements and Results

Neutron and Gamma calibration and testing of the Stilbene detectors were performed at Lawrence Livermore National Laboratory gamma-ray check sources and neutron sources of varying energy, including ^{252}Cf source and AmLi, and D-D, and D-T neutron generators.

The figure-of-merit (FOM) of the Stilbene detector was optimized with a few hardware and software parameter settings. The MDPP short and long integration-time parameters and signal width setting (FWHM) were varied to determine effect on FOM. The integration times can be varied in steps of 12.5ns. Setting the long integration gate to 375 ns and integrating the leading edge with a short gate width of 25 ns, a good separation of neutron and gamma events became possible. The integration parameters do not affect the signal gain, i.e. the end point of the measured energy spectrum. The Stilbene signal FWHM was measured to be 30 ns. The Signal Width parameter was the set to 30 ns and varied around this value to optimize FOM. This parameter does affect the signal gain.

The Stilbene detectors were calibrated to units of keVee using ^{22}Na (511 and 1274 keV), ^{137}Cs (661.7 keV), and ^{232}U (2614 keV) sources. The location of the Compton edge was taken to be 85% of the height of the Compton shoulder. Figure 15 shows the 2D pulse-shape discrimination plot vs. energy for one of the Stilbene detectors. The spectrum was taken with a source to detector separation of 5 cm with a 2.2e4 n/s ^{252}Cf source shielded by 5 cm of lead. Figure 16 shows the Figure-of-Merit results for both detectors.

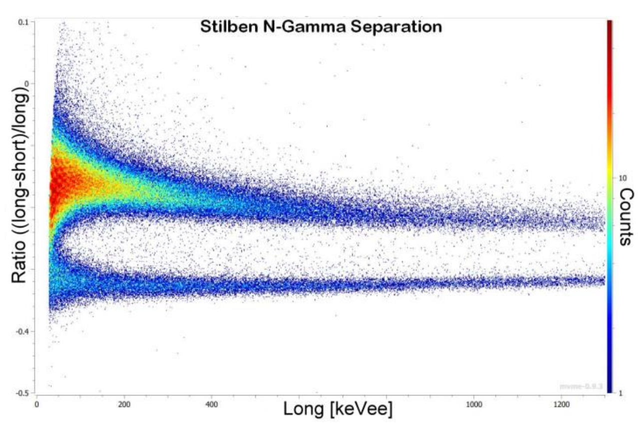

Figure 15 2D plot of Pulse shape discrimination vs. energy for a 4”x2” Stilbene crystal

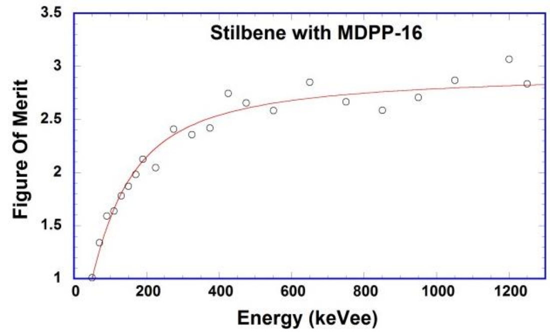

Figure 16 2D Pulse-shape discrimination FOM vs. energy plots for EJ-301 (left) and Stilbene (right)

The FOM from the MDPP-16 is also compared to historical data using a previous Mesytec pulse-shape discrimination module[7]. The historical PSD data was acquired using a 4-channel MPD-4 NIM-based module. The comparison is qualified by the fact that a different detector, source, and distance were used. Figure 17 shows a comparison plot for the EJ-301 FOM from the two systems with the MDPP-16 showing better performance than the MPD-4 module, at least at lower energies.

Figure 18 shows the spectrum for a ^{22}Na gamma-ray source and Figure 19 shows the 2D PSD spectrum for the DT source.

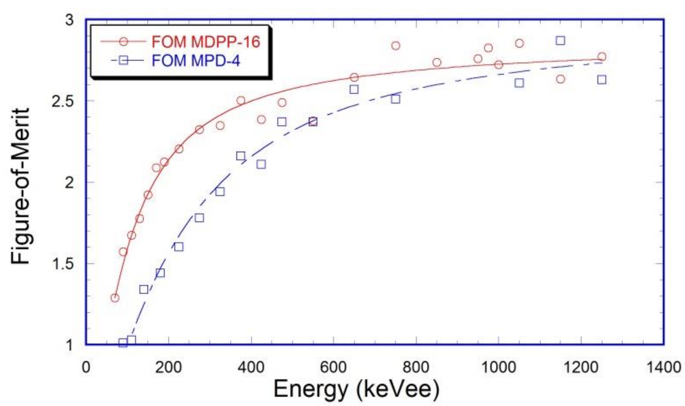

Figure 17 Comparison Plot of EJ-301 Figure-of-Merit using MDPP-16 and MPD-4 modules

[7] T. Hoagland, R. Fox, P. Kerr, G. Montermann, A. Ruben, R. Schneider, "A New Four Channel Pulse Shape Discriminator", IEEE/NSS Poster Presentation, October, 2007.

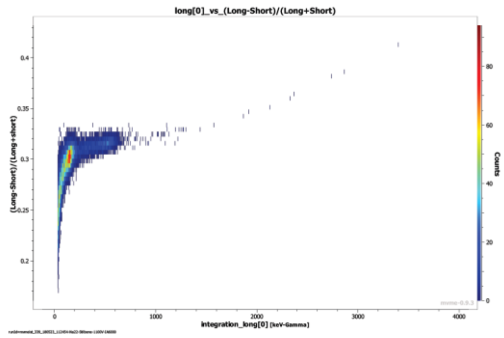

Figure 18 Stilbene detector response to ^{22}Na gamma-ray source

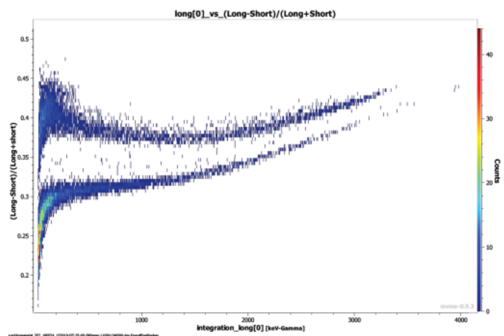

Figure 19 Stilbene detector response to D-T neutron generator

Applications

This section describes some prior and ongoing applications of the Stilbene detector array.

Stilbene Neutron Collar for IAEA

The neutron detection system used by the IAEA to monitor fresh fuel is the He-3 detector-based Uranium Neutron Collar (UNCL). The UNCL system uses an AmLi neutron source to actively interrogate a fuel assembly. The AmLi source is moderated by being embedded in a block of polyethylene. The thermalized AmLi neutrons interrogating the fuel have a high probability of inducing fission in the ^{235}U component of the fuel assembly and therefore allow an assay of the ^{235}U content along the axis of the assembly.

The thermalized AmLi interrogating neutron source signal is large compared to the signal from the induced fission on ^{235}U and reduces the sensitivity of the system. The LLNL proposed Stilbene neutron collar will be almost blind to the thermalized interrogating neutron source since it is a threshold detector that only sees neutrons exceeding an energy threshold of 0.5 to 1 MeV. Therefore, a stilbene based neutron collar will clearly see the induced fission on ^{235}U without being blinded by the interrogating source.

The fast response time permits a much shorter timegate (measurement time window) for counting neutrons and their correlations with very few ‘accidentals’. The improved signal-to-noise (S/N) of stilbene is a key motivation for using stilbene. This leads to an accurate and rapid assay in significantly lower measurement times. This high efficiency is critical to it being competitive with He-3 based neutron detectors[8]. Figure 20 shows the LLNL prototype stilbene neutron collar with 30 stilbene cells. This design has a form factor similar to the commercially available He-3 based UNCL. Also shown is a MCNP model of the stilbene collar with a fuel assembly and an AmLi interrogating neutron source. The poly bank in which it is embedded is not shown.

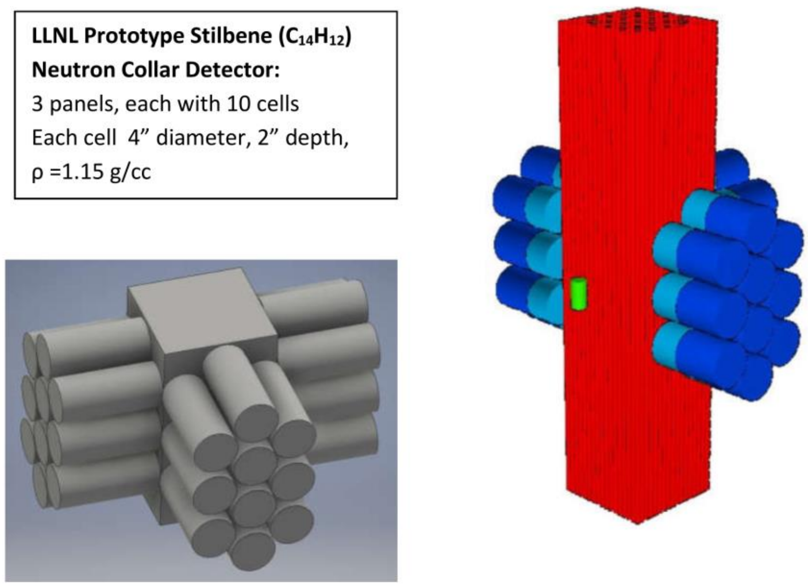

Figure 20 Figure of proposed 30-cell Stilbene detector array for new IAEA neutron collar

Enhanced Neutron Multiplicity Counter for Arms Control and Diagnostics

The Enhanced Neutron Multiplicity Counter (ENMC) is an assay system for special nuclear material (SNM) deployed during the NA-22 End-to-End Warhead monitoring campaign (E2E), 2015-2016. The system consists of a thermal neutron detector, a fast neutron scintillator array, and a D-D neutron generator to actively interrogate the material. Figure 21 shows a schematic diagram and photograph of the system as deployed in the DAF at the National Nuclear Security Site (NNSS). The system was configured to verify the ^{240}Pu source mass was unchanged before and after a dismantlement operation. In addition, the system could verify the moderating

[8] Manoj Prasad, Dan Shumaker, Neal Snyderman, Jerome Verbeke, And James Wong, “Prototype Stilbene Neutron Collar,” Lawrence Livermore National Laboratory, LLNL-TR-707598 (2016).

material around the SNM was reduced. The system used separate passive and active neutron multiplicity counting (NMC) measurements to enhance the accuracy of the mass measurement. The active NMC allowed measurement of the multiplication of the system without needing to solve the NMC equations for source mass. The approximate multiplication determined from the active measurement was feed into the analysis of the passive data to reduce the uncertainty in the one parameter, thereby reducing the degeneracies between multiplication and efficiency that can occur in NMC analysis.

The scintillator array of the ENMC deployed for the E2E project consisted of 24 Xylene based liquid scintillator detectors. As with the neutron collar described above, the liquid scintillators could be replaced by Stilbene detectors. This would simplify shipping and facility acceptance of the system as use of Stilbene would eliminate the flammability and combustibility hazard of liquid scintillators.

As an assay system, the ENMC and variations of the technology are suitable for other applications such as nuclear material control and accountancy (NMC&A) and emergency response diagnostics. The latter application will require some research and development to reduce the size and weight. The MDPP-16 data-acquisition board described above is one step in the further savings in weight and power over the ENMC system deployed in 2016 for the E2E project.

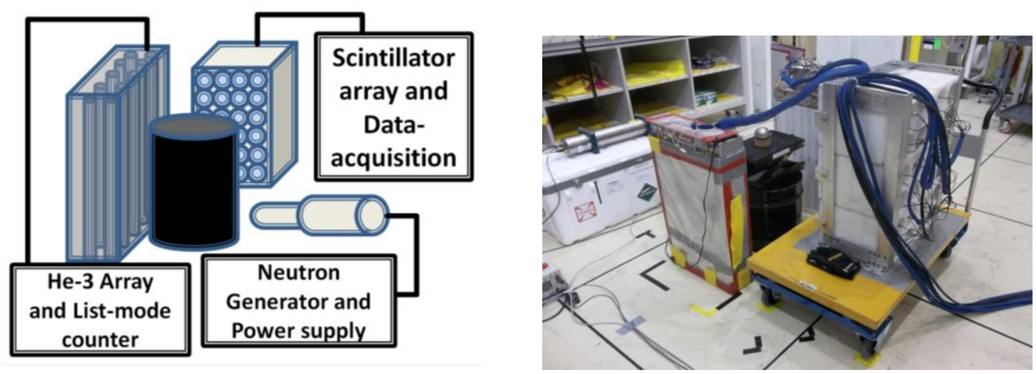

Figure 21 Schematic diagram and photograph of the Enhanced Neutron Multiplicity Counter deployed for the NA-22 End-To-End Warhead Monitoring demonstrator project

Neutron Spectroscopy and Scattering Cross Section Measurements

An additional application of the fast-neutron technology in general are general physical measurements such as the neutron spectrum of items and scattering cross sections of neutrons through materials.

If the response functions of a scintillator detector to neutrons of different mono-energetic neutron sources are measured, complex neutron energy spectra can be determined through a mathematical unfolding process. The energy spectrum is a quantity useful in understanding nuclear processes in fissioning assemblies. Obtaining an estimate of the expected neutron spectrum for a planned system under study is also useful in selecting the most efficient detector for the mission.

Another use of neutron energy information is the measurement of the scattering cross section Σ_s of neutrons through materials. The neutron scattering cross section refers to the ability of the material to remove full-energy neutrons from the original beam of neutrons. Measurements of this type involve a source of neutrons, a material under study, and a detector to monitor neutrons escaping the material. For example, in order to measure the neutron scattering cross section of D-T (14.1 MeV) neutrons through a material, the detector should ideally be sensitive to just the full-energy 14.1 MeV neutrons.

Scattered neutrons lose energy but can still be detected by a detector sensitive to a wide energy range. Such detectors include moderated He-3 detectors, as well as fast-neutron scintillator detectors. The advantage of the fast-neutron scintillator detectors is that the energy information from the detector can be used to filter for only the highest energy neutrons. This is then a measure of unscattered neutrons. Figure 22 shows an example scintillator energy spectrum from a source of D-T neutrons incident on polyethylene. Calculation of the neutron scattering cross section was performed using just the highest energy band shown. Note that this type of measurement is not sensitive to the absorption cross section Σ_a since those thermal neutrons would be below the 500 keV detection threshold and not detected whether absorbed or transmitted.

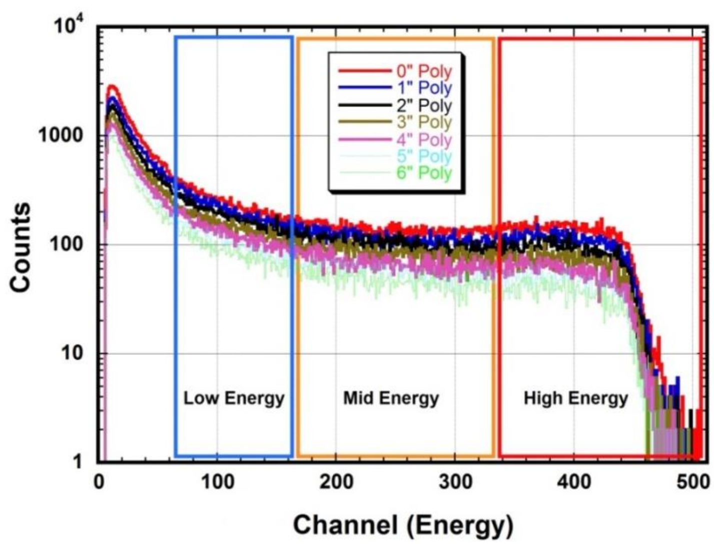

Figure 22 Plot of the 2m EJ-301 energy spectrum for D-T neutron generator for all seven HDPE configurations

Summary

A 10-Cell array of 4” x 2” Stilbene crystal detectors has been built and calibrated at LLNL. Stilbene is of interest due to excellent PSD properties and low hazard (non-flammable). Stilbene has the highest organic scintillation efficiency with much higher light yield than plastics or liquids. The excellent PSD capability of Stilbene implies excellent gamma-ray rejection, which in turn implies high neutron counting efficiency. This array is currently funded through NA-241 to investigate upgrading the existing IAEA uranium neutron collar (UNCL) to Stilbene for assay of uranium in fresh fuel.

Fast-neutron multiplicity with scintillators provides a new window into neutron fission-chain physics. Applications of fast-neutron scintillation neutron and gamma-ray multiplicity counting include arms control, safeguards, NMC&A, emergency response diagnostics, and basic physical measurements.

A new Mesytec MDPP-16 digitizer VME module using a hybrid digital/analog algorithm has been tested for use with the Stilbene array and shows both ease of use and deployment, as well as excellent PSD performance and a low demand on data transfer rates. The high resolution in time (<100ps) and energy (<0.5%) enable use in neutron multiplicity counting as well as energy spectroscopy for a wide range of detectors including HPGe.

Acknowledgements

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