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ABSTRACT

Technology is needed to prevent diversion of fissile material and further special nuclear material accounting, control, and safeguards efforts under the Treaty on the Nonproliferation of Nuclear Weapons. Most measurement systems used in these applications still rely on He-3, whose shortage drives the need of a replacement technology for neutron detection in the near future. In He-3 counters, fission neutrons need to be moderated. Complex electronic circuitry and unfolding procedures are also needed to derive fission correlated counts from detected neutrons in order to quantify fissile mass. Fast neutron counting systems are capable of shortening the coincidence window by several orders of magnitude and thereby provide a direct estimate of fissile mass. We present the design and experimental validation of a fast-neutron multiplicity counter, using a matrix of liquid and stilbene scintillators. The system is based on eight 7.62 cm x 7.62 cm EJ-309 liquid and eight 5.08 cm x 5.08 cm stilbene detectors, arranged in a checkerboard assembly. Given the superior gamma-neutron discrimination capabilities of stilbene, assay of higher-order multiplicity and of samples with high (α,n) source terms are made possible, suitable for the analysis of newer fuels containing high concentrations of plutonium and other fissionable actinides. The system measured separate Cf-252 and PuBe sources to evaluate its capability to detect and discriminate fission neutrons. Neutron, photon, and joint neutron and photon multiplicities were measured, acquiring detected pulses by means of fast electronic digitizers (sampling rate up to 500 MHz). Measured results have been also used to validate a Monte Carlo model of the system, developed in MCNPX-PoliMi code, and to fully characterize signal crosstalk component due to false positive coincidence counts following scattering in nearest-neighbor detectors. The detailed Monte Carlo model also allowed optimizing crosstalk rejection during system design stage. In the full paper, we show that the fast neutron multiplicity counter presented in this work achieves good efficiency for the measurement of neutron, photon, and joint neutron and photon multiplicities, and is a promising alternative to He-3 based systems.

1. INTRODUCTION

MULTIPLICITY counters traditionally rely on helium-3 (^{3}He) based detectors. ^{3}He shortage raises the need for a replacement technology. Fast organic scintillators are not only considered one of the most promising replacement detectors, but their fast response can offer a great improvement over standard time-correlated measurements. We developed a system based on organic scintillators. The capability of similar passive systems to quantify the mass of special nuclear material (SNM) in a variety of different samples, i.e. fresh and irradiated nuclear fuel assemblies, was already proven by our group (Dolan et al. 2014). In this work, we present the design and experimental validation of a system based on eight EJ-309 and eight stilbene detectors. The use of organic scintillators allows an exceptionally short gate time up to 200 ns, compared to hundreds of microseconds needed for moderated systems based on thermal neutron detectors. This approach results in a reduction of accidental neutron coincidence. In addition, stilbene detectors have been introduced in the system to exploit their superior gamma-neutron

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discrimination properties.

2. METHODS

The University of Michigan multiplicity counter includes eight liquid organic scintillators (EJ-309 7.62 cm Φ x 7.62 cm, by Eljen, Sweetwater, TX) and eight crystalline organic scintillators (stilbene 5.08 cm Φ x 5.08 cm, by Lawrence Livermore National Laboratory and Inrad Optics, Northvale, NJ) in a checkerboard arrangement (Fig. 1). We assembled a prototype system, including 8 EJ-309 detectors and acquisitions electronics and tested it using spontaneous fission and (α, n) radionuclide sources. We developed a Monte Carlo model of the system, including detectors and source term, using MCNPX-PoliMi (Pozzi, Padovani, and Marseguerra 2003), and validated it with experimental results. We then included 8 stilbene detectors and thoroughly characterized them, in order to select and optimize their operational conditions. We updated the Monte Carlo model including the stilbene detectors and implemented measured stilbene intrinsic properties.

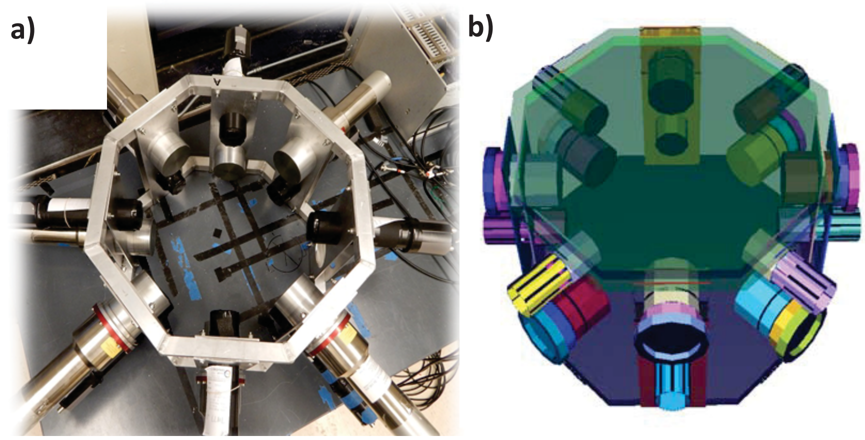

Figure 1. Multiplicity counter assembly (a) and its MCNPX rendering (b).

2.1 Experimental validation

A prototype of the well-counter was assembled and tested at the University of Michigan. The prototype is based on 8 liquid organic scintillators (EJ-309 7.62 cm Φ x 7.62 cm). The acquisition electronic was based on 250 MHz sampling rate, 12 bit analog-to-digital converter (mod. V1720, CAEN, Italy) (CAEN 2014). The system encompasses a single ring of detectors with a source-to-detector distance of 17 cm. This system was used to measure neutron and photon cross-correlated counts emitted by two sources: ^{252}Cf (31 μCi) and PuBe (1 Ci).

2.2 Stilbene detector characterization

Organic liquid scintillators based on EJ-309 as detection medium have been studied and well characterized (Enqvist et al. 2013). However, the eight stilbene crystals included in the well counter were grown by manufacturers using a solution-based technique developed in 2013 (Carman et al. 2013). Detailed specifications of these detectors were not available in the literature. We characterized them in terms of intrinsic efficiency and energy resolution. Calibration was performed before each characterization measurement, using a 137-Cs source and matching 80% of the Compton edge peak with 0.4 V pulse height and setting a 36 keVee detection threshold. Table 1 shows voltage gain per each stilbene detector.

Table 1 High voltage gain used per each detector. Stilbene manufacturer and photomultiplier tube (PMT) models. PMT are distributed by ET Enterprises, Sweetwater, TX. The two used models 9214A and 9214B (ET Enterprises, n.d.) have a maximum operating voltage of 2 kV and 2.3 kV respectively.

Channel	*Manufacturer*	*Photomultiplier tube model*	*HV Gain (V)*	*Efficiency (%)*
1	Lawrence Livermore National Laboratory	9214A	1066	27.0
2	Inrad Optics	9214A	969	26.0
5	Inrad Optics	9214A	1152	25.0
6	Inrad Optics	9214A	1089	25.5
9	Inrad Optics	9214A	1206	25.1
10	Inrad Optics	9214B	1352	22.8
13	Inrad Optics	9214A	1427	25.0
14	Inrad Optics	9214A	1315	24.2

2.2.1 Intrinsic efficiency characterization.

The neutron intrinsic efficiency (Table 1), defined as the ratio between the number of neutrons counted by the detector and those impinging on its front face, was measured using a 3 μCi point ^{252}Cf source, 20 cm source-to-detector distance and detection threshold of 36 keVee.

2.2.2 Energy resolution characterization.

Detector energy resolution, defined as the full-width-at-half maximum of peak response function divided by peak center value (i.e. mean of the peak distribution, if assumed Gaussian), was measured using the Compton spectrometry technique (Knoll 2010, Pausch et al. 2011). At least two detectors are used in this technique. In our experiment, we used a stilbene and a NaI(Tl) detector. The prevailing mode of interaction is Compton scattering in the stilbene, and photoelectric effect in the NaI(Tl). A fraction of the gamma rays scattered within the stilbene is detected by the NaI(Tl). The energy spectrum of these two events in coincidence represents single Compton scattering events within the stilbene. The relative position of the two detectors selects a single scattering angle (θ). According to Compton scattering physics, a constant amount of energy (E_e) is transferred to the electron within stilbene in coincidence with photoelectric absorption within the NaI(Tl) detector (Knoll 2010). The energy deposited (E_e) is a function of the scattering angle (θ), the impinging photon energy (E_γ), and the electron rest energy ($m_e c^2$), under the hypothesis that the electron is close to rest before scattering (Eq. 1). We applied this technique using 137-Cs and 54-Mn laboratory check sources, experimental details are reported in Table 2. Table 2 reports the measured light output in keVee, which is proportional to the energy deposited by photon interaction (light yield of 1 keVee/keV), in the energy range of interest.

$$E_e = E_\gamma \left(\frac{\frac{E_\gamma}{m_e c^2}(1-\cos(\theta))}{1+\frac{E_\gamma}{m_e c^2}(1-\cos(\theta))} \right) \quad 1$$

Table 2. Radionuclide sources and experimental conditions used to estimate stilbene detector energy resolution.

	Source type and activity	*Source-to-stilbene detector distance (cm)*	*Scattering angle (deg)*	*Measured light output (keVee)*
Configuration 1	137-Cs 1μCi	100	45	182
Configuration 2	137-Cs 1μCi	5	180	470
Configuration 3	54-Mn 1μCi	5	180	641

2.3Monte Carlo model of the system

We have developed an MCNPX-PoliMi model of the system. Figure 1.b shows an illustration of the simulated full geometrical model in Visual MCNPX. The model also includes some structures of the irradiation room, not shown in Figure 1.b, such as the floor and the table, to take in to account radiation scattering. The detector resolution function, measured as discussed in Section 2.2.2 was included in the model using MPPost, the MCNPX-PoliMi post processor software. MPPost simulates the detector response based on the particle transport information provided by MCNPX-PoliMi and user provided input data. MCNPX-PoliMi incorporates several source models. We simulated the ^{252}Cf source using one of the spontaneous fission models implemented in MCNPX-PoliMi (IPOL(1)=1), whose angle and energy distribution depends on multiplicity. To simulate the PuBe neutron source, we combined a standard MCNPX energy defined source with a ^{240}Pu spontaneous fission source available in MCNPX-PoliMi (IPOL(1) = 3), in order to take into account a small amount of ^{240}Pu inside the source (0.16 g, less than 2% of the total plutonium mass). The code also includes a specific treatment for induced fissions. The first Monte Carlo model included 8 EJ-309 detectors and it was then upgraded to include 8 stilbene detectors in a checkerboard assembly.

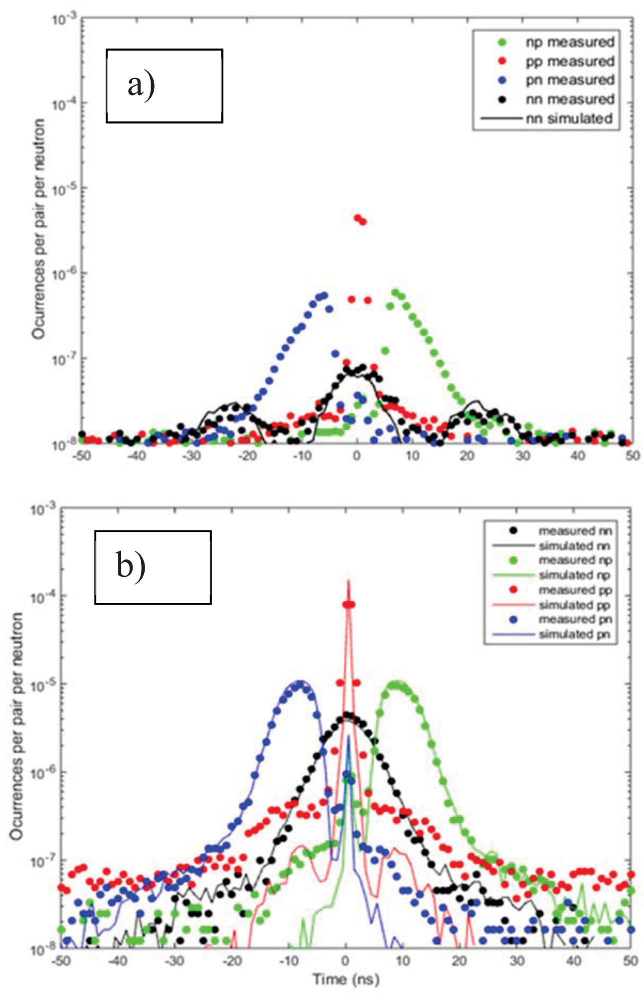

Figure 2. Cross correlated pairs for a PuBe (a) and a 252-Cf (b) source.

3. RESULTS AND DISCUSSION

Figure 2 shows cross-correlated counts measured using the single-ring prototypal version of the counter for the two above-mentioned sources: ^{252}Cf (31 μCi) and PuBe (1 Ci). Photon and neutron correlated measured data within a 50 ns time window are in very good agreement with simulated data. Figure 3 shows single and multiple counts up to fourth order detected by the well counter for the two sources. The number of single counts per unit source strength is slightly higher for 252-Cf compared to PuBe. This difference is due detector intrinsic efficiency energy dependence (Kornilov and Kagalenko 2000). The doubles and multiplets measured for 252-Cf are at least one order of magnitude higher than PuBe. Multiple counts in PuBe source are mostly due to cross talk, traces of 240-Pu in the source (0.16 g, less than 2% of the total plutonium mass) and (n,2n) reactions on beryllium.

Stilbene detectors were then characterized and included in the multiplicity counter and showed an average intrinsic efficiency of 25.5% with an inter-batch standard deviation of 7%, using a 3 μCi point ^{252}Cf source (Table 1). This wide dispersion around the average value is probably due to intrinsic detector differences due to the relatively recently developed manufacturing process. The energy resolution of the detectors is shown in Figure 4. An inverse function of the energy (Eq. 2) was used to fit the measured data in the 200-700 keVee light output range. Previously published data for 3.81 cm Φ x 3.81 cm stilbene detectors reported an energy resolution in the 0.33-0.16 in the same energy range (Cialella and Devanney 1968).

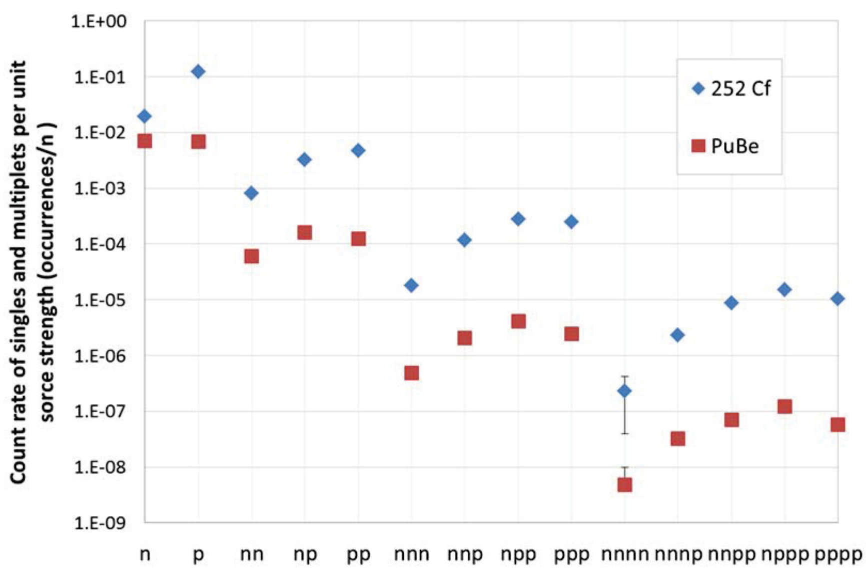

Figure 3. Measured count rate of single and multiple counts per unit source strength.

$$(\Delta E/E) = \sqrt{(3.45\text{E-}4)^2 + (5.01\text{E-}2)^2/E + 1.77\text{E-}2/E^2} \qquad 2$$

The resolution function in Eq.2 was implemented in the MPPost post processing code and yielded the simulated results for the whole system shown in Figure 5, where neutron doubles are compared, for a PuBe source and a 252-Cf. Figure 5 shows neutron doubles detected within the 60 ns coincidence window in two detectors, at an angular displacement of 45º (Figure 5.a) and 180º (Figure 5.b). Coincidence time distributions are unavoidably affected by cross-talk, whose effect is more relevant at small angles. Cross talk is the major contribution to coincidence events in (α,n) neutron sources, PuBe in this case, together with (n,2n) reactions on beryllium and traces of 240-Pu, which undergoes spontaneous fission.

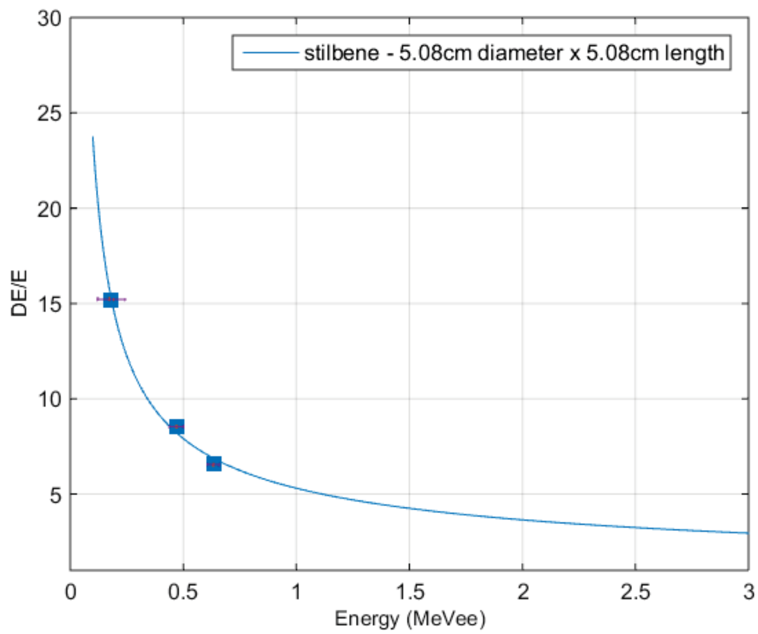

Figure 4. Stilbene resolution as function of light output

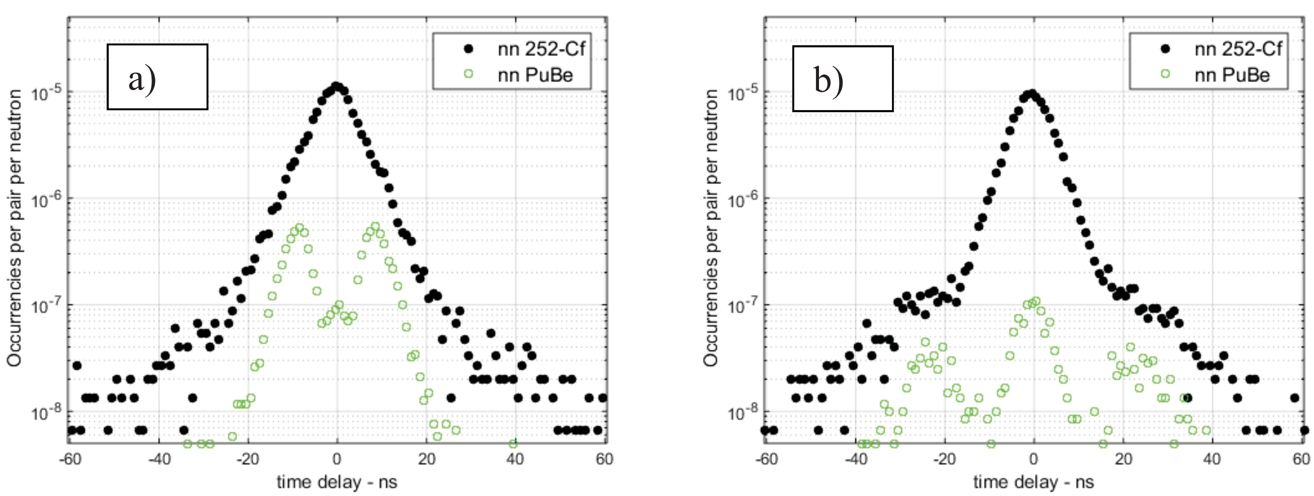

Figure 5. Cross correlated neutron doubles for a PuBe and a 252-Cf source. Cross correlations are reported for detectors placed at 25º (a) and 180º (b).

4. CONCLUSIONS AND FUTURE WORK

In this work we proved the viability of a multiplicity counter based on organic scintillators to discriminate time-correlated fission neutrons from random, uncorrelated neutrons. A prototype of the system was tested with spontaneous fission and (α,n) neutron sources, 252-Cf and PuBe respectively. Measured time correlated photon and neutron doubles and multiplets were about one order of magnitude higher in the spontaneous fission source source than the (α,n) neutron source. The final version of the well counter includes also stilbene detectors, which have been characterized in terms of intrinsic efficiency and energy resolution. Measured energy resolution has been included in the Monte Carlo model of the full system and confirmed the cross correlation trend measured with a single ring of EJ-309 detectors. An experimental campaign will be also carried out at Idaho National Laboratory in August 2015, where the multiplicity counter described in this work will be used to characterize several assemblies of plutonium fuel plates with 239-Pu mass in the 0.1-2.5 kg range. This multiplicity counter can greatly improve verification capabilities of SNM material inventory declarations. For this application, the system will be ruggedized and we will study and optimize its performances in harsh environments.

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