Characterization and use of Stilbene scintillator for neutron metrology and spectrometry from 100 keV to 22 MeV

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Abstract—Stilbene scintillators have now the capability to extend the neutron energy measurement from 100 keV to 1 MeV. To be used as spectrometers, scintillators have to be characterized in entire energy range covered. For the photon characterization, usual calibration source determined the calibration and the resolution function. To do the photon matrix, MCNP PTRAC simulations were done between 59 keV up to 7 MeV. Using MCNP PTRAC allows the calculation of the photon response matrix from the tracking detail of the electron energy deposition in the crystal, including the effect of the Aluminum protection around. Resolution and sensibility obtained from gamma sources are applied later to the calculated photon matrix.

For neutron characterization, Time-Of-Flight measurements on white neutron spectra were realized. However, the fluence reference on these spectra begins at 1 MeV. So, the shape of the response function was measured using stilbene scintillators on 100 keV-1 MeV decade. After, the monoenergetic neutron reference of AMANDE facility was used to give neutron fluence normalization below 1 MeV. This method could characterize these scintillators for neutron energy from 100 keV up to 22 MeV.

Keywords — Scintillator characterization, digital acquisition, neutron, Time-Of-Flight, stilbene

I. INTRODUCTION

The laboratory for micro-irradiation, neutron metrology and dosimetry (LMDN) is in charge of establishing metrological references in France for neutron fluence energy distributions and associated dosimetric quantities, on behalf of the LNE which is the French National Metrological Institute (NMI). One of the laboratory purposes is to maintain and improve metrological references and calibrations.

The laboratory has two facilities for establishing metrological references. The first, named AMANDE [1], has an ion accelerator which can produce mono-energetic neutron fields between 2 keV and 20 MeV. Metrological references by spectroscopy are difficult to cover below 1 MeV due to the lack of efficiency of usual scintillators [2]. For the same reason, Time-Of-Flight references are usually not covered below 1 MeV.

Stilbene with higher purity demonstrated their capability to detect neutrons energy until less than 100 keV [3]. So, the use of stilbene scintillator could allow the extension of the reference measurements, with or without Time-Of-Flight, to the 100 keV-1 MeV energy range.

The second facility, named CEZANE, has two sources of reference \((^{241}\text{AmBe}\) and \(^{252}\text{Cf}\)) used as calibration standard [ISO 8529 standard]. As \(^{252}\text{Cf}\) source is isotopically impure [20], regular source calibration [4], not possible on site, is required. To overcome this difficulty while maintaining metrological references, the use of reference scintillators for source decay monitoring is planned.

To cover these needs, two stilbene scintillators, coupled to a digital acquisition system, were purchased. However, neutrons characterization of these stilbene scintillators is needed. As all neutron fields are accompanied with a photon field, the stilbene scintillator characterization for photons is interesting. Ultimately, this work will result in a high-resolution neutron spectrometer with a large range of energy [3] and a low-resolution photon spectrometer [5].

II. STILBENE SCINTILLATORS AND DIGITAL ACQUISITION

A. Stilbene scintillators

Stilbene scintillators are 2”x2” cylindric organic scintillators composed of Stilbene crystal (C\(_{14}H_{12}\)). Photomultipliers were adapted for each scintillator energy range target. The new stilbene growth method allows a more homogeneous crystal [6-7] with a size compatible with our needs. This purer crystal can detect neutron energy down to 100 keV and discriminate with photon [3, 8-9].

The LMDN stilbene scintillators (named SX and SX2) were purchased at Scionix [10] in 2020. SX has a fast photomultiplier response time [11] and is dedicated to a large range of energy (500 keV-22 MeV). SX2 has a large light collection photomultiplier with relatively low time response and is dedicated to low neutron energy range (100 keV-3 MeV).

B. Digital acquisition

The digital acquisition is a DT5730 [12] distributed by CAEN [13]. The measure with scintillators at higher energy range is limited by the DT5730 digital range amplitude (0-2 Volt). In order to increase the operating range of the SX, the
signal at the scintillator anode is split in two signals both with halved amplitude. One signal amplitude is reduced with a factor of four by a passive attenuator before being digitized. The second signal is digitized on the same digitizer directly after the splitter.

This method makes a channel for the "high energy" neutron and the second channel for "low energy" neutron with a full energy range recovery. The high tension of scintillator and the passive attenuator are optimized and chosen to have a low channel energy limit upper than 7 MeV and a high channel energy limit lower than 7 MeV [14]. The entire energy range of SX can be measured in one go.

The discrimination between neutron and photon is realized with the Pulse Shape Discrimination (PSD) using the Charge Comparison Methods (CCM) as described in [3]. The PSD is directly obtained by CoMPASS software [15]. The long integration gate was adjusted to limit statistical drift at low energy due to an incorrect estimate of the baseline. The fast gate was adjusted to optimize the discrimination threshold. The Constant Fraction Discrimination (CFD) of both scintillators were optimized by a dedicated study on a $^{22}$Na photon source [14].

Scintillator main characteristics are presented in Table I.

<table>
<thead>
<tr>
<th>Characteristics</th>
<th>SX (2 channel)</th>
<th>SX2 (1 channel)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Used neutron energy range</td>
<td>500 keV – 22 MeV</td>
<td>100 keV – 3 MeV</td>
</tr>
<tr>
<td>Temporal response</td>
<td>1 ns</td>
<td>2 – 4 ns</td>
</tr>
</tbody>
</table>

### III. THE PHOTON CHARACTERIZATION

The electron energy deposition will be calculated by a simulation. Many experimental photon spectra will determine sensitivity and resolution of the measurement device. Photon response matrix will be obtained after parametrization of calculation with experimental sensitivity and resolution.

A. The electron energy deposition by simulation

The electric signal at the scintillator anode is named “Light Output (LO)”. The simulation is realized with Monte Carlo N-Particle code (MCNP) [21]. In the simulation, the scintillator is modelled with the size, composition and density (1.15 g.cm$^{-3}$) of IRSN stilbene and a 0.4 mm thickness of Al covering the crystal. The air around the detector is modelled, and the composition and density are those presented in [16]. The isotropic photon source, assumed to be point-like, is positioned at 20 cm from the crystal on the cylinder edge. The photon energy varies between 50 keV up to 7 MeV with a step of 1 keV.

MCNP PTRAC is used to determine the electron energy deposited in the crystal. Indeed, even if MCNP F8 tally is often used to simulate the photon response matrix [3], it is a pseudo-kerma which does not calculate the exact electron energy deposit.

A root program reads the output files and determines the electron energy deposited in the crystal, considering all contributions, including the Al cover contribution. Different cases are possible and must be considered.

The first case is the simplest one: the photon interacts with an electron of the crystal, which deposes all its energy in it. In the second case, the photon interacts with an electron of the crystal, which gets out it and so without depositing all its energy. In the third case, the photon interacts with two or more electrons in the crystal. In this case, the electron energy deposits in the crystal are summed. In the fourth case, the photon does not interact with electron but creates electron pair. The electron/positron pair is considered for energy deposition as previous case. An additional contribution from the two 511 keV photon annihilation from positron is also considered as they can lead to electron energy deposition in the crystal. The contribution of Al is also considered, with photon pair creation or electron coming in crystal.

Cases are resumed on Fig. 1. The aluminum protection is in grey, and the crystal is in blue. The black arrows correspond to the photons emitted by the source. These photons collide in dark blue with electrons in red, or pair off in orange with a positron emitted in green. Several cases can occur during the same interaction.

**Fig. 1: Different cases in the crystal and aluminum**

A spectrum for each energy between 50 keV and 7 MeV with a step of 1 keV, will be obtained. These spectra will be used to construct a photon response matrix without sensitivity and resolution.

The experimental spectrum from a $^{137}$Cs source positioned at 20 cm is shown on Fig. 2. It is presented with the result of the equivalent simulation at 662 keV from the MCNP PTRAC and the equivalent MCNP tally 8. All simulation is presented without considering the stilben resolution and its absolute normalization considering the well know source activity (379.59 kBq).

The Compton front on the MCNP PTRAC analysis and MCNP tally 8 can be identify. The MCNP tally 8 is clearly too low up to the Compton edge, especially at low electron energy. Whereas the MCNP PTRAC analysis enables to reproduce accurately the Compton shape.
IV. THE NEUTRON CHARACTERIZATION

The neutron characterization will be experimentally made using a Time-of-flight (TOF) method from white spectra. TOF is the metrological method to determine the neutron energy. The neutron speed, so its energy, is determined by the time difference between its emission and its detection, knowing the distance between both places. White spectra are issued from Neutron For Science (NFS at Caen in France) and Physikalisch-Technische Bundesanstalt (PTB at Braunschweig in Germany). These white spectra are obtained with a pulsed beam interacting with a thick target [17-18]. White spectra at PTB and NFS are shown on Fig. 3.

A. Time-Of-Flight

The scintillators will be positioned between 10 and 30 meters away at NFS and 14 meters at PTB. The distance at NFS and the beam pulsation are adjusted to allow the low neutron detection limit before the photons of the next pulse may arrive. Indeed, photons are produced at the same time as neutrons by particle beam interaction in the target. The target energy, \( E \) is calculated according to (1) [19]: with \( m \) is the neutron mass, \( c \) the velocity of light and \( L \) is a distance between the detector and the target.

\[
E = mc^2 \left(\frac{1}{\sqrt{1-\beta^2}} - 1\right) \quad \text{with} \quad \beta = \frac{v}{c} = \frac{L}{cT},
\]

(1)

\( T \) is given by (2) but is calculated with (4). Indeed, the photon peak time gives the time reference production, knowing the distance between the target and the detector (3). In this equation, \( t_0 \) is the arrival time of the pulsed beam in the target. It is assumed that both neutrons and photons are emitted from the target at \( t_0 \). \( t_{\text{photon}} \) and \( t_{\text{neutron}} \) are respectively the photon and the neutron time detection in the scintillator, respect to the beam pulsation.

\[
T = t_{\text{neutron}} - t_0 ,
\]

(2)

\[
t_{\text{photon}} = \frac{L}{c} ,
\]

(3)

\[
T = t_{\text{neutron}} - t_{\text{photon}} + \frac{L}{c} ,
\]

(4)

B. The white Spectra

In order to determine the neutron response of the scintillator at a given energy, time windows are made in neutron time spectrum. The width of the time windows gives the energy resolution of the response matrix. To be valid, the number of events must be sufficient. Also, a compromise between energy resolution and statistics must be found. All the neutron-induced events in the time window are grouped together to obtain the LO profile assumed at the average neutron energy windows. All the windows are used to obtain, after correction, the response matrix over the whole energy range.

To limit the pile up effect and thus having to make a large and difficult correction, it is necessary to limit the number of events per pulse. In practice, an event rate lower than 1 per 100 pulses seems to be a good compromise.

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B. The resolution and the sensitivity of scintillators

In order to complete the photon response matrix, the calculated photon matrix is convolled with experimental parameter. The sensitivity and the resolution function of scintillator are deduced by monoenergetic photon spectra. The list of photon sources and rays is shown in Table II.

For all sources except \(^{241}\text{Am-Be}\) with polyethylene, the sources are at 20 cm from scintillators in the same configuration as the MCNP-Simulation. The \(^{241}\text{Am-Be}\) with polyethylene source size is a cube of around 20 cm edge and the edge is only 1 cm from scintillators.

<table>
<thead>
<tr>
<th>Photon Sources</th>
<th>Photon rays used [keV]</th>
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<tbody>
<tr>
<td>(^{241}\text{Am})</td>
<td>59</td>
</tr>
<tr>
<td>(^{133}\text{Ba})</td>
<td>80, 302 et 356</td>
</tr>
<tr>
<td>(^{137}\text{Cs})</td>
<td>661</td>
</tr>
<tr>
<td>(^{22}\text{Na})</td>
<td>511 et 1274</td>
</tr>
<tr>
<td>(^{207}\text{Bi})</td>
<td>569, 1063 et 1770</td>
</tr>
<tr>
<td>(^{56}\text{Co})</td>
<td>1173 et 1332</td>
</tr>
<tr>
<td>(^{241}\text{Am-Be})</td>
<td>4438</td>
</tr>
<tr>
<td>(^{241}\text{Am-Be + Poly})</td>
<td>2223</td>
</tr>
<tr>
<td>(^{152}\text{Eu})</td>
<td>121, 244, 344, 778, 964, 1085, 1112 et 1408</td>
</tr>
</tbody>
</table>

The sources, except the \(^{152}\text{Eu}\), are used to determine the sensitivity and the resolution of scintillators at different energies. Due to the photon energy range of SX, the 59 keV peak of the \(^{241}\text{Am}\) and the 80 keV peak of the \(^{133}\text{Ba}\) are used only for SX2. Due to the photon energy range of SX2, only energy below 511 keV can be used. When the photon response matrix is completed, the spectrum will be used to check the deconvolution method and the matrix. The \(^{152}\text{Eu}\) will be used as a test spectrum for the deconvolution [3].

The MCNP PTRAC simulation result is convoluted with gaussian function which width at half height function is adjusted to fit the experimental Compton edges or photoelectric peak. Using the same methods as described in [5], the sensitivity and the resolution of the scintillators will be determined.
After a detection, the time to return to the operating point depends on the energy deposit and the type of particle. The greater the energy deposit is, the longer the return to the operating point will be. In this case, a secondary trigger is possible, so the acquisition blocking time must be increased.

Fluence references are established on the white spectrum of the PTB by its reference detector. This reference will therefore be used to determine all the reference fluences, including beyond its high energy limit using both spectra results, extrapolation, and simulation.

For neutron characterization between 100 keV and around 1 MeV, there is no fluence reference at PTB or NFS as neutron reference detectors have no efficiency. However, neutron at this energy obviously exists both at PTB and NFS. So, the LO shape of the neutron detectors responses will be measured. The fluence determination for LO shapes will be done using monoenergetic neutron fields from at AMANDE facility.

V. CONCLUSIONS

In neutron metrology, scintillators are often used as secondary standards due to their specific qualities - very fast response, high neutron efficiency and response, neutron-photon discrimination, etc. However, their "high" detection threshold around 1 MeV induces that white (or large energy range) neutron fields with a component below this limit relatively unknown. Recent progress in crystallography on stilbene crystals has led to the development of scintillators with threshold detection around 100 keV, so one decade lower. Neutron characterization of such stilbene scintillators will provide a directional neutron spectrometer over an extremely wide energy range (100 keV-22 MeV), so including 100 keV-1 MeV decade. As this energy range is particularly wide, various experimental techniques are required. Data analysis will also require a specific study to establish neutron response matrices and carry out deconvolution. Photon characterization of the same scintillators will enable the evaluate the contribution of photons in a neutron field. All of this work will complement the time-of-flight references on AMANDE facility and improve the characterization of the metrological references of LMDN.

ACKNOWLEDGEMENTS

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