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Extended time-of-flight measurements down to 100 keV at the AMANDE facility with a stilbene scintillator

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Abstract

The time-of-flight (ToF) method with scintillators is routinely used for determining neutron energy. However, a technical difficulty related to the loss of scintillator efficiency below 1 MeV makes this technique difficult to implement for the energy decade [100 keV–1 MeV]. New crystal production techniques provide stilbene scintillators efficient in this low neutron energy region, making it possible to extend the ToF technique below 1 MeV. In this manner, measurements of secondary reactions (d,n) on carbon or oxygen nuclei in this range become feasible, which should lead to improved reference calibration conditions in neutron fields produced by a deuterium ion beam.

Introduction

The LMDN (*Laboratoire de micro-irradiation, de métrologie et de dosimétrie des neutrons*) of the IRSN (*Institut de Radioprotection et de Sureté Nucléaire*) is missioned by the LNE (*Laboratoire National de Métrologie et d'Essai*, i.e. the French NMI, National Metrology Institute) to ensure the French references in neutron metrology and associated dosimetry quantities. To this end, the LMDN operates the AMANDE facility (*Accélérateur pour la Métrologie et les Applications Neutroniques en Dosimétrie Externe*), which produces monoenergetic neutron fields between a few keV and 20 MeV⁽¹⁾ using an ion beam (proton or deuteron), which can be pulsed or continuous, with a maximum energy of 4 MeV. The AMANDE pulse structure allows the time-of-flight (ToF) method⁽²⁾ to be used to measure and connect neutron energy to the SI (International System of Units). In practice, this method is generally implemented with a scintillator, as at AMANDE above 1 MeV in neutron energy. Indeed, scintillators have two decisive advantages: a short response time on the order of a nanosecond and

the capability to measure and provide discrimination information for both photons and neutrons.

However, below 1 MeV, the sharp drop in scintillator efficiency and the loss of neutron/photon discrimination limits their use. This is why, in 2018, a dedicated project to extend the ToF method to the [100 keV–1 MeV] energy range was launched in partnership with the LNE.

Stilbene scintillator

Stilbene organic scintillators are made of large single crystals and are known to have one of the highest light outputs compared with other scintillators⁽³⁾. Their anisotropic response is not an issue for measurements in monodirectional neutron fields. Until recently, stilbene (C₁₄H₁₂) crystals produced by the so-called ‘melt grown method’ did not provide a decisive advantage over usual and well-known liquid scintillators, such as a BC501A, for neutron detection below 1 MeV. However, N. Zaitseva⁽⁴⁾ demonstrated in 2015 the possibility of making large stilbene crystals with increased purity using the ‘solution grown’ method.

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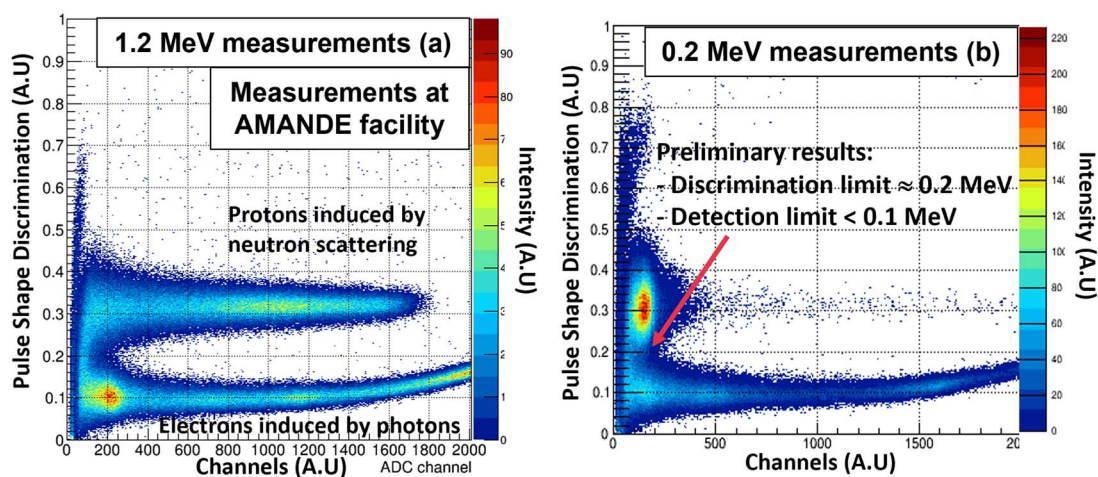


Figure 1. Neutron measurement at AMANDE in a neutron field produced by the ${}^3\text{H}(p,n){}^3\text{He}$ reaction at 2.04 MeV proton energy: (a) at 0° (i.e. with 1.2 MeV neutrons) and (b) at 150° (i.e. in with 170-keV neutrons). The neutron-photon discrimination limit is around 200-keV neutron energy, although the detector is still able to detect recoil protons generated down to less than 100 keV.

Consequently, in 2017, a 25.4 mm \times H 25.4 mm solution-grown stilbene detector assembly was acquired and placed in a neutron field of 565 keV at AMANDE⁽⁵⁾. At that energy, barely measurable with a classical scintillator in the best configuration, the stilbene detector, coupled with an analog acquisition, showed a reduced but still useable neutron/photon discrimination capability.

In 2019, additional tests were carried out at AMANDE with this stilbene detector coupled to a digital acquisition system⁽⁶⁾. These experiments showed that the set-up was capable of detecting neutrons down to less than 100 keV⁽⁷⁾ using the ToF technique.

In 2020, a larger 50.8 mm \times H 50.8 mm solution-grown stilbene detector assembly was ordered to cover the energy range [100 keV–1.5 MeV]. This detector was equipped with a wide light-collection pre-amplifier with reduced amplification in order to improve neutron/gamma discrimination at low energies. Its rise time is relatively long (20 ns) with a temporal resolution of about 1 ns. These temporal performances are not a drawback with AMANDE (beam burst of 2 ns) and for the targeted energy range (neutron velocity of about 70 ns per meter at 1 MeV). Measurements carried out in February 2022 at AMANDE, after optimisation of the digital acquisition parameters and of the photomultiplier high voltage, showed the ability of the detector to work well in the targeted energy range, as illustrated in Figure 1. Below 200 keV, the detection of a recoil proton remains efficient but the lack of clear discrimination limits the detector ability to correctly assess the fluence.

Finalisation of the flight time extension

The extension of the ToF technique at AMANDE between 100 keV and 1 MeV will be carried out between 2022 and 2024 using a pair of stilbene detectors coupled to a digital acquisition system. This work requires the following:

- adapting the experimental set up to each measurement,
- connecting all quantities to the SI (time, length and mass),
- determining the detector efficiency.

Connection to SI

ToF measurements will be made using the digital acquisition time stamp. In order to connect to the SI, it will be necessary to obtain a calibration of the digital acquisition time stamp from a metrology laboratory such as LNE-SYRTE⁽⁸⁾. This calibration principle is rather standard but requires a specific methodology currently under development. The other quantities required for connection to the SI (i.e. length, mass and speed of light) are already connected to the SI at AMANDE. Given the energy limitation of the dedicated low-energy stilbene ([100 keV–1.5 MeV] for neutrons and [30–500 keV] for photons), a second stilbene detector will be used to cover the neutron energy range between 1 and 20 MeV, and up to about 7 MeV energy for photons.

Characterisation of the stilbene detector response functions

The characterisation of a neutron field (i.e. the determination of the neutron fluence energy distribution),

with or without time of flight, requires the neutron response functions. Moreover, every neutron field is accompanied by a photon field. Therefore, the characterisation of photon fields is a necessary step in neutron metrology studies.

Neutron response functions

A recent PhD work⁽⁹⁾ has shown that simulation tools alone are not capable of predicting the full stilbene neutron response function. Indeed, the necessary physics models for characterising and reproducing anisotropy of the neutron response are not yet well known. Therefore, it is necessary to determine the response function experimentally using, for example, a white neutron spectrum. However, the lack of knowledge of the [100 keV–1 MeV] component of the white spectra calls for a specific study.

Photon response functions

The work reported in⁽¹⁰⁾ demonstrated that a stilbene detector could be used as a ‘low resolution’ photon spectrometer using simulated gamma response functions. Here, we report similar work, adapted to the [100 keV–1.5 MeV] neutron measurement range.

Example of low-energy neutron ToF utility at AMANDE

Neutrons production by secondary reactions

Deuterium beams can easily generate ‘undesired’ secondary neutron production reactions in target materials. Examples are the reactions $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ and $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}^*$, which have been observed⁽¹¹⁾ for TiT or TiD targets (i.e. titanium matrix in which tritium (^3H) or deuterium (^2H) is inserted), which were put on a silver backing. Such targets are available at the AMANDE facility. Although we took special care to avoid interactions with carbon or oxygen in the target, we have observed the same neutron-producing reactions. Table 1 shows the energies of the neutrons produced at 0° from the ^{12}C and ^{16}O nuclei as a function of the incident deuteron energy.

ISO 8529-1 standard

Standard 8529-1⁽¹²⁾ recommends several accelerator neutron production reactions to achieve different energies between 8 keV and 19 MeV. At 19-MeV neutron energy, the $^3\text{H}(\text{d},\text{n})^4\text{He}$ production reaction at 0° is recommended. However, the ISO 8529 standard states that, for this energy: ‘*In the 19 MeV field, parasitic neutrons might be present because of the high deuteron energy used to produce this field*’. So, some precautions must be made such as ‘[...] *the time-of-flight technique should be used to check the equivalence*

Table 1. Neutron energy at 0° from secondary reactions on the target nuclei ^{12}C and ^{16}O as a function of incident deuteron energy.

Reaction ¹	Neutron energy at 0° (MeV) ²	Deuteron energy (MeV) ³
$^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$	3.66	[0.33–1.31]
$^{12}\text{C}(\text{d},\text{n})^{13}\text{N}^*$	1.11	[3.08–3.89]
$^{16}\text{O}(\text{d},\text{n})^{17}\text{F}$	2.27	[1.83–2.73]
$^{16}\text{O}(\text{d},\text{n})^{17}\text{F}^*$	1.75	[2.38–3.26]

¹*On the first excited state ²Neutron energy for 4 MeV deuterons. ³First value is the reaction threshold and the second is for 1 MeV neutrons at 0° .

of the targets [...]’. Indeed, this reaction requires a deuteron energy of about 2.6 MeV for a TiT target of $800 \mu\text{g}/\text{cm}^2$. This implies, among other things, the presence of secondary reactions on oxygen at 870- and 305-keV neutron energies at 0° , from the $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}$ and $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}^*$ reactions. These energies are within the energy range targeted by the ToF extension at AMANDE.

Conclusions and perspective

Neutron energy distribution can be measured at AMANDE using the well-known ‘time-of-flight’ method. This technique is usually performed in combination with a liquid scintillator. However, the low scintillator efficiency below 1-MeV neutron energy limits the method application range. This limitation is not dramatic when the production reactions are well known and induced by proton such as ($\text{p} + ^7\text{Li}$ or $\text{p} + ^3\text{H}$). However, in white or complex neutron fields, the poor knowledge of this energy range makes calibration difficult or impossible. It can also make the interpretation of measurements difficult. Recent developments in the production of large and pure organic crystals have made it possible to obtain stilbene scintillators with a decade lower detection threshold (about 100 keV). This opens up the possibility of extending the ToF method from 1 MeV down to 100 keV. Furthermore, these new possibilities could also help better characterise nuclear data of secondary reactions that remain rather poorly known, despite some measurements⁽¹¹⁾ and recent evaluations⁽¹³⁾. Ultimately, the improved knowledge of these secondary reactions should improve the quality of the calibrations carried out in neutron fields generated by a deuterium ion beam.

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