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Editorial board
Nuclear Instruments & Methods in Physics Research

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Dear Editors,

Please find enclosed the manuscript: “PULSE SHAPE DISCRIMINATION OF CLYC SCINTILLATOR COUPLED WITH A LARGE SIPM ARRAY” by Nesrine Dinar, Damiano Celeste, Marco Silari, Alberto Fazzi and Vincenzo Varoli for your consideration for publication in Nuclear Instruments & Methods in Physics Research (section A).

All authors contributed significantly to the submitted work and approved to the revisions made in the manuscript. The results of this research have not yet been published elsewhere and we hope that you will find our manuscript suitable for publication in Nuclear Instruments & Methods in Physics Research (section A). We would like to publish the manuscript with colour figures in the online version, and with black and white figures in the paper version.

The manuscript describes original work and is not under consideration by any other journal. We appreciate your time and look forward to your response.

Yours sincerely,

Nesrine Dinar

Pulse shape discrimination of CLYC scintillator coupled with a large SiPM array

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***Abstract*—This paper discusses measurements performed to evaluate the possibility to build a neutron probe based on a CLYC scintillator coupled with a large SiPM array. The use of a large SiPM array allows optimising the light collection, necessary for spectrometry However, a large SiPM array leads to huge capacitance, whose main impact is to slow down the input signals. Therefore, pulse shape discrimination (PSD) is lost due to the electronic process. To solve this problem, an electronic board was designed to extract the fastest scintillation decay times from the crystal without cutting the crystal response. The impact of using a large SiPM array instead of a PMT is evaluated by comparing the PSDs and the scintillation decay times. A figure of merit (FOM) of 2.3 was found with the SiPM versus a value of 2.64 for the PMT. In addition, the neutron efficiency was measured and compared to the one simulated with the MCNP code. The angular dependence and the linearity range in dose rate were also evaluated.**

Keywords—neutron, scintillator, pulse shaping methods, SiPM, simulation, efficiency.

1. INTRODUCTION

Radiation detectors with dual neutron/γ-ray sensitivity and robust discrimination features are in great demand nowadays to reduce the size and the complexity of conventional systems, generally provided with two different detectors for independent measurements. The advent of the CLYC scintillator material has drawn the attention of the scientific community due to its excellent energy resolution, sensitivity to both neutrons and γ-rays and pulse shape discrimination (PSD) features. CLYC has a density of 3.31 g/cm^3 and a peak-emission wavelength around 373 nm, which well matches the response of many commercially available photomultipliers (PMTs). In the framework of the B-RAD project, the development of a radiation survey meter capable of operating in the presence of a strong magnetic field [1], we started the development of a portable neutron probe based on a CLYC coupled with a SiPM array. The choice of SiPMs is justified by crucial requirements: the insensitivity to external magnetic fields and the low operating voltage with respect

to PMTs. The gain is similar to a PMT. The small size allows building an extremely compact, light and robust mechanical device. The PSD performance and decay times of the CLYC are evaluated with the SiPM and compared to those obtained with a PMT. The measured and simulated efficiencies are also compared. The linearity in dose rate as well as the angular response of the CLYC are evaluated.

2. EXPERIMENTAL DETAILS

2.1. Material

A cylindrical CLYC crystal 25.4 mm in diameter and 25.4 mm thick from Radiation Monitoring Devices [2] and a SiPM array J-30035-64P-PCB from SensL [3] were used. A CLYC crystal with the lithium element enriched in ^{7}Li at >99% is used in this work. This enrichment suppresses the sensitivity to thermal neutrons allowing a better detection of fast neutrons through the $^{35}Cl(n,p)^{35}S$ and $^{35}Cl(n,\alpha)^{32}P$ reactions. The Q values of these reactions are 615 keV and 937 keV, respectively. The energy deposition of the products linearly depends on the energy of the incident neutron. Thus neutron spectrometry without resorting to an unfolding code is in principle possible up to 8-10 MeV. Above this energy, multiple reaction channels and excited states result in a spread out of the response function [4]. Since the CLYC is hygroscopic, a cylindrical housing protects it from moisture. The SiPM array is made up of sixty-four 3x3 mm^2 silicon elements, each consisting of 5676 APD-cells (Avalanche Photo Diodes of 35x35 µm^2 size). The whole size of the array well matches 1-inch cylindrical scintillators. The response of the CLYC+SiPM was compared with that obtained with a PMT R6231-100 from HAMAMATSU [5].

Taking into account the SiPM standard output, the slow response of the electronics prevents distinguishing the initial differences in the shapes of the n/γ signals from the CLYC. To achieve this goal, we had to increase the speed of the signal by trying to extract the fastest scintillation times from the crystal without cutting its response. The method consisted in compensating the slow component of the signal by exploiting the pole-zero cancellation method. An RC circuit is implemented in series, after the amplification stage, to compensate for the pole due to the high capacitance of the photo-detector and to introduce a new pole at higher frequency. Following this idea, a first prototype of the circuit was built (Figure 1).

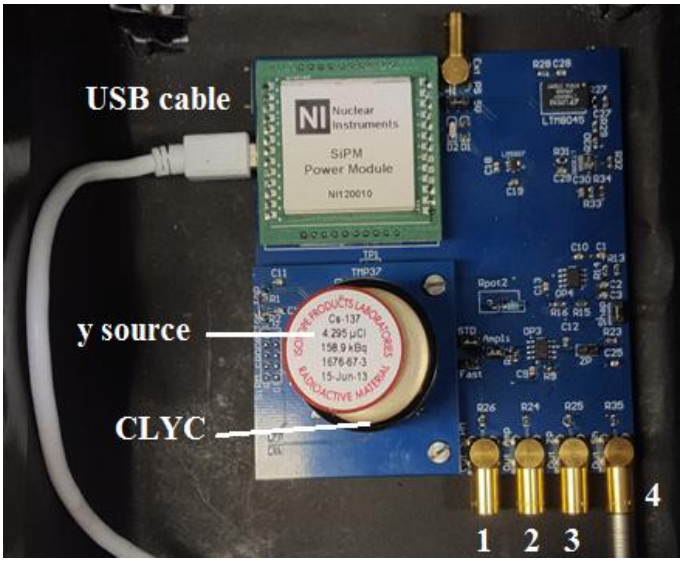


Fig. 1 Prototype of the neutron probe

2.2. PSD

A ^{252}Cf source was used to study the n/γ discrimination. About 100,000 signals were recorded and analyzed off-line. The first step for the PSD analysis was to align all the waveforms to a common zero. However, unlike in the case of the PMT signal output, the Savitzky–Golay low pass filter was applied to increase the signal-to-noise ratio without distorting the signal. Several algorithms for PSD exist, in this work the charge integration method was employed [6]. The principle of charge comparison is to integrate two parts of each pulse: the charge of the entire pulse (Q_{total}) and the charge in the tail (Q_{tail}). The difference in the ratio of the Q_{tail} to the Q_{total} (the Q_{ratio}) is calculated and used to discern which type of radiation generated the pulse.

A figure of merit (FOM) is used to establish the ability to discriminate between pulses generated by photons and neutrons. The FOM is calculated from the histogram of the Q_{ratio} versus peak height data assuming that the pulse distributions are Gaussian. The FOM is calculated after a PSD has been performed, measuring the width and locations of the peaks in the histogram of the pulse shape data. The choice of the two timing intervals, at which the integration of the pulse should be measured, is based upon a series of measurement that optimize the discrimination.

2.3. Neutron efficiency

To estimate the neutron detection efficiency, the scintillator was irradiated with AmBe source neutrons in the calibration laboratory (CALLAB) of the Radiation Protection group at CERN [7]. The activity of the source was 100 GBq emitting 6.4×10^6 neutrons/s. The intrinsic efficiency was defined as the ratio between the neutron fluence detected by the CLYC

and the neutron fluence incident on the crystal. The CLYC was placed at 135 cm distance from the source. The data acquisition was set to approximately 1h30 to obtain the maximum number of files that can be registered by the Lecroy HDO6104 oscilloscope (100,000) and analysed off-line. The measured efficiency was compared to the one simulated with the Monte Carlo N-Particle 6 (MCNP6) code developed by Los Alamos National Laboratory [8], to obtain a theoretical estimate of the efficiency of CLYC to fast neutrons. In the input file, surface, cell and data cards were defined. A 1 inch right cylinder made of CLYC enriched with 99% ^{7}Li, 75% ^{35}Cl and 25% ^{37}Cl was simulated. The evaluated nuclear data file ENDF/B-VII.0 was implemented for these calculations [9]. The scintillator was placed in a concrete room of 13 x 13 x 13 m^3 volume that modelled the CALLAB geometry [10]. The neutron energy spectrum was generated according to the AmBe energy binning defined in the ISO-8529-2 norm [11]. The energy spectrum of the emitted neutrons is a continuum up to around 10 MeV. The source definition was a point source. The number of source particles used in the simulation was 10^7, sufficient to achieve low statistical uncertainty while minimizing the computing time.

Neutron interactions within CLYC-7 result in the production of protons, alpha particles and deuterons [12].The tallying of these products enables evaluating the neutron interactions. The F8 Pulse Height Tally, which records the energy deposited in a cell by each source particle and its secondary particles, was used. The F8 tally was associated to pulse height light (PHL) treatment in order to model the pulse height energy spectrum of the CLYC detector [8]. The PHL tally treatment allows for the conversion of the energy deposition (F6 tally) into detector pulse height (F8 tally).

The intrinsic efficiency was calculated as the ratio between the total number of particles created inside the CLYC (protons, alpha particles and deuterons) and the number of neutrons impinging on the CLYC surface.

2.4. Scintillation decay times

The neutron and photon signals were averaged and normalized to their peak amplitude according to the highest FOM, to produce the so-called standard pulse. The standard pulses contribute to eliminate noise fluctuations in individual signals and reveal the details on the prompt and delayed shape. To study the decay time under photon and neutron irradiations, the standard pulses were fitted with exponential decay curves, each decay time constant corresponding to a specific scintillation mechanism [13]. For Ce3+ doped crystals, up to four mechanisms contribute to the scintillation corresponding to four different energy transfers, which occur at different timescales:

- Direct e-/hole capture by Ce^{3+} (Ce^{3+}): after absorbing a γ-ray, free electrons and holes are created in the conduction band and valence band, respectively. These free electrons and holes may be trapped by a Ce^{3+} ion and the observed decay

time is characteristic of the lifetime of the excited state of Ce^{3+}. This type of fast energy transfer is desired for impurity Ce^{3+} activated inorganic scintillator materials.

- Binary e-/hole recombination (V_k): instead of being captured by the Ce^{3+} ion, the hole produced initially in the valence band is unstable and will create a molecular complex called V_k center.

- Self-trapped exciton (STE) diffusion/emission: if the V_k centre is captured by an electron from the conduction band, before being trapped by a Ce^{3+} ion, a STE is formed. STE is luminescent itself and has a lifetime of several microseconds.

- Core-to-valence luminescence (CVL): in case of chloro elpasolite (such as the CLYC), a fourth mechanism may occur. When the incident high-energy photon excites an electron from the upper core band into the conduction band, the resulting hole is short lived and will recombine with an electron from the valence band. If the process is radiative, it is called CVL. CVL is characterized by a short decay time (0.6 to 3 ns).

2.5. Linearity in dose rate

The count rate linearity of a CLYC + photodetector system is determined by its time resolution, which is defined as the minimum time between successive scintillation pulses that the detection system can distinguish. This time interval may be limited by the decay time of the scintillator, the frequency characteristics of the signal processing circuit, and the timing characteristics of the photodetector's electronics. The decay time of the scintillation pulses from CLYC detectors is as long as a few microseconds, resulting in significant pulse pile-up at moderate input count rates.

To study the linearity range of CLYC and the effect of SiPM (instead of traditional PMT) on this range, measurements were performed with γ-sources. The main component of the output signals are from γ-interactions with the crystal, the neutron efficiency being <1%. Therefore, the saturation will occur with γ-signals. The discriminator threshold affects the linearity range. Thus, in order to compare the linearity range of the CLYC when coupled with the SiPM or PMT, the same discriminator threshold (330 keV) was fixed through an energy calibration measurement. Several waveforms from ^{137}Cs and ^{60}Co sources were registered with the oscilloscope. Data were then manipulated via software (Python script) in order to obtain source spectra. Four ^{137}Cs sources of different activities were used (300 MBq, 3 GBq, 30 GBq and 300 GBq). The dose rate was changed by modifying the distance to the source and the activity of the source.

2.6. Angular dependence

The energy states of molecules consist of a series of discrete levels, which affect the scintillation mechanism in inorganic materials. Therefore, the mechanism is dependent on the crystal lattice, which means that the response of the scintillator can be anisotropic. To study the angular response of the detector, the CLYC was coupled with the PMT and irradiated in a pure γ (^{137}Cs) and a mixed γ/neutron (AmBe) field. The system CLYC + PMT was placed at different angles from -90º to +90º at interval of 30º (see Figure 2).

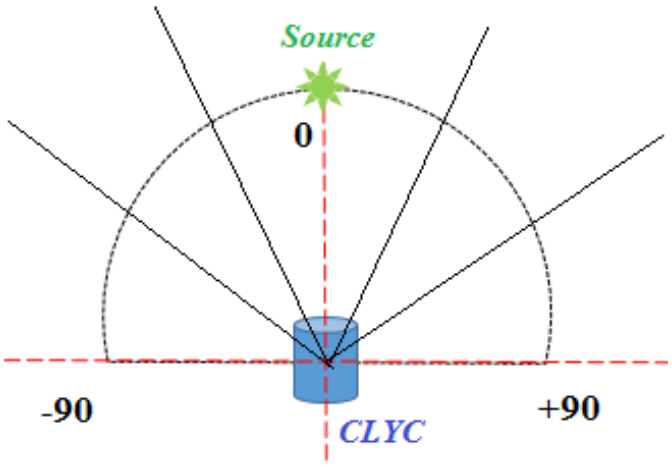
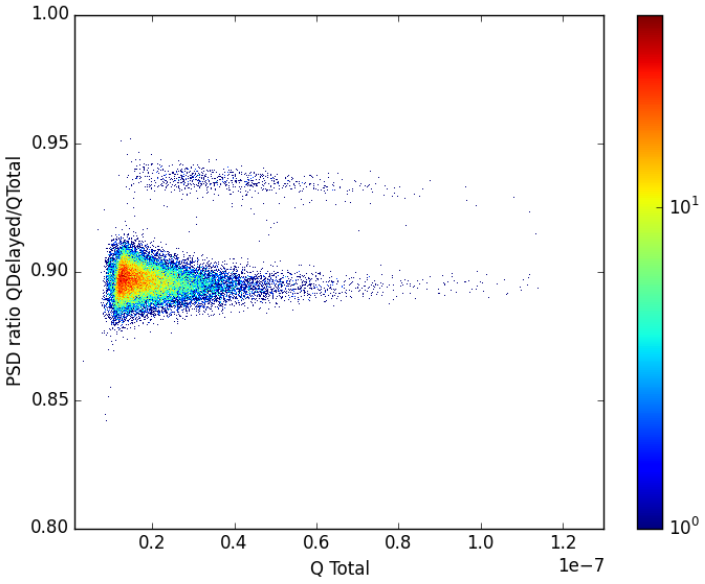


Fig. 2 Schematic of the irradiation set-up for determining the angular response of the CLYC.

3. RESULTS AND DISCUSSION

3.1. PSD

With a cut at the Q value of the neutron reaction (615 keV) the best FOM are 2.97 (PMT) and 2.30 (SiPM). Figure 3 shows the two-dimensional PSD plot obtained with the CLYC coupled with the SiPM and the specific board (see Figure 1). Two populations are clearly visible, the upper one corresponding to neutrons. The ^{252}Cf neutron spectrum is extracted by integrating the neutron signals and it is shown in Figure 4. For a qualitative comparison, Figure 5 shows the ^{252}Cf reference neutron spectrum of ISO8529-1. The first approximation of the measured neutron energy distribution is coherent.



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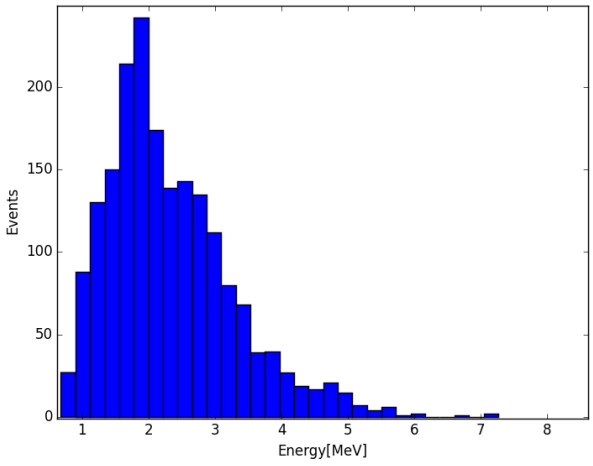
141 Fig. 3 2D PSD plot with γ (lower) and neutron (upper) populations.



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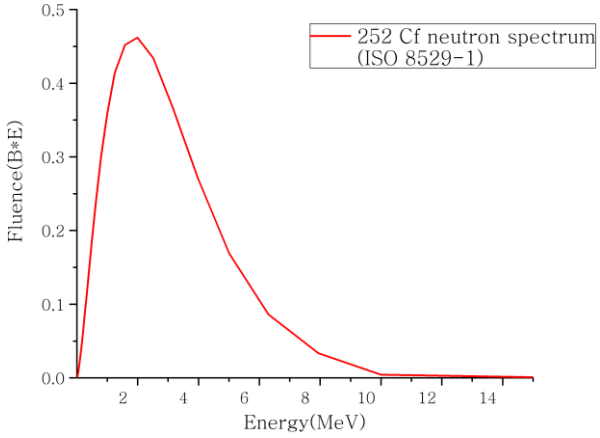
143 Fig. 4 ^{252}Cf neutron spectrum from CLYC coupled with the SiPM array J-30035-64P-PCB.



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145 Fig. 5 ^{252}Cf reference neutron spectrum from ISO 8529-1.

3.2. Neutron efficiency

The optimal time window to optimize the FOM was applied (90 ns - 1 µs) and only the neutron Gaussian peak was integrated from the projection of the PSD ratio on the x-axis. The neutron intrinsic efficiency was found to be 0.8% for an AmBe source. To obtain a calibration factor the fluence was folded with the fluence to dose conversion factor [11]. An experimental calibration factor of 12 nSv/count was found. This value is in excellent agreement with both the simulated efficiency (0.7% equivalent to a calibration factor of 11 nSv/count) and with literature data ([14] and [15] giving 0.7% and 0.8%, respectively).

3.3. Scintillation decay times

Table 1 presents the results of the fitting procedure. For the PMT, the gamma waveform reveals four decay times: 2 ns (T1), 48 ns (T2), 637 ns (T3) and 4.8 µs (T4). Considering the two exponential decay fits, the neutron waveform at room temperature presents a component with 598 ns (T3) and 4493 ns (T4). Correlating these results with the literature, these mechanisms may be attributed to T1=CVL, T2=Ce^{3+}, T3=V_k, T4=STE+Ce^{3+} [13].

To study the influence of the photodetector response on the CLYC decay times, the CLYC was coupled with the SiPM. As with the PMT two decay times were found for the neutron waveform (517 ns and 4.2 µs). These times are comparable to the PMT (598 ns and 4.4 µs) and can be attributed to the same mechanism. However only three decay times were found for the γ-pulse. The T1 component of the signal due to CVL is absent. Moreover the T2 component is slower with the SiPM than with the PMT (131 ns against 48 ns). The slow recovery time of the SiPM is responsible for the disappearance of T1 and the decrease of T2 of the measured waveforms. This result shows that only the fast component of the signal (hundred ns) is slowed down by the SiPM response.

Table 1. Measured decay times for γ and neutron induced reactions on CLYC at room temperature.

		T1 (ns)	**T2 (ns)**	**T3 (ns)**	**T4 (ns)**
PMT	γ	2	48	637	4821
	Neutron	-	-	598	4493
SIPM	γ	-	131	494	4215
	Neutron	-	-	517	4208

Figure 6 shows the photon and neutron standard pulses measured with the PMT and the SiPM. The neutron waveforms are quite similar but the absence of CVL is clear on the photon signal obtained with the SiPM. The difficulty to

discriminate the γ and neutron signals when the CLYC is coupled with the SiPM was reported by several authors (see for example ref. [16]). However, although the fast components are affected by the SiPM and consequently are responsible for the reduction in the PSD capability as compared to a PMT, the PSD is still possible with an excellent FOM of 2.3.

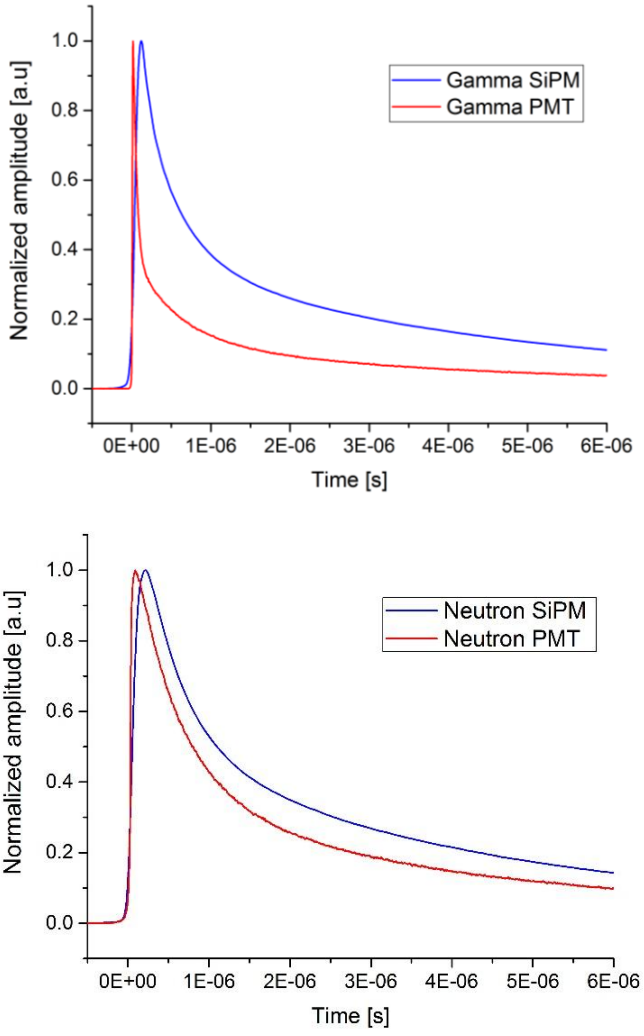

Fig. 6 Neutron and γ signals with PMT (red) and SiPM (blue).

3.4. Linearity in dose rate

Figure 7 shows the count rate as a function of dose rate for both the PMT and SiPM at the same threshold. For clarity, only the fit to the PMT data points is shown. The saturation appears at about 1 mSv/h for the SiPM while the PMT is still linear. This discrepancy may be explained by the difference in the timing of the signals. As seen in Figure 6, the γ signals with the PMT are faster than the ones obtained with the SiPM. The larger the signal width the higher the probability of pile up effects, thus the range of linearity of the detector is shortened.

3.5. Angular response

Figures 8 and 9 show the angular response of the CLYC when irradiated with ^{137}Cs and AmBe sources, respectively. The first effect seen is that the response of the CLYC is symmetric around its 0º axis. For example the count rates at -30º and +30º are very similar. Another effect is that the count rate increases with angle (up to 17%), the minimum being at 0º. This effect can be attributed to the smaller solid angle provided by the cylindrical geometry of the sensitive volume when viewed head-on. Indeed the difference of solid angle when irradiating the CLYC head on as compared with irradiating it side on is about 20%.

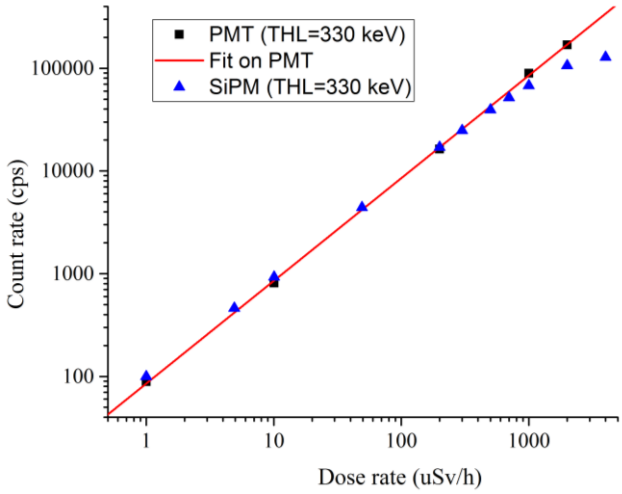

Fig. 7 Count rate as a function of dose rate obtained with the SiPM (blue) and PMT (black) at the energy threshold of 330 keV. The red line represents the linear fit to the PMT data.

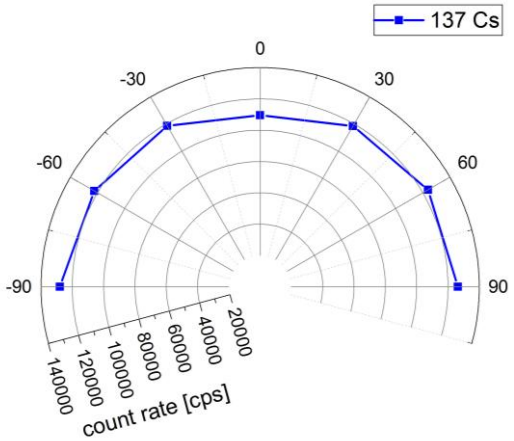

Fig. 8 Directional response of the CLYC to a ^{137}Cs source.

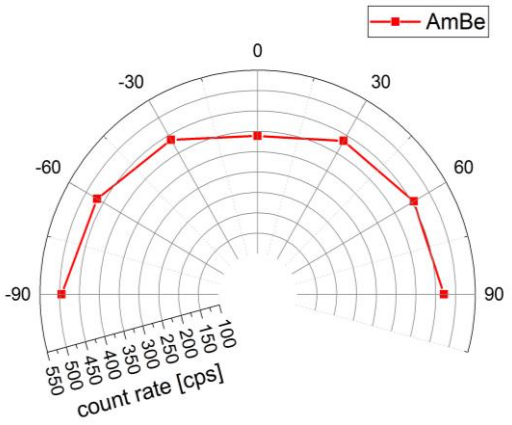

Fig. 9 Directional response of the CLYC to an AmBe source.

4. CONCLUSIONS

A CLYC scintillator was investigated for its potential use in fast neutron detection/spectrometry. The CLYC was coupled to a SiPM as well as a PMT and the results were compared.

The capacity of the CLYC+SiPM to discriminate γ and neutron signals was the more challenging part when dealing with such a large array, since the total capacitance increases with the number of pixels. The extraction of the fastest scintillation decay time constants from the crystal and the shortening of the falling time of the signal without cutting the crystal response was achieved through the development of specific electronics. An excellent FOM of 2.3 was found approaching the result obtained with PMT. Shortening the signals allows increasing the linearity range and the frequency of the events at which the CLYC is sensitive.

The system CLYC+SiPM is linear in dose rate up to 1 mSv/h where it starts showing a slight saturation effect. The angulation response was measured with both γ and neutron sources. A variation of 20% in count rate was found between front and lateral irradiation (0° and 90°).

Finally, the fast neutron efficiency was simulated with MCNP and the results were validated experimentally. Similar efficiency values of approximately 0.8% were found (i.e about 0.2 counts per second for a dose rate of 1 uSv/h). The CLYC could be used as a stationary detector to monitor both γ and neutron dose rates in workplaces. Another potential application for the CLYC is its use for homeland security (non-proliferation, border security) to improve the detection of special nuclear materials (SNM). Spontaneous and induced fission events in SNM produce neutrons and γ-rays, which

can be detected and well separated thanks to the excellent discrimination capabilities of the CLYC. The fission products have an energy between 500 keV and 10 MeV that fit the measuring range of the CLYC. CLYC neutron detectors can therefore be used as dual mode to detect γ-ray and neutron in SNM search and monitoring applications.

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