Characterization of new scintillators: SrI$_2$:Eu, CeBr$_3$, GYGAG:Ce and CLYC:Ce

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Abstract.
We measured the performance of a 2”x2” tapered cylindrical SrI$_2$:Eu, a 2” x 3” cylindrical CeBr$_3$, a 2” x 0.3” cylindrical GYGAG:Ce. The gamma-ray energy resolution was measured up to 9 MeV. A scan along three axes of each scintillator was performed using a collimated source of $^{137}$Cs. The signals of the detectors were also digitized and compared. We tested two 1” x 1” cylindrical CLYC crystals. One was enriched with $^6$Li at 95% while the other was enriched with 99% of $^7$Li. The response of the two detectors to gamma rays, thermal and fast neutrons was measured. The PSD performances have been tested with a different types of PMTs.

1. Introduction

There is an intense R&D work on CeBr$_3$ and SrI$_2$:Eu crystals as larger crystals are becoming available [1-2]. The development of new ceramic scintillator materials (such as GYGAG:Ce) offers the possibility to have high-resolution gamma-ray spectroscopy at low cost [3]. The scintillator Cs$_2$LiYCl$_6$:Ce (CLYC) is a very interesting material for radiation detection because of its ability to measure gamma rays and neutrons simultaneously [4]. CLYC scintillators are suitable for thermal neutrons detection, owing to $^6$Li content and they can also be used as fast neutron spectrometers, owing to $^{35}$Cl content. The gamma rays and neutrons can be discriminated by pulse shape discrimination methods [5].

In this work the results of the characterization measurements of SrI$_2$:Eu, CeBr$_3$ and GYGAG:Ce are described in section 2. The measurements performed with two samples of CLYC scintillators, characterized by different Li ions concentrations, are discussed in section 3; in particular, the general properties, the fast and thermal neutron detection capability, and the PSD performances with different PMTs are presented in section 3.1, 3.2 and 3.3, respectively. The conclusions are given in section 4.
2. Characterization of SrI$_2$:Eu, CeBr$_3$ and GYGAG scintillators sample

The SrI$_2$:Eu and the GYGAG:Ce samples have been provided by the Lawrence Livermore National Laboratory while the CeBr$_3$ crystal by the Institut de Physique Nucleaire Orsay. The measurements were performed in the gamma spectroscopy laboratory of the University of Milan. The 2” x 2” cylindrical tapered SrI$_2$:Eu and the 2” x 3” cylindrical CeBr$_3$ were coupled to a HAMAMATSU R6233-100sel PMT powered at 800V and 850V, respectively. The 2” x 0.3” GYGAG:Ce was coupled to a HAMAMATSU R6231-100mod (HV=800V). The used Voltage Dividers (VD) were HAMAMATSU E1198-27 for all the detectors. Since CeBr$_3$ signal is very similar to that of LaBr$_3$:Ce, for this scintillator we used as well the LABRVD. This dedicated VD was developed by the electronic workgroup of the University of Milan for LaBr$_3$:Ce scintillators to reduce the PMT induced non-linearity at high energies [6].

The scintillation response was measured using standard gamma ray sources ($^{22}$Na, $^{60}$Co, $^{88}$Y, $^{133}$Ba, $^{137}$Cs, $^{152}$Eu) and an AmBe(Ni) composite source. The signals of the three detectors were sent to a spectroscopic amplifier (TENNELEC TC244) and, in turn, to an ADC (ORTEC ASPEC MCA 926). The $^{152}$Eu spectra, acquired with all tested scintillators, are compared in Fig. 1, while the AmBe(Ni) energy spectra are compared in Fig. 2. At 662 keV we measured a FWHM-energy resolution equal to 4.0%, 4.4% and 5.2% for the SrI$_2$:Eu, the CeBr$_3$ and the GYGAG:Ce respectively[7].

![Figure 1](image1.png) **Figure 1.** The $^{152}$Eu energy spectra acquired using a standard spectroscopic amplifier for the three tested scintillators. The SrI$_2$:Eu has the best energy resolution among these scintillators. The spectra are normalized on the area of the 344 keV peak of the $^{152}$Eu source.

![Figure 2](image2.png) **Figure 2.** The measured SrI$_2$:Eu, GYGAG:Ce and CeBr$_3$ acquired using a PMT, a standard spectroscopic amplifier and an ADC. The used sources were AmBe(Ni), $^{88}$Y and $^{60}$Co (only for CeBr$_3$).

The detector anode pulses were also digitized using a 12 bit LeCroy HDO 6054 oscilloscope. A set of pulses (~1000) at a fixed energy were averaged to produce the reference signals. The signal properties (rise time and fall time) of all detectors were compared and the signals of each scintillator were studied from 662 keV up to 9 MeV. Fig. 3 shows the comparison of the three scintillator signal shapes, at 662 keV. The CeBr$_3$ provides the fastest signal: it has a rise time of ~18 ns and a fall time of ~70 ns. The SrI$_2$:Eu has the slowest signal among the tested detectors (fall time ~7 μs).

No significant change in the GYGAG:Ce and SrI$_2$:Eu signal shapes up to 9 MeV, whereas a small change was seen in CeBr$_3$ at 9 MeV of energy [7].
Figure 3. The normalized pulses of the three detectors for 662 keV gamma rays. The CeBr$_3$ has the fastest signal among these scintillators, whereas SrI$_2$:Eu has the slowest one.

The energy spectra and the pulses were also measured using a collimated beam of 662 keV gamma rays scanning the detector along the x, y, and z axes. A non-collimated $^{88}$Y source, providing two calibration points, was placed nearby and used as a reference. It was possible to study how the position of the centroid, the FWHM, and the area of the 662 keV peak changes as a function of the position of the incident radiation. The detector response was measured to be constant for the CeBr$_3$ and the GYGAG:Ce. No significant variations of the peak position or the FWHM were observed. While SrI$_2$:Eu detector shows variations in the FWHM along the z axis imputable to the self-absorption effects, as shown in Fig. 4. The SrI$_2$:Eu provides the best energy resolution among the studied detectors (4.0 % at 662keV) but this value strongly depends on the position of the gamma-ray interaction point. A FWHM value as good as 22 keV at 662 keV (3.3%) was measured with the collimated source, as shown in Fig. 4 [7].

Figure 4. The FWHM of 662 keV peaks from a collimated $^{137}$Cs source on the SrI$_2$:Eu crystal. The different values were obtained by moving the collimated beam along the z axis of the crystal. The spectra were calibrated using a non-collimated $^{88}$Y sources that was placed nearby as a reference.

3. Characterization of two 1” x 1” CLYC scintillators

Two crystals were used in this work: a CLYC enriched at 95% of $^6$Li (CLYC-6) and a CLYC with an enrichment of $^7$Li larger than 99% (CLYC-7). Both crystals were produced by RMD and have a cylindrical shape, a diameter of 1” and a thickness of 1”. By visual inspection the crystals present some small internal structures. However, they do not seem to affect the energy resolution. These two samples of CLYC scintillators were coupled to a HAMAMATSU R6231-100mod PMTs and to standard voltage dividers (HAMAMATSU E1198-26 and HAMAMATSU E1198-27 for CLYC-6 and CLYC-7, respectively).

3.1. General Properties

The CLYC scintillators exhibit good energy resolution. We measured 4.8% and 4.5% at 662 keV for CLYC-6 and CLYC-7, respectively. The CLYC scintillators can identify and measure gamma-rays
and neutrons via pulse shape discrimination. The CLYC internal radiation was measured to be at least two order of magnitude smaller than that of an equivalent LaBr$_3$:Ce detector [8].

3.2. Thermal and fast neutron identification

The thermal neutrons were measured with both detectors, using an AmBe source. The spectra of Fig. 5 and Fig 6 were acquired in the same conditions and are normalized on the $^{137}$Cs peak. The 3.2 MeV peak, induced by thermal neutrons, is practically absent in the CLYC-7 spectrum (see Fig. 6), but it is visible in the CLYC-6 one (see Fig. 5). The thermal neutron detection efficiency for the CLYC-7 is 0.3% with respect to the CLYC-6 sample of identical size.

![Figure 5](image)

**Figure 5.** The energy spectra measured with CLYC-6 and a AmBe(Ni) source in a paraffin box. A $^{60}$Co and a $^{137}$Cs were also present.

![Figure 6](image)

**Figure 6.** The energy spectra of CLYC-7 acquired in the same condition of the spectra of Fig. 5.

Both crystals are capable to detect, identify and measure the kinetic energy for fast neutrons. The ability to separate between neutron and gamma-rays does not depend on the neutron energy while the measurement of neutron kinetic energy using the CLYC scintillators can be performed up to approximately 8 MeV. In the case of neutrons of higher kinetic energy three body reactions have a larger cross section of the reaction on $^{35}$Cl. A continuum energy spectrum was measured caused by a superimposition of different reaction mechanisms [8].

3.3. PSD with different PMTs

The CLYC light emission has a fast and slow component related. The fast component, mainly in the UV-blue, is significant only when gamma rays are measured and it may be used for effective pulse shape discrimination. Timing or spectroscopic PMTs provide different responses to a fast signal because of their intrinsic rise time; in addition, the entrance window (quartz or borosilicate glass) filters in a different way the CLYC fast emissions.

The CLYC-6 crystal was coupled to the different PMTs and irradiated using gamma-rays sources and the AmBe source. The used PMTs are all from HAMAMATSU: i) H6533 (timing PMT with borosilicate glass window); ii) R2059 (timing PMT with quartz window); iii) R6233-100sel (spectroscopic PMT with borosilicate glass window) and iv) R6231-100mod spectroscopic PMT with quartz window. The fast component is more visible for PMTs with fast timing response (R2059 and H6533). A figure of merit (FOM), defined as the difference between the centroid of the neutron and gamma peak divide by the sum of the FWHM of the two peak, by projecting a PSD matrix, like the
one of Fig. 7, along the y axes. The FOM describes the capability to discriminate between gamma rays and neutrons and it ranges from 2.8 to 3.4 was found. It is important to point out that the FOM of a $^3$He tube is $\sim 2$. In optimized conditions using custom electronics and digital filters FOM of 3.8 was found for a HAMAMATSU R6231-100 PMT, as shown in Fig. 7 [8].

![Figure 7](image_url)

**Figure 7.** The PSD matrix of CLYC-6 coupled with an HAMAMATSU R6231-100 mod PMT. The logarithm of the counts of the matrix is plotted on the z axis. The measured FOM is 3.8.

4. Conclusions

In this work, GYGAG:Ce (0.3” x 2”), SrI$_2$:Eu (2” x 2”) and CeBr$_3$ (3” x 2”) samples were characterized by studying the pulse shape, the energy resolution and the crystal responses to a collimated source. The SrI$_2$:Eu has the best energy resolution but it presents a strong self-absorption. The CeBr$_3$ has no internal activity and a fast signal, similar to LaBr$_3$:Ce. The GYGAG:Ce seems to be a very good detector, with high efficiency, good energy resolution and the possibility to be produced in every dimension and shape. Moreover, two samples (enriched with $^6$Li and $^7$Li) of CLYC scintillators were tested with gamma rays, thermal and fast neutrons. The performances of CLYC scintillators with different PMT were measured.

References

[1] Cherepy N., et al., 2013, *IEEE Tran. on Nucl. Science*, 60, 955-958.
[2] Quarati F. G. A., Dorenbos P., van der Biezen J., Owens A., Selle M., Partther L and Schotanus P., 2013, *Nucl. Instr. and Meth. A*:729, 596-604
[3] Cherepy N., et al., 2013, *IEEE Tran. on Nucl. Science*, 60, 2330-35.
[4] Glodo J., Hawrami R. and Shah K.S., 2013 *J. Crys. Growth*, 379, 73–78
[5] D’Olympia N., Chowdhury P., Lister C. J, Glodo J., Hawrami R., Shah K. and Shirwadkar U., 2013, *Nucl. Instr. and Meth, A* 714, 121-127.
[6] Riboldi S., et al., 2011, *Nucl. Science Symp. Conf. Record* (NSS/MIC), 776-778.
[7] Giaz A., Hull G., Fossati V., Cherepy N., Camera F., Blasi N, Brambilla S., Coelli S., Million B. and Riboldi S., submitted to *IEEE Trans. on Nucl. Science.*
[8] Giaz A. et al., in preparation to be submitted to *Nucl. Instr. Meth. A.*