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D9.1 - STATUS OF NEW SCINTILLATOR MATERIALS AND THEIR BASIC CHARACTERISATION

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LIST OF ACRONYMS AND ABBREVIATIONS

$LaBr_3$:Ce	A transparent scintillator material that offers the best energy resolution obtained so far.
$CeBr_3$	An alternative to LaBr, not exhibiting the internal radiation of La
CLYC	Cs_2LiYC_6:Ce crystal belonging to the elpasolite scintillator family
CLLB	$Cs_2LiLaBr_6$(Ce)) scintillator material sensitive to gamma and neutron radiation
CLLBC	$Cs_2LiLa(Br6)_{90\%}(Cl6)_{10\%}$ elpasolite crystal with excellent neutron and gamma radiation response
SPring8	Laboratory near the city of AIOI, Japan. http://www.spring8.or.jp/en/
NewSubaru	Facility inside SPring8 http://www.spring8.or.jp/en/about_us/whats_sp8/facilities/accelerators/new_subaru/
HECTOR+	High Energy detector, array of 10 LaBr3:Ce detectors
PMT	Photo Multiplier Tube
VD	Voltage divider
PSPMT	Position-Sensitive PMT

Executive Summary

In this JRA, we exploit novel scintillator materials and explore new techniques and concepts such as phoswich detectors and segmented or hybrid scintillators. We focus on developing the capability to detect high-energy gamma rays, neutrons and charged particles simultaneously. The emphasis is on a modular approach both in the scintillator crystals and photosensors as well as in the electronics, where improved throughput and effective data processing will allow for compact scalable devices. This JRA will also investigate the use of this technology, developed for basic science, in societal applications within, for example, the areas of nuclear medicine and homeland security. Depending on the applications, features like energy resolution, position sensitivity, high-rate capability, and insensitivity to magnetic fields or to radiation (radiation hardness) are of different levels of importance.

This deliverable is related to Task 1, Novel Scintillator Materials. The work presented here has mainly been performed by the participating groups of INFN (F. Camera) and CNRS (G. Hull) with some inputs from the rest of the collaboration.

As was discussed in the project application a large number of new promising scintillator materials are becoming commercially available. These new materials have to be characterised with gamma rays, charged particles and neutrons, in large volume. Furthermore, since most of them are highly hygroscopic, they need to be encapsulated. The features of these new materials make it difficult to produce working detectors out of them.

The performances of the new materials, for possible nuclear physics applications, are not well known and thus require specific characterisation. The result of such study is particularly interesting for the companies that produce new scintillator materials, as often they cannot perform such tests themselves, as this requires infrastructures that only Universities and Research Centres possess.

In this first delivery, several new crystal materials have been obtained commercially, and prototypes have been constructed in order to be able to test the material as possible detectors for nuclear science.

Introduction

A number of new promising scintillator materials are recently becoming commercially available. These new materials have to be characterised with gamma rays, charged particles and neutrons, in large volume (at least 1”x1”). Furthermore, since most of them are highly hygroscopic, they need to be encapsulated using the right wrapping depending upon the need for best resolution or highest efficiency. These features of the materials make it difficult to produce working detectors out of them. The performances of different crystals have first to be characterised, e.g., according to energy resolution for gamma rays as function of energy, and possible gamma-neutron discrimination etc.

We report here on some new crystal materials that have been obtained commercially, from which prototypes have been constructed in order to be able to test the material as possible detectors for nuclear science. More details can be found in our articles published in Nuclear Instruments and Methods A, see publications further down.

Section 1 Crystal characterisation

The performances of new scintillator materials, and their possible use as detectors, are not well known and thus require specific characterisation. The results of such studies are of particular interest not only for the researchers but also especially for the producers of these materials in order to be able to make and sell detectors that can be widely used including in applications. The delivery of different materials and especially crystals in sizes useful for building detectors is still a relatively slow process, and therefore this activity will probably continue beyond the ENSAR2 project.

The collaboration has been and is continuously performing a survey of the commercial availability of new scintillator materials. In the first half of 2016, it was possible to order $CeBr_3$ (maximum size 3”x3”) and CLYC (maximum size 2”x2”) while in the second half of 2016 CLLB (1” diameter – 1” height), CLLBC (1” diameter - 1” height) and larger volume ^{7}Li-enriched CLYC scintillator (3” diameter - 3” height) became available as well. In the beginning of 2017, it has also been announced that Co-doped $LaBr_3$:Ce (1” diameter - 1” height) has become commercially available and therefore will be ordered.

The activity has therefore focused on:

a) The preliminary measurements of the CLLB (1” diameter - 1” height) performances (see Subsection 1). Two crystals, of different size, have been acquired by the CNRS unit (with ENSAR2 funds). The smaller volume crystals were delivered in January 2017 and they are now under test. The preliminary results will be presented in Subsection 1. The delivery of the larger volume crystal is expected in March 2017.

b) The analysis of the data focused on the measurement of the fast-neutron detection efficiency of two 1” diameter - 1” height CLYC crystals (one enriched with ^{6}Li and one enriched with ^{7}Li). See Subsection 2.

c) The analysis of the data relative to the measurement of the position sensitivity of a 3” x 3” $LaBr_3$:Ce crystal was performed with the aim of correcting for the Doppler broadening. The crystal has diffusive surfaces to measure medium or high-energy gamma rays (0.5 MeV < $E\gamma$ < 20 MeV). It is important to remember that two of the PSPMT had been borrowed by the CSIC unit of the PASPAG JRA. See Subsection 3.

d) The measurement of the response of large volume (3.5” diameter - 8” height) $LaBr_3$:Ce detectors to almost monochromatic gamma rays from 5 to 38 MeV. See Subsection 4.

e) The acquisition by the INFN Milano unit of a CLBBC crystal (bought with INFN funds). The crystal did not arrive yet and it is expected in spring 2017.

f) The acquisition by the INFN Milano unit of a 3”x3” ^{7}Li-enriched crystal (bought with INFN funds). The crystal is expected in February 2017.

Subsection 1.1 Study of CCLB crystals

The CLLB $Cs_2LiLaBr_6$(Ce) scintillator is a material sensitive both to gamma and neutron radiation. One CLLB crystal, 1" diameter – 1" height, arrived to the CNRS group in January 2017. The study of the detector response to gamma rays and neutrons is currently ongoing. The energy resolution as a function of the irradiation energy, in the range between 85 keV and 1408 keV, is presented in Fig. 1 in comparison with the energy resolution measured for a LaBr3:Ce crystal of the same size. In particular, we measured 3.6% at 662keV, coupling the crystal to a low-gain high-quantum efficiency PMT.

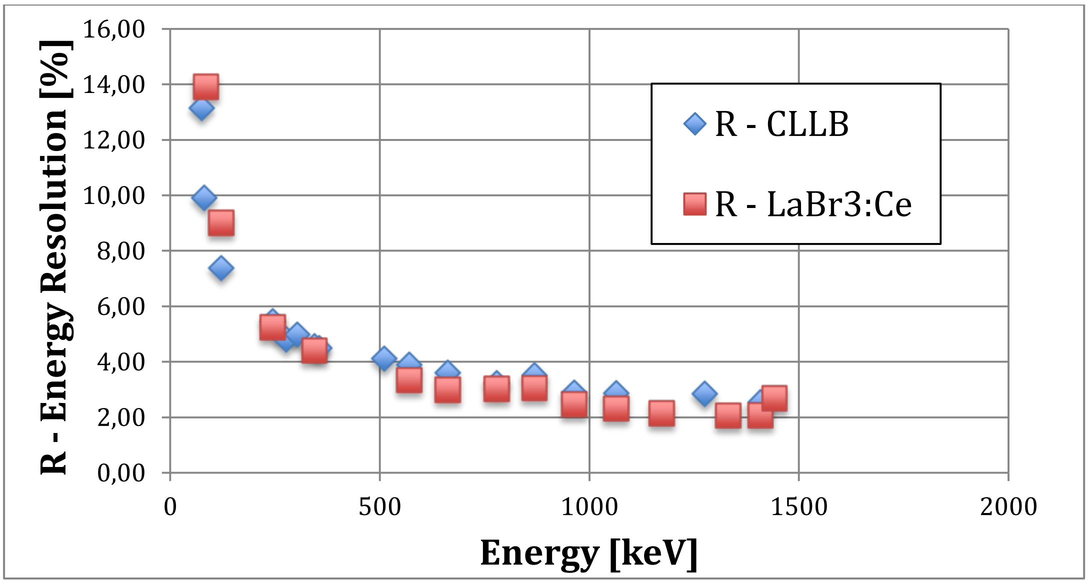

Figure 1. Energy resolution versus energy of gamma-rays measured in CLLB and $LaBr_3$:Ce crystals.

We can conclude that in case of gamma-ray energy resolution the material is more or less equivalent to LaBr3:Ce. The interesting features still to study are the possible gamma-to-neutron separation, the eventual internal radiation and of course, the absolute efficiency, similar to what has been done with CLYC crystals; see next subsection.

Subsection 1.2 Study of CLYC crystals

The CLYC, Cs_2LiYC_6:Ce, as well as the CCLB crystals belong to the elpasolite scintillator family; details on original research on the production of these crystals can be found in http://prod.sandia.gov/techlib/access-control.cgi/2012/129951.pdf and references there in.

The fast-neutron detection efficiency has been measured for two cylindrical crystals (1"x1" in size). The first is enriched with more than 99% of ^{7}Li (CLYC-7) while the second with 95% of ^{6}LI (CLYC-6). The experiment was performed at the L.A.S.A. Laboratory of INFN and University of Milano (Italy), using a calibrated Am-Be source. To identify neutron signals, the PSD (Pulse-Shape-Discrimination) technique has been used. A value of the total relative efficiency for fast neutrons (E_n > 1.5 MeV) of approximately 1% was measured. The work was presented in Dublin at the ANSRI conference by a PHD student, who won the second prize for the best talk between young speakers.

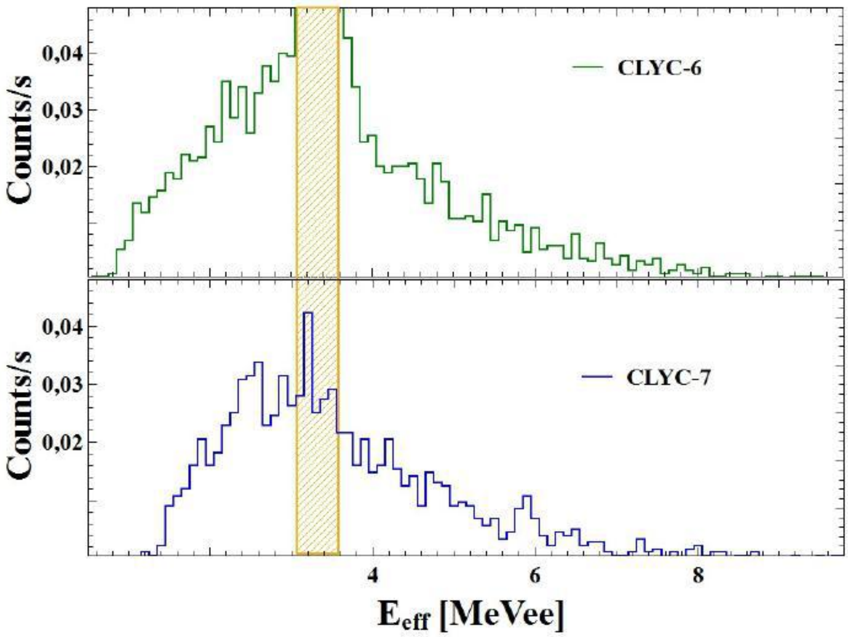

Figure 2. Comparison between the neutron experimental spectra of the two CLYC crystals. Top panel: CLYC-6 measured energy spectrum. The region between 3.1 MeV and 3.6 MeV is completely dominated by the thermal neutron peak (due to the interaction in 6Li). Bottom panel: CLYC-7 measured energy spectrum. Figure is from the ANSRI conference proceedings.

The CLYC-7 detector turns out to be a good neutron spectrometer in the studied energy range, whereas for CLYC-6 the neutron spectrum in the energy range of 3 –3.5 MeV is dominated by the thermal neutron background, so that the TOF measurement is fundamental for the subtraction of thermal neutrons; see Fig. 2 and Publication [1].

Subsection 1.3 Study of response to high-energy gamma rays in LaBr3:Ce

The measurement of the detector response function and relative/absolute efficiency of LaBr3:Ce detectors has been published in several papers. However, measurements have been done using calibration sources, namely gamma rays with energies between 100 keV up to 3 MeV at most. Therefore, any number for gamma rays with energy higher than 3 MeV is extrapolated, using GEANT simulations, from low energy values.

In case of large-volume LaBr3:Ce detectors(in particular detectors with 9 cm of diameter and 20 cm of length) the measurement of the absolute/relative efficiency and linearity for medium- to high-energy gamma rays is even more important as only in this case the full volume of the crystal will be used to stop gamma-rays. In fact, to measure 661 or 1322 keV gamma rays only the first 5-10 cm of the detector are used. The remaining volume is not used since low-energy gammas do not penetrate so deeply into the detector. Any GEANT extrapolation for high-energy gamma rays, therefore, needs to be confirmed by some experimental points.

Using the NewSubaru facility in the SPring8 laboratory, we have measured the response function and efficiency of large-volume (3.5” x 8”) LaBr3:Ce detectors. We used almost monochromatic beams, produced using the backward Compton scattering mechanism. In particular, we have used a beam of electrons with energies ranging from 500 MeV up to 1.5 GeV, circulating in the NewSubaru storage ring and a commercial laser with a wavelength of 1040 nm.
It was possible to produce a collimated beam of “monochromatic” gamma rays with energies from 5 up to 38 MeV. The energy spread of the beam is of the order of 5% and the spectral distribution has a triangular shape.

Two LaBr3:Ce detectors from the HECTOR+ array were used in the test. The PMT and the voltage divider were exchanged between the detectors to disentangle the crystal-induced effect from the PMT ones. In the following picture, Fig. 3, the measured response function is reported for one of the two detectors.

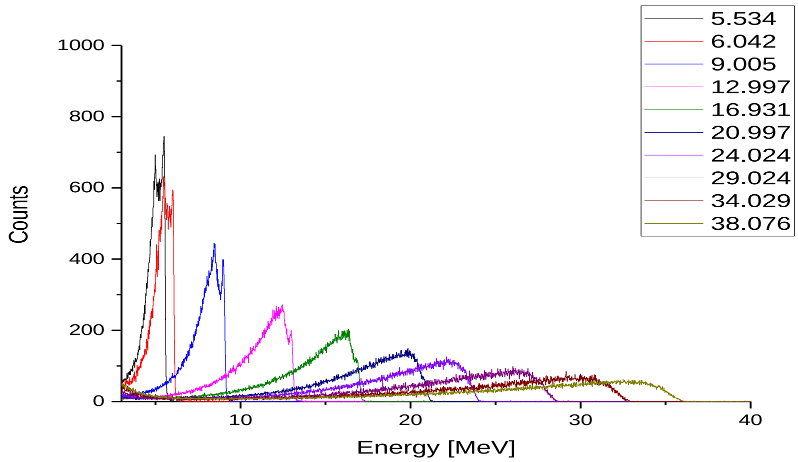

Figure 3. Measured gamma-ray response function with a $LaBr_3$:Ce detector from the HECTOR+ array. The inset lists the gamma-ray energies produced by Compton backscattering.

The figure shows the energy spectra measured by one $LaBr_3$:Ce detector (diameter 9 cm – height 20 cm). The incident gamma rays have the energy indicated in the inset to the right of the figure, with a triangular line-shape and a FWHM of approximately 5%.

In the measurement, we have used two different $LaBr_3$:Ce crystals, two PMTs and two Voltage Dividers. The measured data have shown that the large volume $LaBr_3$:Ce crystals have a rather linear response with gamma rays up to 38 MeV while the non-linear effects are mainly due to the PMT and VD. The measured efficiencies have not yet been extracted as data analysis is still in progress.

Subsection 1.4 Study of position sensitivity using LaBr3:Ce detectors

The position sensitivity properties of a 3" x 3" in $LaBr_3$:Ce crystal having diffusive surfaces was studied on an event-by-event basis. The event-by-event analysis was performed using four position-sensitive Hamamatsu H8500 PMT's, with segments short-circuited in groups of 16, in order to have only four "macrosegments" for each PMT. The crystal surface was then covered by 12 such "macrosegments". By measuring the collimated sources in several positions, we found a position sensitivity with a resolution of the order of 2.3 cm for 662 keV and 2 cm for 1832 keV gamma rays. Two of the PSPMT were contributed by the CSIC group of the PASPAG collaboration.

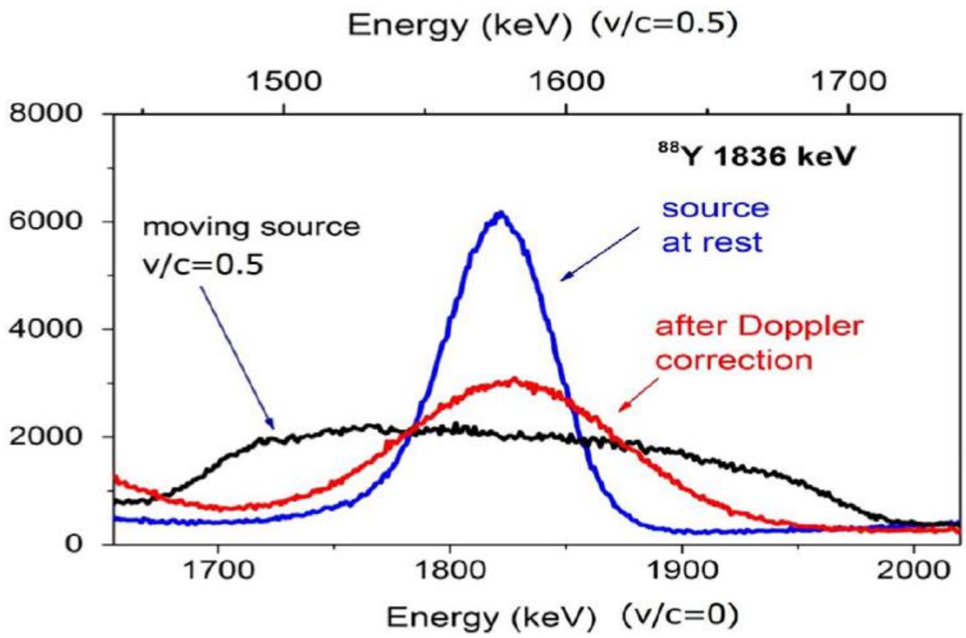

Figure 4: The black line shows the measured spectrum of the 1836 keV simulating a source moving with v/c = 0.5. The energy scale is on the top of the figure. For comparison, the measured original spectrum is also shown (blue line, energy scale on the bottom of the figure). The measured spectrum obtained after Doppler correction (red line) is also plotted. Figure from Ref. [1].

Furthermore, we investigated whether this position resolution would be sufficient to correct for the Doppler broadening, i.e. if the detector could be useful in reaction experiments, where the gamma rays are emitted in flight (see Fig. 4). The energy resolution, which is 50 keV for the source (^{88}Y, Eγ=1836 keV) at rest, becomes 250 keV for a moving source (v/c = 0.5). By reconstructing the gamma interaction point, the Doppler broadening can be corrected and one obtains a resolution of 100 keV, demonstrating that the position sensitivity of large LaBr3:Ce can be used for studies, where an energy resolution of 4 – 5% at 1 MeV is needed; see publication [2] for details.

Section 2 Publications and Outreach

Subsection 2.1 Published papers:

1. A. Giaz et al., Nucl. Instrum. Meth. Phys. Res.-A825 (2016) 51
2. N. Blasi et al., Nucl. Instrum. Meth. Phys. Res.-A839 (2016) 23
3. A. Mentana et al., J. Phys.: Conf. Ser. 763 (2016) 012006 http://iopscience.iop.org/1742-6596/763/1/012006).

Subsection 2.2 Conference contributions and Outreach

- Alice Mentana, Franco Camera et al. "Measurement of fast-neutron detection efficiency with ^{6}Li and ^{7}Li enriched CLYC scintillators".
  ANSRI - Application of Novel Scintillators for Research and Industry.
  Dublin, Ireland, May 11-13, 2016. ***The talk won the second prize for the best talk between young speakers***.

- Agnese Giaz, Franco Camera et al. “Investigation of fast-neutron spectroscopy capability of ^{7}Li and ^{6}Li enriched CLYC scintillator for nuclear physics experiments
  ANSRI - Applications of Novel Scintillators for Research and Industry.
  Dublin, Ireland, May 11-13, 2016.

- Agnese Giaz, Franco Camera et al. ”New Scintillator detectors for nuclear physics experiments”.
  Terzo Incontro Nazionale di Fisica Nucleare INFN 2016.
  14-16 November 2016, Laboratori Nazionali di Frascati

- Agnese Giaz, Franco Camera et al. “A new scintillator detector for nuclear physics experiments: the CLYC scintillator”
  GDS Topical meeting 27-29 January 2017 Laboratori Nazionali di Legnaro

- Agnese Giaz, Franco Camera et al. “New scintillator detectors for nuclear physics experiment”, NUSPIN 2016. Workshop of the nuclear Spectroscopy Instrumentation Network and AGATA physics S. Servolo Venice, 27 June - 01 July 2016

- Agnese Giaz, Franco Camera et al.: “Studio della possibilità di misurare spetti continui di neutroni veloci con scintillatori CLYC”. Congresso Nazionale della Società Italiana di Fisica Padova, 26-30 September 2016.

- Giulia Hull et at.:” New developments on scintillator detectors”. LIA France-Ukraine workshop 2016. 19-21 October 2016, Laboratoire de l’Accélérateur Linéaire, Orsay France

Conclusion

The study of new promising scintillator materials has been initiated. These new materials are being characterised with gamma rays, charged particles and neutrons. The features of the materials are being characterised according to energy resolution for gamma rays as function of energy, and possible gamma-neutron discrimination. So far, crystals of different size of the more promising materials like CLYC, CCLB and LaBr3:Ce are tested with high-energy gamma rays and in high neutron flux. There is still several investigations and comparisons to be done before we can go from crystals to actual performing detectors, and especially to choose the best crystal for a specific application.

The process of testing new materials is not straightforward. This is further complicated by administrative difficulties in obtaining the crystals and by the delay in delivery. Furthermore, the access to beam time at different facilities, where high-energy gamma rays or particles can be obtained, is an additional complication.

The work is ongoing; we have a good starting point, where now also the other tasks of the PASPAG JRA can profit from our findings. The work will continue on a timescale that is dependent on delivery of new material. A yearly update of the report will be made to present progress made with the testing of new material.

ANNEX