Deliverable D10.1

WP10 – JRA04 – Innovative solutions for nuclear physics detectors

R&D on new and existing scintillation materials Report on characterisation of organic and inorganic scintillators for neutron and gamma detection. Results of detector prototype tests

Inorganic scintillators and related photosensors

a) CsI(TI)

CsI(TI) crystals, with a range of desirable characteristics, particularly in regards to intrinsic energy resolution, have been identified as a promising detection material in challenging physics experiments. These experiments demanded the use of very long crystals (to perform calorimetry) in a highly segmented (needed for spectroscopic studies) and compact geometry.
One limitation related to the use of CsI(TI) is the important light output non uniformity that can deteriorate significantly the intrinsic energy resolution of raw CsI(TI). Long crystals with mirror polished facets exhibit light output non-uniformity when irradiated at different positions along the main axis. The effect is caused by light attenuation and optical focusing. Light attenuation yields a decrease in light output with increasing distance from the photosensor, converse to the effect of optical focusing. Depending on the final crystal geometry one effect or the other prevails. For crystals with a uniform dopant concentration and surfaces with mirror reflectivity, this results in an exponential drop in light output along the main axis as the distance to the photosensor decreases. Starting from such a dependence, lapping can be applied in order to achieve a uniform light output (LO). Crystals differ in light output uniformity significantly depending on the manufacturer, but also between samples from the same manufacturer. The R&D performed within this project has been able to determine the need of limiting drastically the variations in the LO to 3-4%. Indeed such values, particularly in the case of long CsI(TI) crystals can only been achieved by individual surface treatment.
Additionally, different readout systems were investigated, particularly SiPM and LAAPDs. The results achieved with the formers even if satisfactory in terms of energy resolution, showed a limitation in the energy range covered (this could be of extreme importance for certain applications). Moreover, the results obtained for LAAPD yield better results in terms of energy resolution and overcame the limitation in the dynamical range coverage. Different CsI(TI) crystal providers were characterised and the most promising for application in nuclear physics identified.

The APD combines the properties of the PIN PD and the PMT. It has a QE between 75-80% in the CsI(TI) emission spectrum, an internal gain of 40-50 and a linear light response in the 400-800 nm region. APDs exhibit gain variations that depend on voltage and temperature. However, these variations can be compensated. The conclusions drawn from the tests performed is that APDs represent a very nice solution for its use in new nuclear detection systems. This is due to their insensitivity to magnetic fields, their high QE, their relatively high gain and the recent development of APDs with large sensitivity. Moreover, the combination of CsI(Tl) and APDs yielded a significant improvement in the final energy resolution offered by CsI(Tl) crystals. Figure 1 shows an example of the gamma-ray energy resolution obtained for a 1cm^3 CsI(Tl) crystal coupled to an Avalanche PhotoDiode (APD), the S8664-1010 from Hamamatsu. The resolution achieved with a ^{137}Cs source is 4.42 % (DE/E) (see M. Gascón et al., IEEE Transactions on Nuclear Science 55(3): 1259-1262 June 2008, and M. Gascón PhD thesis University of Santiago de Compostela 2010)

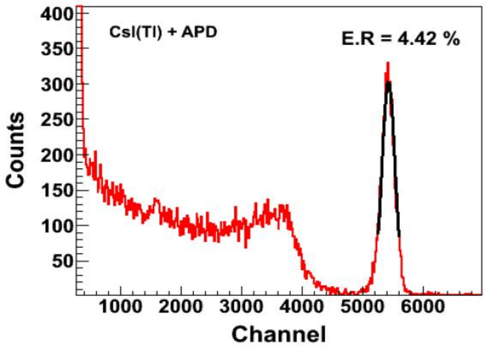

Figure 1: *Example of the best energy resolution achieved for 1cm3 CsI(Tl)coupled to a S8664-1010 Avalanche Photo Diode (APD)*

This excellent result initially motivated the choice of CsI(Tl) as the scintillator material for novel nuclear physics calorimeters presently under construction. They have also been identified as good candidates to detect simultaneously gamma-rays and light charged particles over an even wider dynamic range (see CALIFA BARREL for the R3B/FAIR experiment Technical design report).

The evaluation of small size prototypes of detectors based on the use of long CsI(Tl) detectors has been also addressed within this project, first with radioactive sources. A summary of the achieved performances is summarised in Figure 2.

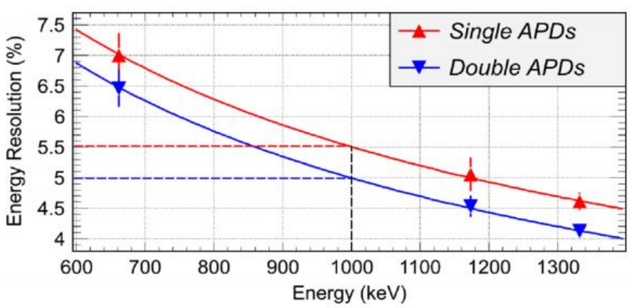

Figure 2: *Energy resolution values taken for standard calibration sources using 180 mm CsI(Tl) coupled to both 10x10 mm single and 10x20 mm double LAAPDs. Values taken from repeat measurements at our laboratory over a set of eight crystals for each case.*

Extensive testing of a detector prototype corresponding to a section of the forthcoming CALIFA calorimeter has been performed at both the MLL laboratory (Technische Universität München) and the Instytut Fizyki Jądrowej, Krakow. These tests probed characteristics pertaining to the physical detector itself, for example the response of long CsI(Tl) crystals over a range of both gamma (up to 15 MeV) and proton (70 - 230 MeV) energies. Additional physical effects such as light crosstalk between the crystals, energy straggling of protons passing through both the ESR optical wrapping and the carbon fibre alveoli support structure were investigated, the results of which were recently published by B. Pietras et al. (NIM A729 (2013) 77) and M. Gascón et al. (Journal of Instrumentation 8 (2013) P10004). Dedicated simulations were performed in order to better understand the underlying physical processes and were found to be in good agreement with the experimental results. Figure 3 summarises the results achieved by this system with high-energy gamma-rays.

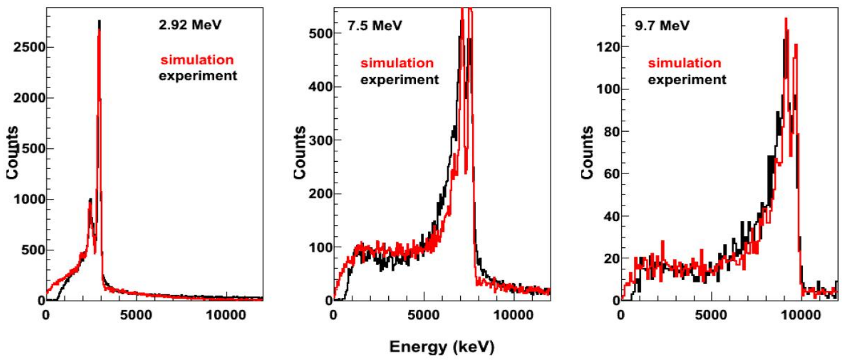

Figure 3: *Comparison of experimental and simulation spectra for three different tagged gamma energies. High energy gamma-rays were produce in the DALINAC-NEPTUN facility and were recorded with a small prototype mounted with 14 CsI(Tl)+ APDs.*

As well as testing the physical characteristics of the crystal array prototype, the FEBEX3 GOSIP digital electronics was employed, enabling traces to be optionally taken. This allows pulse shape analysis to later distinguish between gamma, proton and other particles. The use of this digital electronics setup allowed testing of the capability of phoswich arrays via exploitation of the crystal geometry by varying the direction of proton irradiation. This simple technique gives data over several different crystal dimensions, shedding light on the optimum configuration and limitations of the phoswich concept.

b) LaBr3:Ce

New inorganic scintillators such as LaBr3:Ce have been studied for use in nuclear physics in terms of linearity, energy and time resolution, response to high energy gamma-rays, internal radioactivity, particle identification and position sensitivity.

The work focused on position sensitivity has been done using segmented detectors or shielded PMTs (see for example A. Giaz *et al.* Conference Record NP01-20 IEEE-NSS Seoul 2013, this work was one of the tasks of the first ENSAR fellowship). A code based on GEANT4 libraries, which tracks the scintillation photons up to the photocathode, has been developed for the evaluation of the capability of a 3”x3” LaBr3:Ce detector to reduce the Doppler Broadening effect.

Specific voltage divider “LABRVD” for the PMTs used as readout, have been developed within this project. Large volume LaBr3:Ce crystals have been successfully tested with radioactive sources and high energy gamma rays (see for example L. Pellegri proceedings of Varenna Conference – 2012).
The characterization of the properties of large volume $LaBr_3$:Ce (which started in the first part of the project) was concluded and the results have been published in the paper ¨Characterization of large volume 8″ x 8 ″ LaBr3:Ce detectors¨ by A.Giaz et al. (NIM A 729 (2013)910). The studied detector is among the largest ones available and many properties, not directly scalable from the ones of smaller detectors, have been analyzed. We tested the $LaBr_3$:Ce detectors using two different voltage dividers coupled to the PMTs. We proved that $LaBr_3$:Ce detectors are capable to measure transitions at 5 keV (see figure 4) and at the same time clearly separate the full energy peak from first escape peak up to at least 25 MeV gamma-ray energy. This is a unique feature for a scintillator detector to date.

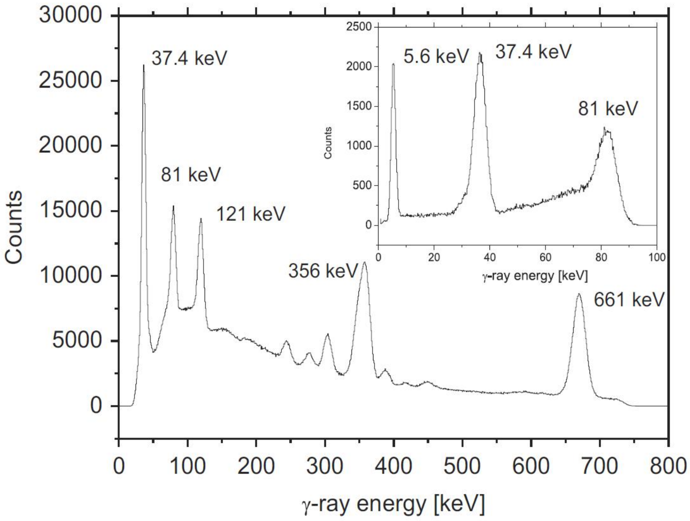

Figure 4. *A low energy spectra measured with the 3.5″_8″ LaBr3:Ce detector and the "LABRVD" active voltage divider using ^{137}Cs, ^{152}Eu and ^{133}Ba sources. The leftmost energy peak (at 37.4 keV) corresponds to the X-ray K shell of Ba, while the 81 keV energy peak comes from the ^{133}Ba source. In the inset spectrum it is also shown the 5.6 keV peak corresponding to the X-ray L shell of Ba. The plot is taken from A.Giaz et al. NIM A 729 (2013)910.*

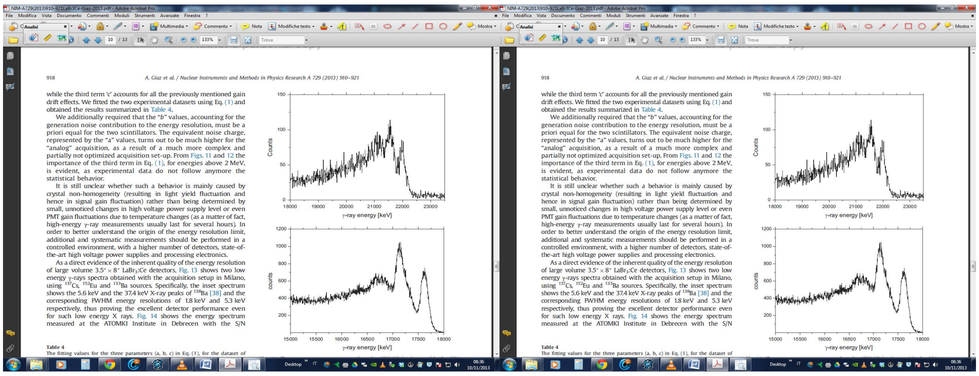

Figure 5. *The energy spectra measured at the ATOMKI Institute, with a 3.5" x 8" $LaBr_3$:Ce detector and the "LABRVD" active voltage divider in case of 22.6 MeV and 17.6 MeV monochromatic γ rays. The plot is taken from A.Giaz et al. NIM A 729 (2013)910]*

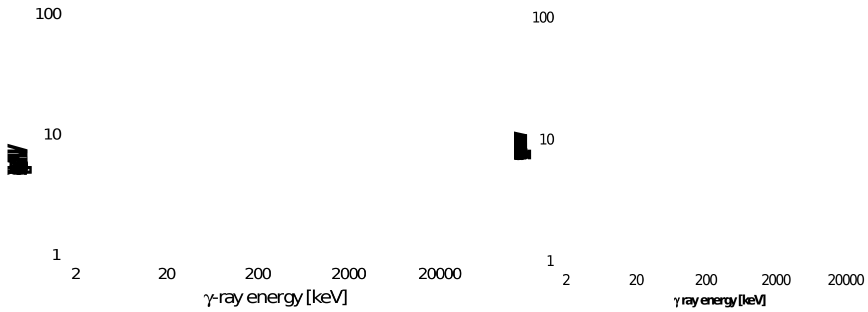

Figure 6: *The energy resolution measured in large volume $LaBr_3$:Ce detectors for γ-rays ranging from 1 to 22600 keV. In the left panel, the measurements with digital electronics are shown while in the right panel those taken with the analogue one are displayed. The dashed line shows the expected $(E)^{1/2}$ trend while in the continuous line a term linear with energy was added [13]. The plot is taken from F. Camera et al. proceedings of the INPC-2013 conference (Florence) and A. Giaz et al. NIM A 729 (2013)910.*

In both the plots of figure 6, the energy resolution of the $LaBr_3$:Ce detectors deviates from a strictly statistical behavior in the case of high-energy γ rays. The energy resolution of $LaBr_3$: Ce detectors tends, in fact, to saturate at a constant value around 0.5-1%. This was already reported in the literature and confirmed by this work. The saturation behavior can be understood adding a linear dependence in the energy resolution equation, namely $FWHM^2 = a+bE+cE^2$. In this equation the first term '*a*' represents the electronic noise, the second term '*b*' modulates the contribution of the scintillation light production while the third term '*c*' can account for gain drift or non--homogeneities effects.

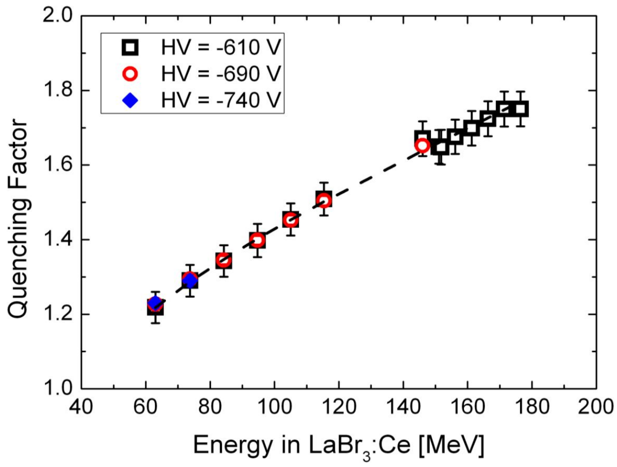

Figure 7: *The measured quenching (preliminary data) factor measured for protons in a LaBr3:Ce 3"x3" detector. The plot have been taken from A.Giaz et al. Conference Record NP01-21 IEEE-NSS Seoul 2013.*

The use of LaBr3:Ce combined in phoswich detectors with other materials has also been explored. The coupling of these detection systems to different PMT has also been evaluated, including readout by segmented PMT.

In particular, a Phoswich prototype (PARIS Cluster prototype, composed of 3x3 phoswiches of LaBr3:Ce + NaI) has been tested in the ALTO facility in Orsay using calibration sources and in beam gamma-rays. In this kind of detector, on an event by event basis, we have to identify if the gamma-ray interact in NaI only, in LaBr3:Ce only or in both. The energy deposited in each crystal type need to be measured, calibrated and separately treated. In the performed tests we have successfully used the electronic and DAQ designed for large volume LaBr3:Ce especially modified for PARIS phoswiches. The data analysis for the characterization of the cluster is still in progress.

The test of other phoswich prototype (LaBr3+NaI) has also been studied in the view of digital data acquisition system and front-end electronics requirements.

The emphasis with Phoswich was on the discrimination of two different components (LaBr3 vs NaI) and their properties in the view of energy spectrum, since their signal time development properties are quite different.

c) Other materials

One 1" x 1" sample of the very new CLYC scintillator detector has been studied. The crystal shows an energy resolution of 4.7 % (at 661 keV) using 12 microseconds of shaping time was measured. The contribution from the internal radiation was measured using three different kind of Pb shielding (see fig. **8**). It was found that the measurements show that the CLYC internal radiation is approximately at least two order of magnitude smaller than that of an equivalent $LaBr_3$:Ce detector.

Prototype tests

Along the project duration several irradiations have been performed on both scintillators and different size detector prototypes built. We provide the list of experiments performed together with the publication list result of these irradiations. The list is subdivided by the nature of the scintillator/photosensors used.

CsI(TI)+ APD

- detector irradiation

- CMAM tandem accelerator in Madrid Spain, production of high energy gamma rays

- Neptun tagger, S-Dalinac Darmstadt (Germany) , production of bery high energy gamma rays (up to 20 MeV)

- TSL Upsala, Sweden (3 days beam time), High-energy protons

- 32 channel prototype irradiation

  - Tandem accelerator TUMunich, Germany, production of high energy gamma rays
  -Kracow Cyclotron Laboratory (One measurement four days of beam time)

- 64 channel prototype irradiation

  - Kracow Cyclotron Laboratory (One measurement four days of beam time)
  - Cave C/ GSI Darmstadt Germany (10 days experiment) : detection of high-energy protons (up tp 400 MeV) and gamma-rays produced in QFS reactions

LaBr(Ce) with custom Voltage divider developed in this project

EXPERIMENTS

- ATOMKI in Debrecen (one week of beam time) for the production of high energy gamma rays
- Kracow Cyclotron Laboratory (Two measurements for three days of beam time)
- Data Taking with AmBe neutron source or AmBeNi composite source (several weeks)

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WP10 – JRA04 – Innovative solutions for nuclear physics detectors

Organic scintillators and related photosensors

The main objective of the activities of this deliverable was to achieve the optimal performance for detecting neutrons with existing and new scintillation materials, and to determine the best combination between the scintillating materials and optical conversion devices.

Organic scintillator materials have been widely used as the best candidates for fast neutron detection by time-of-flight technique. The main advantages behind this choice are the relatively high intrinsic efficiency, the very fast response and in some cases a discrimination capability between neutron and gamma events based on pulse shape. The key requirements of neutron detectors, for example those to be used as a TOF spectrometer, are to have high detection efficiency, a high energy resolution, neutron/γ discrimination capabilities and modularity. The study of these requirements, including scintillation materials and geometrical configurations, has been carried out for reaching an optimal design of a TOF spectrometer.

Among different organic scintillator materials, plastic, crystalline and liquids, two organic scintillation liquids, BC501A and EJ301, offer at present time the best performance in terms of efficiency, time response and neutron/γ discrimination. On the other hand, flexibility in the geometric configuration is demanded for achieving the requirements and reducing cross-talk. Therefore, such liquids have been tested with two geometrical configurations, rectangular bar cells and cylindrical cells.

Both geometric configurations have been investigated experimentally and by Monte Carlo simulations with GEANT4, in order to optimize the cell dimensions in terms of efficiency, energy resolution and light collection. In the case of rectangular bar detectors, the best performances have been achieved for modules with a 5 cm x 5cm section and 50 cm length. A bar prototype was filled with EJ301 liquid scintillator and coupled to both ends with two Photonis XP2262B photomultipliers. Several tests performed with standard gamma-ray and neutron sources yield a large signal attenuation of 35% between the centre and the extremes of the bar. In fact the number of photoelectron per energy deposited determined at those positions followed a similar behaviour with values around 150-200 phe/MeV. Time resolution and position resolution has been determined to be of the order of 2 ns and 14 cm respectively. A poor value on neutron/gamma-ray separation has also been obtained with a ^{252}Cf neutron source. The figure of merit (FOM) showed also a large dependence with interaction position obtaining values of 0.7 at the center and 1.0 at the end of the bar at energies of 500 keVee.

In the case of cylindrical geometry, the optimal trade-off between efficiency and energy resolution was achieved for a cell diameter of 20 cm and a length of 5 cm, for flight distances around 2 m. In order to couple the largest available photomultiplier of 5 inch diameter, a light guide has been included in the design. A light collection efficiency of around 45% is achieved with a 3cm thickness conical light guide with diffuse coating covering the lateral surface. A prototype of cylindrical cell filled with BC501A has been tested with standard gamma-ray sources and a ^{252}Cf neutron source. The cell was coupled to different photomultiplier models, fast and slow types. A summary of the main results obtained with PMT are reported in the table below.

PMT model	**N_{phe}/MeV**	**Timing (ns)**	**FoM @ 500 KeVee**
XP4512B	1175 (60)	0.7 (1)	1.8 (1)
R877-MOD	1580 (50)	1.2 (1)	1.7 (2)
R4144	1050 (50)	0.9 (1)	1.5 (1)
E9823KB	800 (60)	1.1 (1)	1.0 (1)

The optical performance has also been evaluated with two of the photomultipliers, XP4512 and R4144, and the results have shown an excellent agreement with the simulation, obtaining a relative variation of the light collection efficiency around 10% between the central position and the edge of the scintillator surface. A dedicated robotic scanner was built and operated at CIEMAT for determining the 2D spatial dependence of the light collection efficiency.

The response function of the cylindrical cell filled with BC501A liquid scintillator has been characterized by irradiation with mono-energetic neutron beams at two similar facilities, PTB (Germany) and CEA/DAM (France). Neutron beams of energies between 0.144 and 14 MeV were produced through the D(d,n), T(p,n) and ^{7}Li(p,n) reactions. The response function at several energies and the intrinsic efficiency has been determined experimentally by the time-of-flight technique. The response functions at each energy were simulated with a modified version of the GEANT4 code specifically developed for such a neutron detector. A general good agreement with experimental responses has been achieved.

Other scintillation materials have also been characterized with standard gamma sources and ^{252}Cf neutron source, in particular stilbene organic crystal and EJ309 liquid scintillator doped with Boron, which allows discriminate thermal neutron events by capturing in B-10. Furthermore, the new plastic scintillator EJ299-33, synthetized for the first time by Zaitseva et al. with neutron/γ discrimination capability has also been tested. Those detectors have been irradiated with mono-energetic neutron beams at the PTB neutron facility.