# TESTING AND DEVELOPMENT OF A NOVEL PHOSWICH SCINTILLATOR DETECTOR FOR PARIS\* \*\*

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 $LaBr_3(Ce)$  is a novel scintillator that holds a lot of potential in  $\gamma$ -ray spectroscopy due to its high energy resolution and timing properties, despite exhibiting self-activity due to the <sup>138</sup>La isotope in the crystal (0.09% abundance). However, due to the high cost of these scintillators, a phoswich detector was acquired as a more cost effective approach to constructing the new Photon Array for the study of Radioactive Ion and Stable beams (PARIS), intended for the SPIRAL2 beam-line at GANIL. Tests on the timing and energy response of a  $1" \times 1" \times 2"$  LaBr<sub>3</sub>(Ce) and  $1" \times 1" \times 6"$  CsI(Na) phoswich detector is presented. Little is known about what happens to these crystals during neutron activation, and this was also investigated, where the study of the pulse shapes from neutron and gamma sources were used to show that  $(n, \gamma)$  discrimination was not possible. An activated spectrum from a  ${}^{241}Am/{}^{9}Be$  source was acquired where neutron activation due to excited states of lanthanum and bromine (<sup>140</sup>La, <sup>80</sup>Br and <sup>82</sup>Br) were found. The timing of the phoswich was also investigated, and found to be ~ 650 ps when used in a start-stop set-up with  $BaF_2$ . A marginal improvement of around 40 ps was achieved despite the set-up being poorly time matched.

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## 1. Introduction

PARIS<sup>1</sup>, is a newly formed collaboration, the aim of which is to design and build a high efficiency detector consisting of two shells of crystals for medium resolution spectroscopy and detection of  $\gamma$ -rays over a large range of energies.

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<sup>&</sup>lt;sup>1</sup> PARIS Collaboration website can be found at http://paris.ifj.edu.pl

The inner shell of crystals will be highly granular and made from  $LaBr_3(Ce)$ , while the outer hemisphere of crystals will have a lower granularity, larger volume and higher stopping power (*e.g.* CsI(Na)), used primarily to measure high energy photons. Due to the high cost of purchasing  $LaBr_3(Ce)$  scintillators, another cost effective solution would be to purchase smaller  $LaBr_3(Ce)$  scintillators in a phoswich arrangement with a cheaper scintillator. The response of this arrangement under several experimental conditions are discussed and presented.

## 2. The phoswich detector

A 1" × 1" × 2" LaBr<sub>3</sub>(Ce) crystal was coupled to a 1" × 1" × 6" CsI(Na) scintillator to form the detector, which was acquired from Saint Gobain crystals. The difference in the decay times of both signals (~ 20 ns and ~ 600 ns for LaBr<sub>3</sub>(Ce) and CsI(Na) respectively<sup>2</sup>) allows both signals to be discriminated and read off one PMT, resulting in high detector efficiency due to a reduction in dead space. CsI(Na) typically has a resolution that is of the order of 7% at 662 keV, compared to ~ 3% found in the LaBr<sub>3</sub>(Ce) crystal.

## 2.1. Self-activity and actinide contamination

"Self-activity" is due to the presence of the radioisotope <sup>138</sup>La in LaBr<sub>3</sub>(Ce), a naturally occurring isotope with an abundance of 0.09% and half-life of  $1.05 \times 10^{11}$  years. In 66.4% of its decays, <sup>138</sup>La undergoes electron capture to an excited state of <sup>138</sup>Ba via photon emission of a 1436 keV  $\gamma$ -ray. However, this is displaced by 32 + 5 keV due to the reoccupation of the K and L electron shells respectively, that result in coincident barium X-rays [2]. The remainder of <sup>138</sup>La decays 33.6% of the time via beta emission to <sup>138</sup>Ce, emitting a 789 keV  $\gamma$ -ray from the 2<sup>+</sup> state.

Alpha contamination is revealed by 4 or 5 broad peaks due to the presence of <sup>227</sup>Ac<sup>3</sup> ( $T_{1/2} = 21.77$  years [4]), which  $\beta$ -decays to <sup>227</sup>Th, and subsequently  $\alpha$ -decays to <sup>223</sup>Ra, <sup>219</sup>Rn, <sup>215</sup>Po and <sup>211</sup>Pb. However, here the  $\alpha$ -decay chain becomes disturbed due to the  $\beta$ -decay of <sup>211</sup>Pb to <sup>211</sup>Bi, before the chain ends with the  $\alpha$ -emission of <sup>211</sup>Bi to <sup>207</sup>Tl. When calibrated with  $\gamma$ -rays,  $\alpha$  particles were found to produce 65% less light ( $\frac{\alpha}{\gamma} = 0.35$ ), which is in agreement with a similar finding for LaCl<sub>3</sub>(Ce) scintillators by Hartwell and Gehrke [3]. This quenching for  $\alpha$  particles is possibly due to the sensitivity of the scintillation mechanism.

<sup>&</sup>lt;sup>2</sup> These values for decay times are dependent on the quantity of dopant present in the scintillator. Typically for LaBr<sub>3</sub>(Ce), a decay time of 23 ns is seen with a Ce<sup>3+</sup> concentration of 0.2% [1].

<sup>&</sup>lt;sup>3</sup> <sup>22</sup>Ac is in the same periodic group (group IIIB) as lanthanum.

## 3. Neutron response in the phoswich

## 3.1. Neutron activation

The phoswich was placed within 20 cm of a sealed 10.5 GBq  $^{241}$ Am/ $^{9}$ Be source, where a bias of -1200 V was applied to the detector, and subsequent amplification was done with an ORTEC 472A NIM module. The neutron response was then investigated to study how activation affects the response of the detector, where in-beam tests were seen to activate the detector material.

It was anticipated that coincident  $\gamma$ -rays, as well as detecting neutrons, would be detected. The 4.4 MeV  $\gamma$ -ray due to the de-excitation of the 2<sup>+</sup> state in <sup>12</sup>C, produced by the <sup>241</sup>Am/<sup>9</sup>Be source in the reaction:<sup>9</sup>Be+ $\alpha \rightarrow$ <sup>12</sup>C<sup>\*</sup> was used to determine the FWHM efficiency of the energy resolution at high energies. However, due to the size of the crystal, the FWHM of the 4.4 MeV  $\gamma$ -ray photo-peak was found to be poor, however it was used for calibration purposes which allowed for the identification of excited states; <sup>140</sup>La and <sup>80</sup>Br,<sup>82</sup>Br. This is due to the large thermal neutron cross-sections of naturally occurring bromine and lanthanum; <sup>79</sup>Br, <sup>81</sup>Br and <sup>139</sup>La with abundances of 50.69, 49.31 and ~ 99%, respectively [4]. The calibrations were performed with various sources, and by obtaining a time lapse of spectra offline to identify the excited states and the time it took for their decay.

#### 3.2. Pulse shape discrimination

Pulse shape analysis was carried out to see if  $(n, \gamma)$  discrimination was possible. Generally, organic scintillators provide such discrimination due to delayed remission of energy; however, inorganic alkali halide crystals, such as LaBr<sub>3</sub>(Ce) and CsI(Na) do not. A previous study [5] performed with a  $1.5" \times 1.5"$  LaBr<sub>3</sub>(Ce) detector shows that the discrimination was essentially non-exsistent. In the phoswich detector, a similar result is observed, where it was possible to extract information from the fast and slow components of the pulse by gating on 3 parts of the pulse, and fitting the resulting data.

#### 4. Timing measurements: set-up

Coincidence timing tests of the phoswich detector were investigated by irradiating a 1" Bicron BaF<sub>2</sub> detector and both scintillators in the phoswich detector separately. BaF<sub>2</sub> has two components, a fast component with a decay time of approximately 600 ps and a slow component of 630 ns [6]. The phoswich detector was coupled to a fast Hamamatsu R7057 PMT, operated at -1200 V with 511 keV positron annihilation  $\gamma$ -ray pairs emitted by a <sup>22</sup>Na source. The BaF<sub>2</sub> detector formed the "start" channel in this timing circuit, while the phoswich detector formed the "stop" channel. The signals from both detectors were processed by NIM electronics. Data Acquisition was performed with an ORTEC Multi-Channel Analyser (MCA) unit, with pulses saved on a fast 500 MHz LeCroy Waveform oscilloscope at a sampling rate of 5 GS/s for rise time analysis. Data was accumulated using a LeCroy coincidence timing unit and ORTEC-996 Time Counter. Both channels were triggered by the coincidence unit externally to allow for better statistics.

#### 4.1. Timing measurement results and analysis

Two sets of data were collected; one set determining the timing information from the front end of the detector, and another set from the side of the detector, where scintillation is primarily within the CsI(Na) crystal. It was seen that the count rate of coincidences decreases substantially as the BaF<sub>2</sub> detector is moved along the side of the phoswich detector due to the bias voltage being too low to record CsI(Na) pulses. As a result, the bias was increased to -1300 V on both the phoswich and BaF<sub>2</sub> detectors, and a ORTEC 474 Timing Filter Amplifier (TFA) was added to optimise the signal. Using delays of 100 and 300 ns with a TAC range of 400 ns, it was found that  $45.0 \pm 0.1$  ps/ch resolution was possible with the front end of the detector, and  $167 \pm 4$  ps/ch was attainable when scintillation occurs primarily in the back of the detector. A coincidence timing resolution plot acquired in the manner described is shown in figure 1, where a timing resolution for the front end of the detector of  $696 \pm 13$  ps was recorded for a delay of 100 ns. Similarly, a timing resolution of  $24 \pm 1$  ns was obtained for the CsI(Na) scintillator in the phoswich detector by placing the  $BaF_2$ detector near the back of the detector with a TAC range of 2 ns.

The resolutions due to the electronics were found to be  $125 \pm 1$  ps and  $1.4 \pm 0.1$  ns for the front and back respectively when a pulser was used. These results are displayed in the following table, noting that the initial and intrinsic resolution for the electronics are the same.

TABLE I

Segment	Initial res.	Timing [ps/ch]	Intrinsic res.	Delay [ns]
LaBr <sub>3</sub> (Ce) Front Elec. CsI(Na) Back Elec.	$696{\pm}13$ $125{\pm}1$ $23851{\pm}434$ $1436{\pm}110$	$\begin{array}{c} 45.0{\pm}0.1\\ 45.0{\pm}0.1\\ 167{\pm}4\\ 167{\pm}4 \end{array}$	$685 \pm 13$ $125 \pm 1$ $23810 \pm 426$ $1436 \pm 110$	$100 \\ 100 \\ 400 \\ 400$

The initial and intrinsic timing resolutions of the scintillators and electronics. All measurements in ps unless otherwise stated.



Fig. 1. Timing responses for both scintillators with 400 ns delay (approx. 200 ns internal).

Although these resolutions are not as good as previous findings where approximately 260 ps [1] was obtained for a  $1" \times 1" \times 1"$  LaBr<sub>3</sub>(5%:Ce) scintillator, the recently recorded values from this study are still fast enough to be used effectively in  $\gamma$ -ray spectroscopy.

## 5. Summary

It was observed that exposure to an AmBe neutron source showed substantial activation of numerous isotopes due to large thermal neutron crosssections in both lanthanum and bromine. This was identified and presented, with  $(n, \gamma)$  discrimination not being possible using pulse-shape analysis. A degradation of the signal was seen and found to contribute to worse-thanexpected timing and energy resolutions, where care needed to be exercised when applying the bias and gain for the LaBr<sub>3</sub>(Ce) as an optimised set-up for this scintillator might result in too little gain in the CsI(Na) component, resulting in very low statistics. The timing resolution for the phoswich was found to be in the region of 600 ps when used in a start–stop set-up with a BaF<sub>2</sub> detector. Improvement of this value was limited by the fast timing component of the BaF<sub>2</sub> scintillator, and the set-up was poorly time matched. By strobing the fast coincidence between the two outputs a cleaner TAC output was achieved. The result was only a marginal improvement of around 40 ps, with an initial resolution of 665 ± 2 ps recorded.

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