Neutron Response of 1.5″ $LaBr_3$:Ce Crystal Scintillators for PARIS

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Abstract

$LaBr_3$:Ce crystals are high resolution scintillators that hold a lot of potential in gamma-ray spectroscopy. The crystals have a self-activity due to the ^{138}La isotope in the crystal (0.09% abundance). This was observed and used to better understand the inner dynamics of the detector. Little is known about what happens to the crystals due to neutron activation and this was investigated by studying the pulse shapes from neutron and gamma sources as well as determining its activated spectrum from a ^{241}Am/^{9}Be source. It was determined that (n,γ) discrimination was not possible, and that the majority of gamma emission in the spectra after neutron activation were due to excited states of La and Br (^{140}La, ^{80}Br and ^{82}Br).

1 Motivation

PARIS or the Photon Array for the studies with Radioactive Ion and Stable Beams, 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 to detect gamma-rays over a large range of energies.

The inner shell of crystals will be highly granular, and made from $LaBr_3$:Ce, with the readout done in either a Phoswich arrangement with another crystal, APDs or with another configuration involving digital electronics and possible light-guide to limit dead space and allow for pulse shape analysis.

The outer hemisphere of crystals will have a lower granularity, larger volume, and large stopping power (e.g. CsI:Na). This outer shell of inorganic alkali halide crystals will be used to measure high energy photons, and act as a Compton suppressor.

1.1 Lanthanum Halide Scintillators

$LaBr_3$:Ce is a relatively new inorganic scintillator that has been gaining popularity for use in gamma-spectroscopy due to its excellent timing resolution (200 ps depending on the size of the detector), energy resolution (3% at 662keV), high efficiency and light output (60,700 photons/MeV with Ce^{3+} concentration of 0.2%). The decay times are generally quick with the fast components having a value of between 15-23ns depending on the Ce^{3+} concentration[1]. The rise time is typically between 3.5 and 0.7ns. The peak emission wavelength of the $LaBr_3$:Ce scintillator is in the blue-UV part of the electromagnetic spectrum, with λ_{max}=350nm (characteristic of Ce^{3+} luminescence). A shortcoming of these detectors is that they exhibit "self-activity" which can be seen as deceptive lines in experimental data. These spurious lines could limit the practicality of the detector and its uses, especially with regards to low level counting.

Figure 1: A picture showing the detector set-up with the 10.5Gbq $^{241}Am/^{9}Be$ neutron source

1.2 Self-activity of $LaBr_3$:Ce

This "self-activity" is due to Lanthanum-138, a naturally occurring radioisotope of Lanthanum with an abundance of 0.09% and a large half life of 1.05x10^{11} years. In 66.4% of its decays, ^{138}La undergoes electron capture to an excited state of ^{138}Ba via photon emission of a 1436keV gamma ray. The refilling of the electron shell results in a coincident 35keV Barium X-ray. The remainder of ^{138}La decays 33.6% of the time via beta emission to ^{138}Ce, emitting a 789keV gamma ray in the 2^+ state. A spectrum of these features in $LaBr_3$:Ce is shown below outlining its decay processes[2].

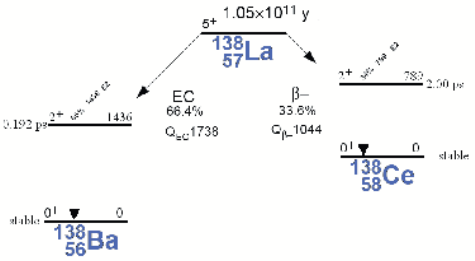

Figure 2: An Energy level diagram showing the decay branches of the metastable state ^{138}La [2]

A Background spectrum was then obtained to show the effects of the decay process of the ^{138}La metastable state in the crystal. In the spectrum, there is an off-set of 45keV as the number of counts from the ^{138}Ba 32keV X-ray were large compared to the rest of the spectrum. These X-rays occur due to reoccupation of K-shell electrons. The spectrum is a linear and not logarithimic representation of the background, as the many interesting features that are attributed to this structure can be more easily seen in this way.

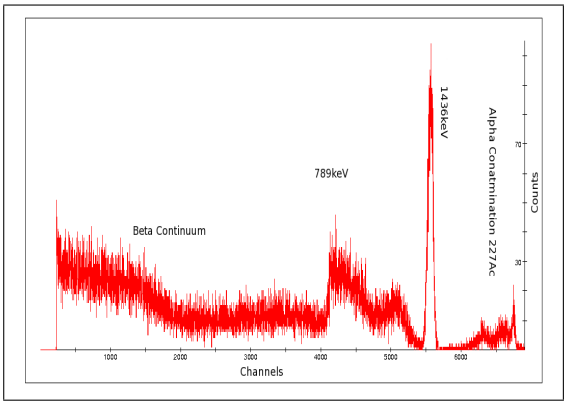

Figure 3: The background spectrum of the $LaBr_3$:Ce detector

1.3 Detector Setup

A 1.5"x1.5" $LaBr_3$:Ce scintillation detector coupled with a Photonis 2" XP20D0 PMT was obtained from Saint-Gobain. Operational voltage recommended by Saint Gobain was -1200V, however this resulting in a highly non-linear spectrum distribution as the Photon yield produced by the crystal is extremely high. Therefore the working voltage was reduced to a rather low -900V. The dynamic range was rather inconsistent and is possibly due to saturation phenomena inside the PMT, thus effecting the overall response in energy. A pre-amplifier with a shaping time of 500ns and low coarse gain was used to shape the anode signal.

The neutron response of these detectors were then tested with the neutron source $^{241}Am/^{9}Be$, where a study of how neutron activation affected the crystals was investigated.

2 Testing for Neutron Response

After measuring the background spectrum of the $LaBr_3$:Ce detector, the detector was then placed roughly 20cm from the neutron source $^{241}Am/^{9}Be$. As well as detecting neutrons, there will be some coincidence with a 4.4MeV γ-ray due to the reaction ^{9}Be and an alpha particle to produce the excited 2^+ state of Carbon-12. Due to the size of the crystal, the 4.4MeV gamma ray was hard to locate, and the FWHM of the photo-peak was rather poor. However, it was used to calibrate the graph, and identify the excited states of both Lanthanum(^{140}La) and Bromine (^{80}Br,^{82}Br). There are two stable states in naturally occuring Bromine that contribute significantly to the activated spectrum of $LaBr_3$:Ce; ^{79}Br and ^{81}Br with abundances of 50.69 and 49.31% respectively. The (n,γ) cross-sections for the formation of metastable ^{79}Br and ^{81}Br states are 2.5 and 2.4b respectively (where $1b = 10^{-28}m$). For the formation of both ^{79}Br and ^{81}Br ground-states, the (n,γ) cross-sections are 8.3 and 0.24b respectively[3]. The resulting calibrated spectrum is shown at the end of the article in figure 7.

Alpha contamination is revealed by 4 broad peaks in the 1750-3000keV energy range (the actual alpha energies range from 5000-7500keV). This ^{235}U decay chain has a long lived ^{227}Ac member ($\tau_{1/2} = 21.2$yrs). ^{227}Ac is also in the same periodic group (group IIIB) as Lanthanum. Heavy charged paritcles such as alphas, exhibit less light than gamma rays of a similar energy. A spectrum with 4096 channels is enlarged to show both ^{227}Ac contamination and the 4.4MeV gamma ray with escape peaks in figure 8 at the end of this article.

2.1 Energy Resolution and FWHM Calculations

The energy resolutions were calculated for the ^{137}Cs, ^{241}Am and ^{60}Co sources that were then used to calibrate the resulting activated spectra. Below is a small table of some of the

peaks in the calibrated spectrum.

Table 1: Energy Resolutions from $LaBr_3$:Ce spectra

Element	γ ray (keV)	FWHM (keV)	E_{Res}(%)
^{241}Am	59.54	9.23	15.50
^{137}Cs	661.73	16.89	2.55
^{60}Co	1173.21	22.18	1.96
^{60}Co	1332.54	24.14	1.81
^{12}C*	4438.90	8.46	0.19
^{140}La	1596.11	24.33	1.52
^{82}Br	1317.47	23.65	1.80
^{82}La	827.83	19.34	2.34
^{82}Br	554.35	14.56	2.63
^{208}Tl	511	14.05	2.75
^{140}La	487.02	13.52	2.78
^{140}La	328.76	10.23	3.11
^{80}Br	11.89	2.22	18.67

The above FWHM were calculated using Radware, and have an error of $\pm$1.15keV. These values of resolution were then taken and plotted against energy to get the graph below.

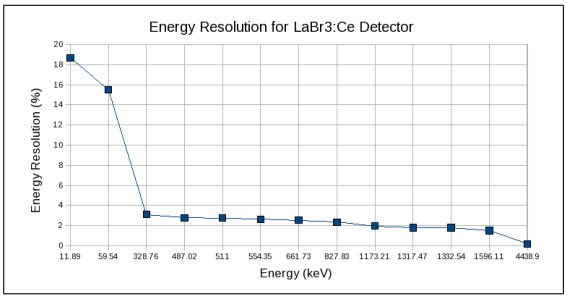

Figure 4: A figure showing the results of the FWHM measurements made, with energy resolution plotted against increasing energy

As we can see, the energy resolution of the $LaBr_3$:Ce becomes worse at lower energies, but is still better than standard NaI detectors (7.0% at 662 and 8.9% at 122keV)[2]. Now that the resolution of the crystal has been thoroughly analysed, the next thing that was studied were the pulse shapes of the crystal when subjected to both gamma and neutron sources.

3 Pulse Shape Discrimination

Scintillator material exhibits a phenomena known as luminescence. If remission of this energy is immediately after the absorption (within 10^{-8}s) then this process is known as fluorescence. However, if the remission is delayed due to an excited state being metastable at the atomic level, another process known as phosphorescence occurs. The time evolution of this remission process can be accurately modeled on the equation:

$$N = Aexp\frac{-t}{\tau_f} + Bexp\frac{-t}{\tau_s}[4] \qquad (1)$$

The finite rise time from zero to the maximum amplitude is usually much smaller or shorter, than the decay time. For most scintillators, one component of the decay constant is much faster than the other, and are referred to as fast and slow components of the pulse shape. The existence of these two components forms the technique for pulse shape discrimination. A diagram of the two pulses is shown below with the gamma pulse having a different shaping time to better illustrate both curves individually, in reality these two curves had a similar pulse height.

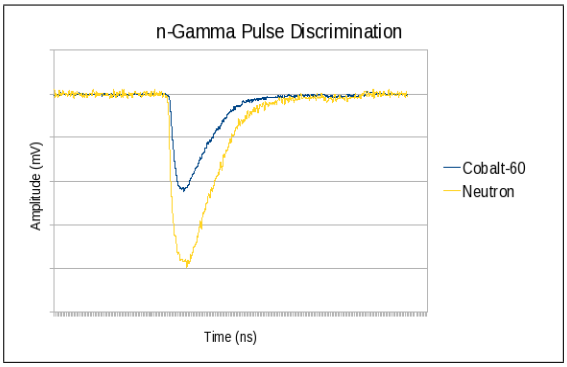

Figure 5: A comparison neutron and gamma pulse shapes

Pulse shape analysis was carried out to see if one could discriminate between gamma and neutron pulse shapes. Generally, organic scintillators provide such discrimination due to delayed remission of energy. Several batches were recorded using 2 well known gamma-ray emitters; ^{137}Cs (662keV) and ^{60}Co (1173 & 1332keV). The files were compiled into a single file and then the slow and fast components of their pulse shapes were obtained by using a code to extract the parameters needed to make the discrimination. At first the discrimination was fairly scattered, dividing the slow component by the sum of both components making the fit better. An example of this with ^{60}Co is shown in the figure below.

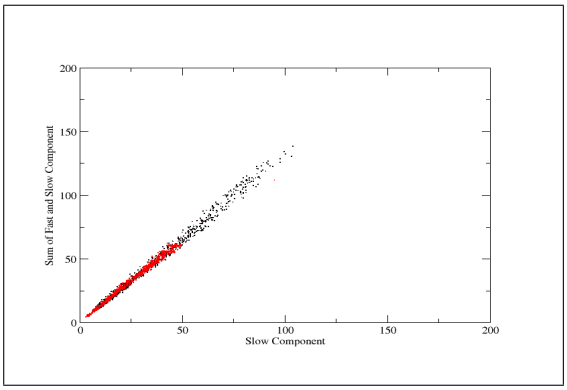

Figure 6: Fast and slow components from gamma and neutron anode pulse shapes to try to observe any discrimination

The gamma points in red show two thresholds that correspond to the gamma peaks at 1132 and 1332keV. As one can see, there is no discrimination between both sets of data unlike what would be expected for organic crystals. The magnitude of the slow component is small, and the discrimination, if any, too small to be useful in experimental conditions. However, there is modest discrimination of both neutrons and charged particles with $LaCl_3$:Ce [5]. More coincidence measurements with a beamline would help verify these results.

4 Conclusions and Future Work

Tests done on the inorganic Lanthanum Halide crystal $LaBr_3$:Ce with the neutron source ^{241}Am/^{9}Be has shown the background spectrum before and after activation due to the natural occurring metastable in Lanthanum, ^{138}La. Calibrated spectra for this crystal were presented, and revealed gamma lines from ^{140}La, ^{80}Br and ^{82}Br after neutron activation. The self-activity present in $LaBr_3$:Ce, does not make the detector a suitable candidate for low-level counts. Pulse shape analysis also revealed that very little or no discrimination between both gamma and neutron slow and fast components was possible, which agrees with previous work done[5].

Tests with larger $LaBr_3$:Ce crystals, such as 3”x3” or even 2”x2” crystals should be conducted to better verify the results, and also allow for better discrimination of gamma rays at higher energies. For now, the group in York will focus next on tests with CsI(Tl) crystals with a High gain Silicon APD from Sensl. The aim will be to determine whether or not this arrangement can be used in a phoswich arrangement with the tested $LaBr_3$:Ce crystals when construction of the PARIS calorimeter starts in 2009/2010.

5 Acknowledgements

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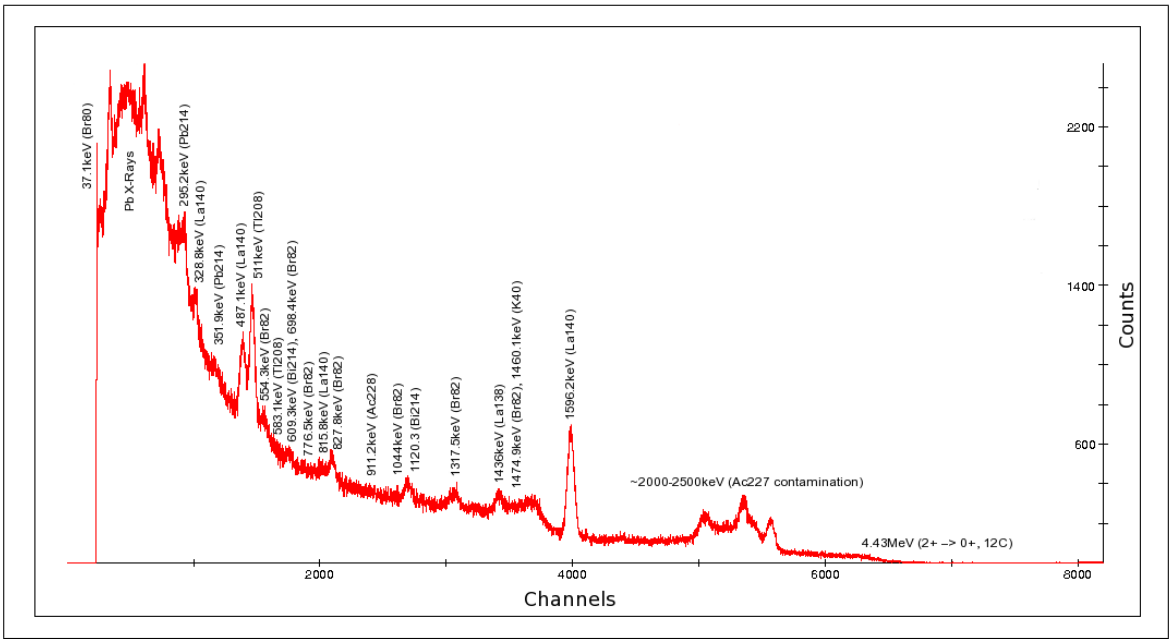

Figure 7: The calibrated spectrum showing both the background spectrum of the lab as well as lines from ^{140}La, ^{80}Br and ^{82}Br

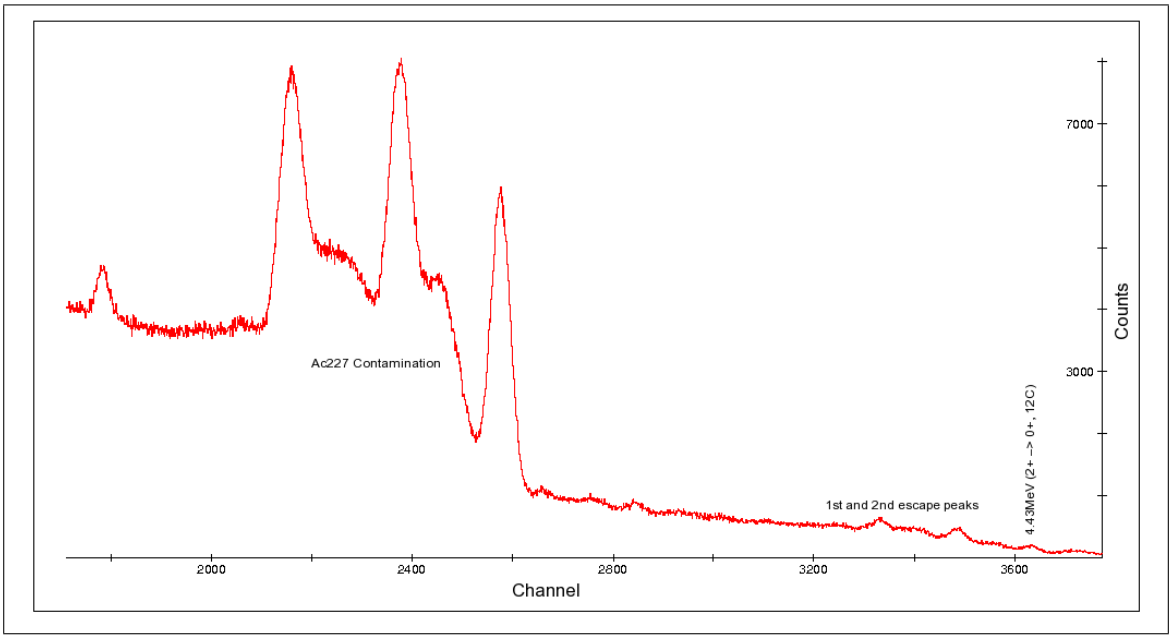

Figure 8: Neutron activation within the $LaBr_3$:Ce detector at high energies