Neutron Response of $LaBr_3$:Ce and CsI:Na Phoswich Crystal Scintillators for PARIS☆

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Abstract

$LaBr_3$:Ce is a novel scintillator that holds a lot of potential in γ-ray spectroscopy due to its high energy resolution and timing properties, despite exhibiting self-activity due to the ^{138}La isotope in the crystal (0.09% abundance). However, due to the high cost of these scintillators, a Phoswich detector is likely to be more cost effective for use in new high efficiency calorimeters. $LaBr_3$:Ce is accompanied with a CsI:Na scintillator, which was used for detection of high energy γ-rays. Little is known about what happens to these crystals during neutron activation, and this was investigated by studying the pulse shapes from neutron and gamma sources as well as determining an 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 was due to excited states of Lanthanum and Bromine (^{140}La, ^{80}Br and ^{82}Br). Similarly, activation of ^{127}I in the CsI:Na scintillator was seen, although rather short lived ($\tau_{1/2}$=24.99 mins). The Phoswich detector was also seen to be sensitive to the location of the source, with reasonable energy resolutions that make it a possible candidate for various other applications such as PET or homeland security.

Keywords: Phoswich, neutron activation, $LaBr_3$:Ce, PARIS

☆This document is a collaborative effort.

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1. Motivation

The Photon Array for the studies with Radioactive Ion and Stable Beams, PARIS[1], 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 γ 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, Si-large area avalanche photodiodes, LAAPDs, or with another configuration involving digital electronics and possible light-guides to limit dead space and allow for pulse shape analysis.

The outer hemisphere of crystals will have a lower granularity, larger volume and higher 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.

2. Preliminary Tests with the Phoswich Detector

2.1. Lanthanum Halide Scintillators

$LaBr_3$:Ce is a relatively new inorganic scintillator that has been gaining popularity for use in γ-ray spectroscopy due to its excellent timing resolution (roughly 200 ps depending on the size of the detector), energy resolution (3% at 662 keV), and high efficiency and light output (60700 photons/MeV with Ce^{3+} concentration of 0.2 %). The decay times are generally quick with the fast components having a value of between 15-66 ns depending on the Ce^{3+} concentration[1]. The rise time is typically between 3.5 and 0.7 ns. The peak emission wavelength of the $LaBr_3$:Ce scintillator is in the blue-UV part of the electromagnetic spectrum, with λ_{max}=350 nm (characteristic of Ce^{3+} luminescence), making it compatible with standard PMTs. A shortcoming of these detectors is that they exhibit "self-activity" which can be seen as deceptive lines in experimental data. These contaminants could limit the practicality of the detector and its uses, especially with regards to low level counting experiments.

2.2. Initial Testing and Signal Optimisation.

A 1"x1"x2" $LaBr_3$:Ce crystal was coupled to a 1"x1"x6" CsI:Na scintillator to form the detector, which was acquired from Saint Gobain crystals. The difference

[1]PARIS collaboration website can be found at `http://paris.ifj.edu.pl`

in the decay times of both signals (roughly 20 ns and 600 ns for $LaBr_3$:Ce and CsI:Na respectively[2]) 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 $LaBr_3$:Ce crystal. No matter where a source is placed in proximity to the detector, it is thought that scattering will occur in both scintillators which will result in one signal with a combination of properties from both scintillators. As a result, one expects a degradation in energy and timing measurements (this is discussed later in more detail).

Method	E_{Res}(%)
TFA	4.36
QVC	5.19
ORTEC 572	5.50

Table 1: Energy Resolutions at 662 keV for scintillation in the front of the Phoswich.

Due to the high light output of the $LaBr_3$:Ce and CsI:Na scintillators (roughly 60000 and 50000 Ph/MeV respectively), the working voltage was reduced from the advised -1500 V to -1200 V. The light was collected in a Hamamatsu R7057 PMT fitted with a voltage divider constructed at the University of York. Several amplifiers were used due to a bipolar signal output when using the unipolar outlet on various amplifier modules. This is possibly due to saturation effects, or impedance mismatching which could be improved with the use of a Charge-Digital Converter (QDC). As none were available, a comparative study of ORTEC 472A, 572 modules, TFA and Charge-Voltage Converter (QVC) was done to try and optimise the signal to the best of our ability. The best results were obtained with a TFA (ORTEC 474) which gave a resolution of 4.4 % at 662 keV. A roughly unipolar signal was observed along with very poor linearity, which ruled this out as a practical solution.

A specially designed NDE Bartek 202 Module intended for Euroball was used, utilising the (QVC) output. The output changes the signal by amplifying it slightly, with no shaping. A ^{137}Cs source was used in addition to a ^{22}Na source for a full energy calibration at various bias voltages ranging from -1200 V to -1500 V. It was found that with a working HV of -1200 V a resolution of 5.2+/-0.8 % was attain-

[2]These values for decay times are dependent on the quantity of dopant present in the scintillator. Typically for $LaBr_3$:Ce, a decay time of 23 ns is seen with a Ce^{3+} concentration of 0.2 %[1].

able.

The QVC was seen to be rather linear, despite a reduction in resolution compared to the TFA. Therefore, the QVC was used with the ORTEC 472A amplifier to see if this could improve the resolution while maintaining the linearity. Due to the square signal structure in the QVC, the amplifier integrates the flat peak, resulting in a bipolar distribution with an extended middle section between both the peak and trough. All information held in the signal for this region is consequently lost in the integration process, thus ruling out this method.

The ORTEC modules were specifically designed for use with Germanium detectors and consequently the shaping times are too long to integrate the $LaBr_3$:Ce signal properly (decay shaping constants are 500ns and 250ns for ORTEC modules 572 and 472A respectively). Placing the source at the front of the detector gives a resolution of 5.5% at 662keV with the 472A module, and a similar resolution with the 572.

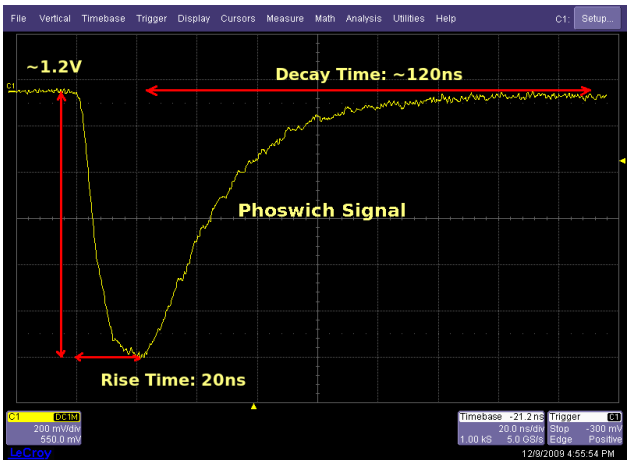

Figure 1: The combined signals from both scintillators in the Phoswich

However, issues with using these modules include the differing values of input impedance. It was found that Z_{in} = 500 Ω for the 572 amplifier, and as a result, the module was modifying the input pulse fall time substantially, thus contributing to the signal degradation output seen. A variable resistor (13 Ω to 10k Ω) was added in parallel to counteract this problem (shunt termination). A similar situation arose with the 472A ORTEC amplifier, Z_{in} =1k Ω. Ultimately, the amplification of the signal was done principally with the ORTEC 472A amplifier.

2.3. Energy Resolution and Position Sensitivity

A ^{137}Cs source was placed at the front end of the detector, resulting in an energy resolution due to interactions predominantly in the $LaBr_3$:Ce crystal. By optimising the shaping time for each scintillator (0.5 μs and 6 μs for $LaBr_3$:Ce and CsI:Na), the resolution was found to be 5.5 % and 8.6 % for $LaBr_3$:Ce and CsI:Na respectively. Similarly, by placing the source near the back of the detector, more scattering occurs in the CsI:Na and energy resolutions of 13.09 % and 11.69 % can be obtained. This is represented in figure 2 along with the position of the source for one instance.

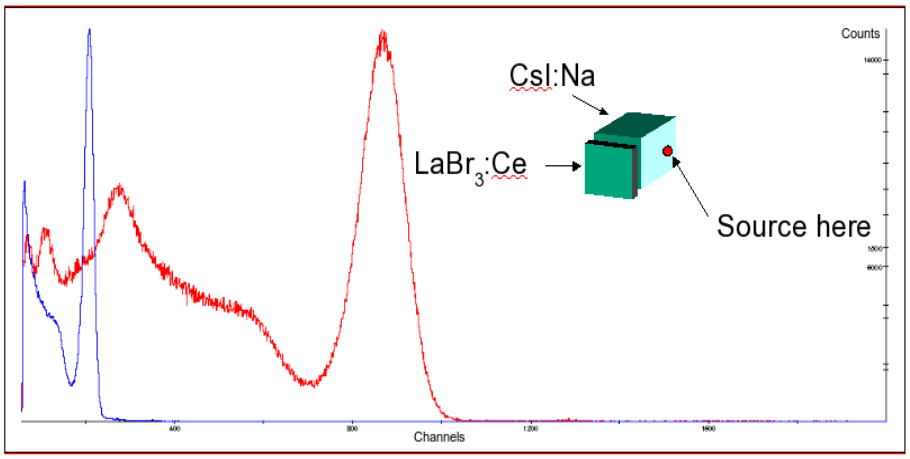

Figure 2: Position sensitivity of the detector when source placed behind

The Phoswich detector is therefore a useful tool with regards to position sensitivity, and could be useful in Positron emission tomography, PET, applications. However, a problem may be encountered if a γ-ray interacts with the middle of the detector close to where the crystals are joined, causing scintillation in both. This is still a problem that is being addressed via addback algorithms to isolate, as best as possible, the tracks of incoming gammas and their subsequent interactions.

Energy measurements were performed using the ORTEC 472A spectroscopy amplifier to determine resolution in the 0.1-2 MeV range. Since it was found that the TFA had previously been seen to give optimal energy resolution, it was compared with the ORTEC 472A amplifier to further any conclusions. Multiple sources were used as calibrations to find the nature of how the energy resolution changed with increasing deposited energy. ^{152}Eu, ^{137}Cs, ^{60}Co and ^{22}Na sources were used, along with activated peaks in ^{80,82}Br as calibration points.

The results were compared to similar studies at higher energies with 2″x2″ $LaBr_3$:Ce scintillators [2], where a fit of $77.10(1.20)E^{-0.5}$ was obtained for energies between 2-20 MeV. However, this fit does not give a good representation of the Phoswich data due to signal degradation, principally from the CsI:Na crystal. Since the resolution is typically higher for this scintillator, the combined signal is worse overall. Large errors were found when calibrating energies less than 100keV due to large amounts of background noise. The best results are given by a logarithmic fit, especially at lower energies. However, with a larger detector, a better fit could be achieved.

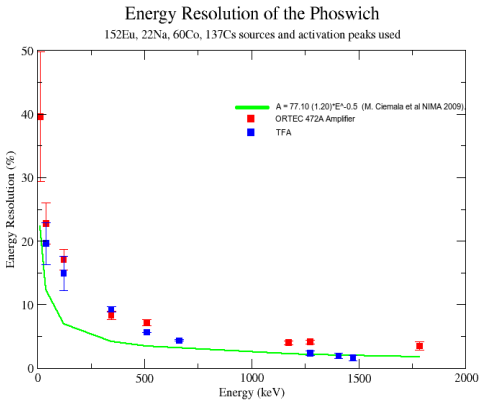

Figure 3: Resulting energy measurements with fitted parameter from Ciemala et al[2].

In addition to this, the position of the source was found to change the energy resolution due to different angles of incident γ rays, which results in scattering and scintillation in both the $LaBr_3$:Ce and CsI:Na detectors. A simple test of moving several sources to different angles and distances was conducted to review this effect. ^{152}Eu, ^{137}Cs and ^{22}Na sources were placed at distances of 0, 5 and 10 cm from the detector, at angles ranging from 0^o to 135^o. Several spectra were then taken to assess where likely points of scattering occured and what effects this had on the resulting energy resolution and subsequent measurements.

These energy measurements were performed with the phoswich detector operating at -1200V, its signal amplified in a standard ORTEC 572 Amplifier. A shaping time of 500ns, and low gain were used in all measurements. While results for angles 0^o to 90^o yielded good results and resolutions with this setup, at larger angles,

more scintillation occured within the CsI:Na segment of the detector resulting in very poor results. In these instances, the shaping time will have to be changed in order to salvage any information about incident γ rays. At the angle of 135^o, the resolution is so poor that no decent results were recorded at this angle. One should also note that another re-occuring problem at angles where scattering in both detectors because increasingly common are double peaks (previously mentioned above).

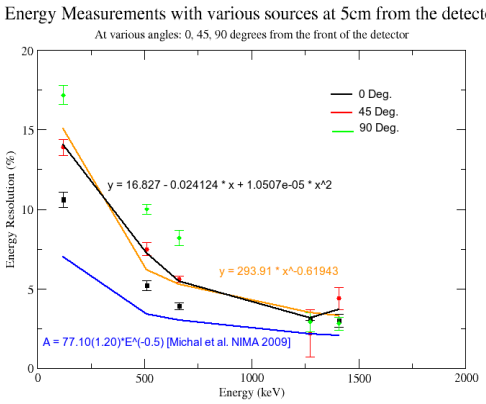

Figure 4: Energy measurements with various sources at 5 cm at various angles.

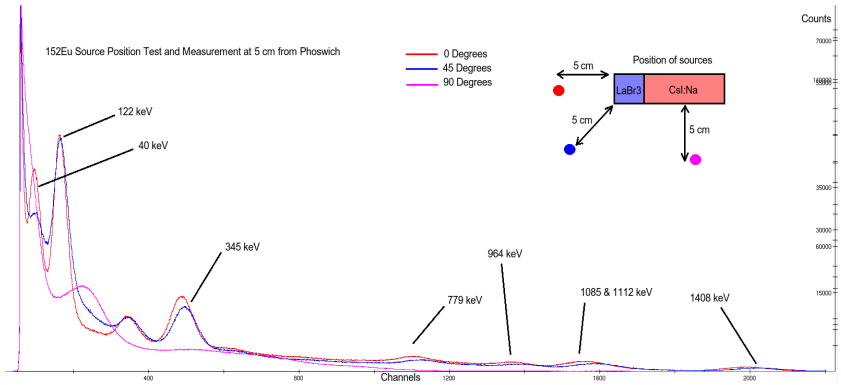

Figure 5: Spectrum showing changes in a ^{152}Eu source at 5 cm due to varying angles.

3. Self-Activity and Neutron Activation in the Phoswich Detector

3.1. Self Activity

"Self-activity" is due to the presence of the radioisotope ^{138}La in $LaBr_3$:Ce, a naturally occurring isotope 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 1436 keV gamma ray. However, this is displaced by 37 keV due to the reoccupation of the K electron shells that result in Barium X-rays. Similarly, coincident L electron shell capture results in the Barium X-rays shifting from the 1436 keV gamma ray by 5 keV, resulting in a slight feature to the left of the main line[3]. The remainder of ^{138}La decays 33.6 % of the time via beta emission to ^{138}Ce, emitting a 789 keV gamma ray from the 2^+ state. A spectrum of these features in $LaBr_3$:Ce is shown in figure 4, outlining its decay processes[3]. This feature will be an underlying problem in the detector, ruling out any practical use in low counting applications. The 1473keV peak was found to have a resolution of 4.7 % with the ORTEC 472A amplifier.

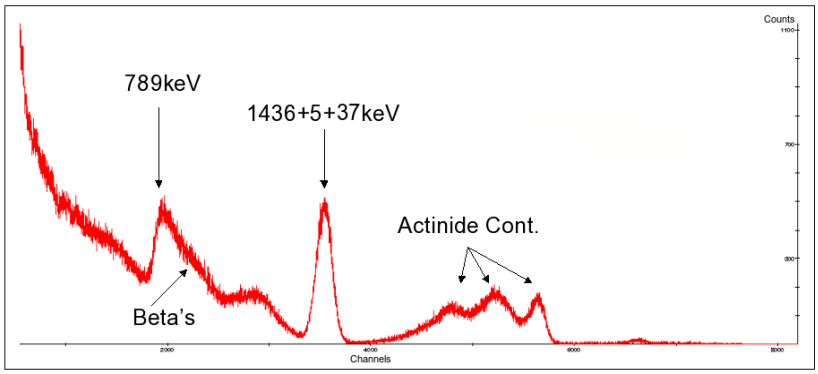

Figure 6: Linear spectrum showing self-activity is gated at lower channels due to the intensity of the K-Shell X-Rays[3].

3.2. Actinide Contamination

Alpha contamination is revealed by 4 or 5 broad peaks. Since ^{227}Ac is also in the same periodic group (group IIIB) as Lanthanum, the presence of alpha decays due to the ^{235}U decay chain is present, with long lived ^{227}Ac being the contributing element ($\tau_{1/2} = 21.2$ years). ^{227}Ac β-decays to ^{227}Th, which subsequently α-decays to ^{207}Tl.

By comparing the true values of these alpha energies and their measured energies, extraction of the properties of alpha scintillation in the detector can be achieved. It was found that alphas produce 65 % less light than gamma rays in the detector; that is, the ratio between the two is 0.35. This is in agreement with a similar finding for $LaCl_3$:Ce scintillators by Hartwell and Gehrke[4].

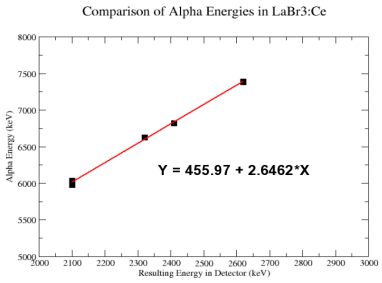

Figure 7: Alpha scintillation properties compared to gamma scintillation

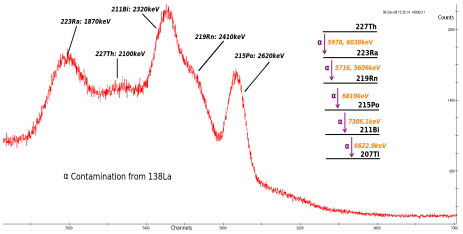

Figure 8: Alpha contamination present in the detector

4. Neutron Response in the Phoswich

4.1. Pulse Shape Discrimination

If re-emission of scintillator luminescence is within 10^{-8}s after absorption, fluorescence occurs. However, if the re-emission is delayed due to an excited metastable state, another process known as phosphorescence occurs. The time evolution of this re-emission process can be accurately modeled:

$$N = Ae^{\frac{-t}{\tau_f}} + Be^{\frac{-t}{\tau_s}} \text{[5]} \tag{1}$$

The finite rise time from zero to the maximum amplitude is usually much shorter than the decay time. For most scintillators, one component of the decay constant is much faster than the other. The existence of these two fast and slow components creates the opportunity for pulse shape discrimination.

Pulse shape analysis was carried out to see if n-γ discrimination was possible. Generally, organic scintillators provide such discrimination due to delayed remission of energy; however inorganic alkali halide crystals, such as $LaBr_3$:Ce

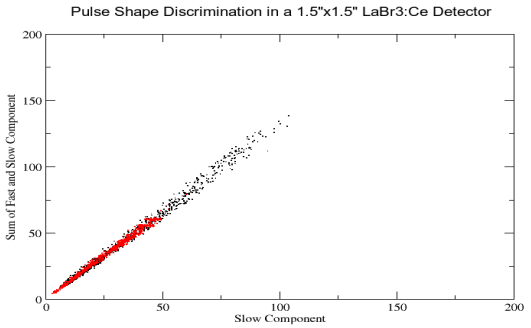

Figure 9: Fast and slow components from gamma and neutron anode pulse shapes to try to observe any discrimination

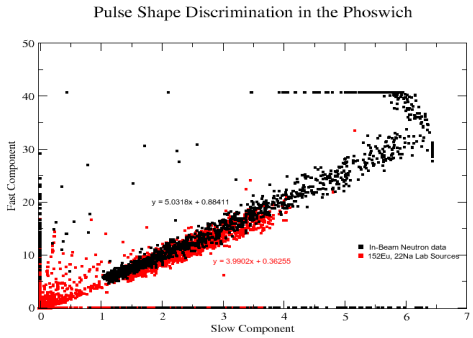

Figure 10: Phoswich Pulse Shape Discrimination over a 2 MeV range

and CsI:Na, do not. Previous experiments were performed with just a 1.5"x1.5" $LaBr_3$:Ce detector using 2 well known gamma-ray emitters: ^{137}Cs (662 keV) and ^{60}Co (1173 & 1332 keV). The slow and fast components of the pulse shapes were obtained 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. The points in red in figure 8, show two thresholds that correspond to the gamma peaks at 1132 and 1332 keV. There is no discrimination between both sets of data, as opposed to 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.

In the Phoswich detector, we see a similar result. It was possible to extract information from the fast and slow components of the pulse by gating on 3 parts of the pulse, of which the 2nd is ignored. This is a good method for discrimination, although the gating requirements need to be strict. The sets of data were fitted twice, each time showing slight differences; this could be due to errors in gating.

4.2. Neutron Activation

It is assumed that there is little n-γ discrimination, if any, as the scintillators are inorganic alkali halides. However, when exposed to a neutron flux, most scintillators will respond at some level. Fast neutrons can be present which will produce prompt signals due to inelastic scattering within the scintillator.

The Phoswich was placed within 20 cm of a sealed 10.5 GBq ^{241}Am/^{9}Be source, having found pile-up a persistent problem at shorter distances. A bias of -1200 V was used to power the detector, and amplification was done with the ORTEC 472A

NIM module. The neutron response was then investigated to study how neutron activation affected the scintillators, and ultimately the detector as a whole. This is seen in the in-beam spectra discussed. However, in-beam tests were seen to activate the detector material.

Elastic neutron scattering resulted in excited states of both Lanthanum (^{140}La) and Bromine (^{80}Br,^{82}Br). The two stable states in naturally occurring Bromine that contribute significantly to the activated in-beam spectrum of $LaBr_3$:Ce are ^{79}Br and ^{81}Br, with abundances of 50.69 % and 49.31 % respectively. The (n,γ) thermal capture cross-sections for the formation of metastable ^{80}Br and ^{82}Br states are 2.5 b and 2.4 b respectively. For the formation of both ^{80}Br and ^{82}Br ground-states, the (n,γ) thermal cross-sections are 8.3 b and 0.24 b respectively[6].

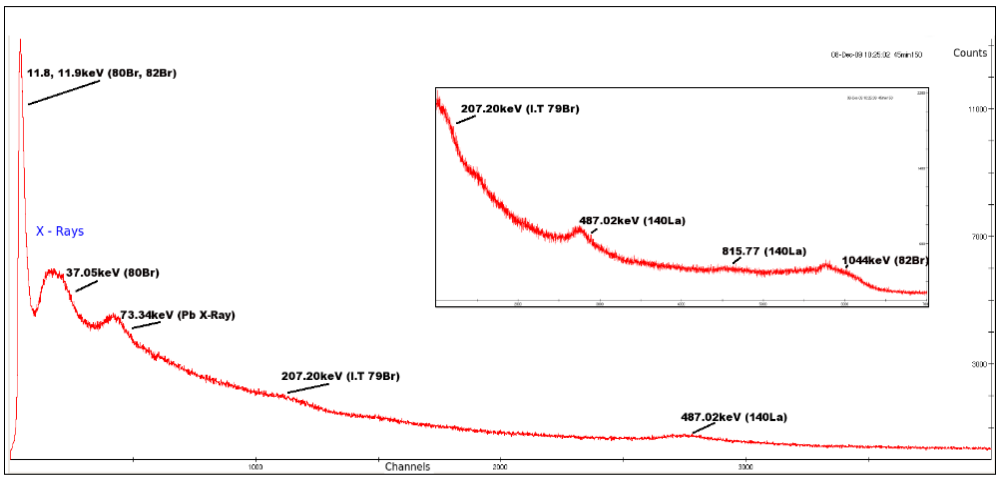

Figure 11: In-beam neutron activation at low energies

When the detector is shielded out of beam in a Pb castle, activation seen from the CsI:Na scintillator is due ^{127}I, and its thermal neutron cross section being sufficiently large. The result is a β-decay with an energy of 832.1 keV and end-point energy of 2119 keV from ^{128}I decaying to ^{128}Xe. While this is a fairly intense decay, it has a short half life: $\tau_{1/2} = 24.99$ min. An 80-hour time-lapse study of the detector in a shielded enviroment shows the result of neutron activation. Multiple 30 minute spectra were taken to show the excited states present in the detector. A few were selected and overlaid to give figure 13.

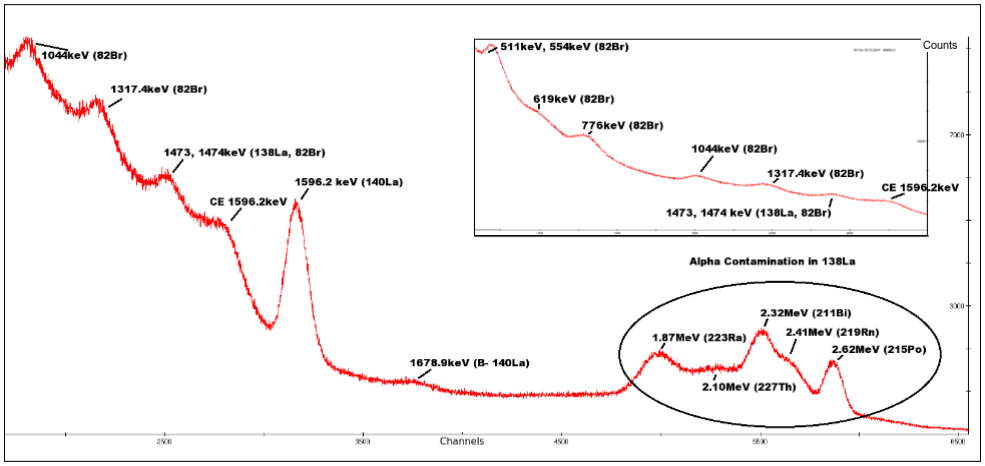

Figure 12: In-beam neutron activation at high energies

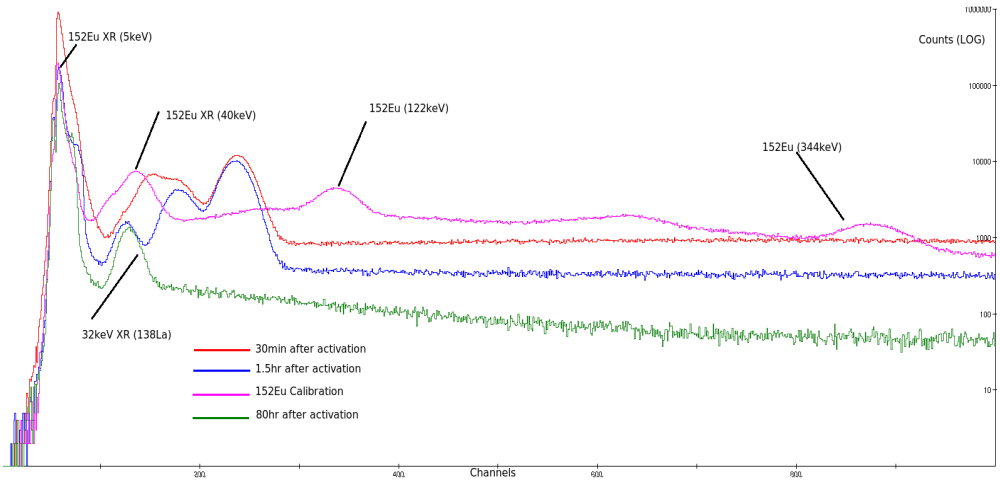

Figure 13: Activated low energy spectra transition back to background over 80hrs

5. Conclusions and Future Work

Tests done on the Phoswich detector show that signals and subsequent data obtained from various NIM amplifiers and other modules resulted in quite poor energy measurements. It is thought a QDC could optimise these results and further this work by discriminating the two pulses and integrating them. A ”fast” scope (1-10 GHz) will be needed as a reliable DAQ, as well as optimisating the output signal. It has been shown that for energies less than 1MeV the fit suggested by

Ciemala et al[2]. for high enrgy γ-rays is in disagreement with the Phoswich data. The position sensitivity was noted as a contributing occurence to signal problems, especially in the case when scintillation in both detectors occur simultaneously. The self-activity, particularly the alpha scintillation properties was examined, obtaining a result similar to what was found for $LaCl_3$:Ce detectors by Hartwell and Gehrke[4]. Activation of neutrons was examined and commented on, showing in-beam activations due to elastic scattering similar to what was seen previously with a similar test in York. Numerous spectra were calibrated and revealed additional gamma lines due neutron capture in both scintillators, and as a result, 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 n-γ discrimination in both the Phoswich and 1.5″x1.5″ $LaBr_3$:Ce was possible, which agrees with previous work[7].

Timing properties will be examined rigourously by performing coincidence measurements with auxiliary detectors. Tests with a larger detector, such as 3″x3″ or even 2″x2″ crystals should be conducted to better verify the results discussed, and also allow for better discrimination of γ rays at higher energies.

6. Acknowledgments

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