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Pulse-shape analysis of CLYC for thermal neutrons, fast neutrons, and gamma-rays

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ABSTRACT

Cs_2LiYCl_6:Ce (CLYC) has been demonstrated to be sensitive to thermal neutrons via the ^{6}Li(n, α)t reaction, and recently to fast neutrons via the ^{35}Cl(n,p) reaction. The scintillation properties of CLYC have been investigated in more detail to further understand its capabilities. Pulses from thermal neutron, fast neutron, and γ-ray induced excitations were captured, digitized over a 16 μs time range, and analyzed to identify the scintillation mechanisms responsible for the observed shapes. Additionally, the timing resolutions of CLYC crystals of different sizes were measured in coincidence with a fast $CeBr_3$ scintillator. The effect of high count rates on fast neutron energy resolution and pulse-shape discrimination was investigated up to 45 kHz.

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1. Introduction

In recent years, Cs_2LiYCl_6:Ce (CLYC) has become a popular neutron/γ scintillator. CLYC functions as both a γ-ray spectrometer with good energy resolution (~4% at 662 keV) and thermal neutron detector, via the ^{6}Li(n, α)t reaction. Its excellent n/γ discrimination has made CLYC especially attractive for a variety of nuclear safeguard applications where ^{3}He tube replacement is needed [1,2].

The response to fast neutrons with CLYC via the ^{35}Cl(n,p)^{35}S and ^{6}Li(n, α) reactions was independently established around the same time by several groups [3–5]. The former reaction is more significant as it leads to relatively sharp peaks (~10%) in the pulse height spectrum in response to mono-energetic neutrons. CLYC can thus function as a fast neutron spectrometer without the need for time-of-flight (TOF), thus opening up an interesting new range of applications. This paper further investigates the timing characteristics for thermal neutrons, fast neutrons, and γ-rays in more detail.

The process of scintillation in CLYC involves several competing mechanisms, which can be dependent on exciting particle, crystal size, and Ce^{3+} concentration [6–8]. For Ce^{3+} doped crystals, the possible mechanisms which are believed to contribute to scintillation include self-trapped exciton (STE) emission, binary V_k-electron diffusion, STE diffusion, direct capture by Ce^{3+}, and core-to-valence luminescence (CVL). Detailed reviews of each mechanism can be found in Refs. [6,7,9,10].

In a direct capture by Ce^{3+}, free electrons and holes are captured by the activator, leading to excitation and 5d→4f optical emission though recombination. Alternatively, free holes produced along the track of ionization are unstable against trapping by the host lattice. Self-trapped holes bound between two anions form what is known as a V_k center. From this point, luminescence may proceed through different mechanisms. The V_k center can migrate through the crystal through thermal motion until it is trapped by Ce^{3+}, forming Ce^{4+}. The subsequent trapping of a free electron leads to luminescence. In Ref. [7] this is referred to as binary V_k and electron diffusion. Rather than migrating after formation, the V_k center may trap a free electron from the conduction band to form a STE. A STE is itself a luminescent center with a typical lifetime on the order of microseconds [7]. The STE is also mobile, and can diffuse through the crystal until it reaches an activator site, resulting in luminescence. Finally, CVL is the radiative recombination of a valence electron from the $3pCl^-$ band to a $5pCs^+$ core hole band [11].

Reports have been published on the lifetimes for some of these mechanisms, primarily under γ-ray excitation, and in some cases for thermal neutrons; however the number of reported components and quoted lifetimes vary [6,8,11–13]. To some extent this

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is expected to be due to the dependence on specific crystal properties. As CLYC has become more popular, the crystal size and Ce^{3+} concentration has become more standardized, making it possible to study the scintillation decay profile and lifetimes for what is now a typical detector. The scintillation produced under fast neutron irradiation has not been previously investigated in detail, so the possibility of fast/thermal neutron pulse-shape discrimination is not certain.

To comprehensively investigate the scintillation response to thermal neutrons, fast neutrons and γ-rays, the pulse-shapes produced by each have been studied for a 2.5×2.5 cm CLYC crystal. Since thermal neutrons result in an α-triton pair and fast neutrons result in a proton, it is not clear whether the pulses produced by each will be the same. High statistics scintillation decay profiles have been collected for all three particle types and fit to extract the lifetimes associated with each mechanism present. Aside from the underlying physics, this information can be used for more advanced pulse-shape discrimination (PSD) techniques such as Gatti's optimal filter [14] and χ^2 pulse fitting.

The timing resolution of CLYC is also important in considering it as a replacement for Li glass in neutron TOF counting. Here, we have measured the timing resolution of five different sized CLYC crystals in coincidence with a fast $CeBr_3$ detector. Finally, the effect of count rates up to 45 kHz in a combined fast neutron/γ environment have been studied to assess its effect on PSD and energy resolution.

2. Experimental setups

2.1. Pulse-shape analysis

A 2.5×2.5 cm cylindrical CLYC crystal, doped with 0.5% Ce^{3+} and enriched with 95% ^6Li, was grown by Radiation Monitoring Devices Inc. (RMD) [15]. It was packaged in an aluminum casing with a reflective inner coating and sealed with a quartz window at one end. The packaging was optically coupled to a Hamamatsu R6231-100 super bialkali PMT.

Fast neutrons were produced using a 5.5 MV Van de Graaff and thin (≈ 10 μm) lithium target evaporated onto a tantalum backing. The target was irradiated with a 3 MeV DC proton beam to produce 1.3 MeV neutrons at 0°. The detector was placed 50 cm from the target.

Data was collected using a 250 MS/s 12 bit CAEN DT5720 digitizer [16] to record pulses directly from the PMT. Signals were collected over a 16 μs time window to sufficiently encompass the entire decay of the pulses.

2.2. Timing resolution

Due to the limited sampling rate of the digitizer, timing resolutions were measured using analog electronics. Five different sized CLYC crystals were measured in coincidence with a 2.5×2.5 cm $CeBr_3$ detector, which has previously been found by the authors to have a resolution of 100–200 ps. The $CeBr_3$ crystal was coupled to a fast Hamamatsu H6610 PMT with a rise time of 0.4 ns. Each CLYC crystal was coupled to a fast PMT made by PR Inc. Four of the CLYC crystals were cylindrical with dimensions of $\emptyset 2.5 \times 2.5$ cm, $\emptyset 5 \times 1$ cm, $\emptyset 5 \times 2.5$ cm, and $\emptyset 5 \times 5$ cm. The fifth had rectangular dimensions of $1.3 \times 1 \times 0.96$ cm, with the shortest dimension in depth. One measurement was made using two $\emptyset 2.5 \times 2.5$ cm CLYC detectors in coincidence.

A CAMAC based multi-parameter data acquisition system was used to record the energies and times of coincident events from a ^{22}Na positron source. This made it possible to set energy gates on the 511 keV photons in each detector and eliminate chance coincident events.

2.3. Count rate dependence

The effect of high counts rates on the 2.5×2.5 cm CLYC crystal was measured using the same setup and neutron energies described in Section 2.1. The beam current was increased to ~ 10 μA to increase the neutron flux. Higher count rates were achieved by moving the detector closer to the target. The detector to target distances used were 50 cm, 25 cm, 12 cm, and 6 cm. Traces were collected in a 2 μs time window. The count rate was determined by the data acquisition software from the observed trigger rate.

3. PSD optimization

Excellent PSD in CLYC can be achieved using the well known charge comparison method [17–19]. The PSD parameter used here was defined as the ratio of prompt to delayed pulse integration windows, $PSD = Q_{prompt}/Q_{delayed}$, where Qrepresents the integrated charge. The prompt integration window starts at the zero point, and the delayed window usually begins at the end of the prompt window. An example 2D plot of the PSD parameter versus pulse height is shown on the left of Fig. 1 for a moderated PuBe source. On the right of the figure is a projection of the PSD parameter axis for events within the boxed region, which encompasses the thermal neutron peak.

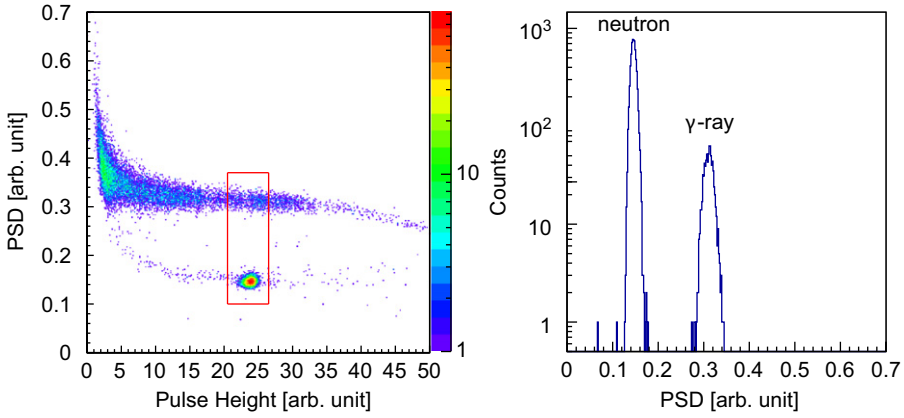

Fig. 1. Left: 2D PSD plot for thermal neutrons and γ-rays. Right: PSD parameter for n/γ events within the boxed region of the PSD plot.