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Comparison of model fitting and gated integration for pulse shape discrimination and spectral estimation of digitized lanthanum halide scintillator pulses

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ABSTRACT

An analysis of digitized pulse waveforms from experiments with $LaBr_3(Ce)$ and $LaCl_3(Ce)$ detectors is presented. Pulse waveforms from both scintillator types were captured in the presence of ^{22}Na and ^{60}Co sources and also background alone. Two methods to extract pulse shape discrimination (PSD) parameters and estimate energy spectra were compared. The first involved least squares fitting of the pulse waveforms to a physics-based model of one or two exponentially modified Gaussian functions. The second was the conventional gated integration method. The model fitting method produced better PSD than gated integration for $LaCl_3(Ce)$ and higher resolution energy spectra for both scintillator types. A disadvantage to the model fitting approach is that it is more computationally complex and about 5 times slower. $LaBr_3(Ce)$ waveforms had a single decay component and showed no ability for alpha/electron PSD. $LaCl_3(Ce)$ was observed to have short and long decay components and alpha/electron discrimination was observed.

1. Introduction

Lanthanum halide scintillators have excellent energy resolution, timing resolution and detection efficiency for gamma rays, which makes them ideal for many gamma ray spectroscopy applications. However, their responses are complicated by self-activity. The key features of the energy spectra of $LaBr_3(Ce)$ and $LaCl_3(Ce)$ in the presence of ^{22}Na and ^{60}Co sources are shown in Figs. 1 and 2 respectively. Spectral features are very similar for the two scintillators. The decay of ^{138}La to ^{138}Ba (66.4% branching ratio) yields a 1.473 MeV sum peak from a 1.436 MeV gamma ray coincident with 0.032 MeV and 0.005 MeV X-rays. The decay of ^{138}La to ^{138}Ce (33.6% branching ratio) yields a continuum from 0 to 0.255 MeV due to the 0.255 MeV endpoint β^- and a continuum from 0.789 MeV to 1.044 MeV due to the coincident emission of a 0.789 MeV gamma ray with the β^- particle [1,2]. Alpha contamination due to ^{227}Ac also provides a broad response with several peaks from roughly 1.700–2.300 MeVee (MeV electron equivalent) [2]. Count rates are approximately 1–2 and 0.1 counts/s/cm^3 for contamination from ^{138}La and ^{227}Ac respectively, although contamination from ^{227}Ac has been significantly reduced in more recently manufactured crystals. Contamination may interfere with low activity-level counting. If the alpha particle response could be distinguished from the electron response, events due to ^{227}Ac activity could be suppressed.

For applications involving thermal and fast neutron-induced gamma rays, a large number of neutron-induced reactions are energetically favourable, which may further complicate the response function. For example, the National Nuclear Data Centre data base lists 87 neutron-induced reactions for La, Br and Cl targets which are energetically favourable for 14 MeV neutrons [3]. Many of these reactions produce heavy (compared to electrons) charged particles, such as protons, deuterons, tritons, ^3He and alpha particles. If the responses of these particles could be distinguished from those due to electrons, neutron events could be distinguished from gamma ray events.

Past studies have yielded contradictory results for pulse shape discrimination (PSD) of alphas and gamma rays in $LaBr_3(Ce)$. Hoel et al. [4] determined that PSD was not possible because the light emission of $LaBr_3(Ce)$ has no measurable slow component. However, other studies [5,6] have found small, but measurable shape differences between alpha and gamma pulses, although the PSD mechanism is not understood. Pulse shape discrimination can definitely be achieved with $LaCl_3(Ce)$ [4]. The PSD method used in that study was inadequate to separate protons from deuterons and tritons, but this does not necessarily preclude the ability to achieve neutron/gamma ray discrimination with $LaCl_3$. If indeed possible,

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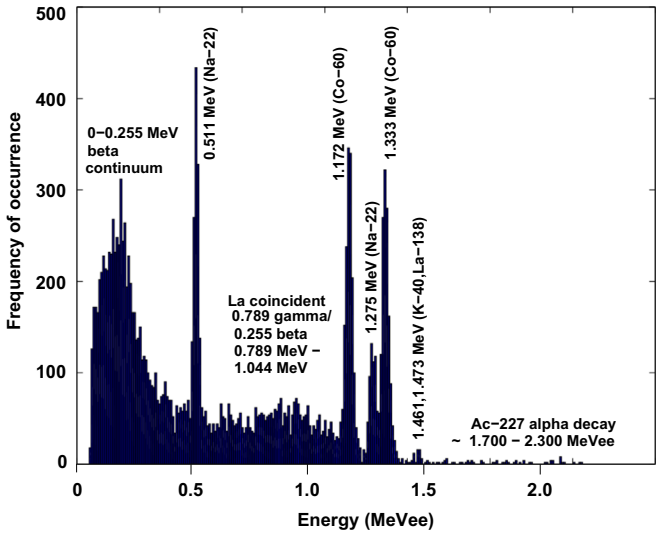

Fig. 1. Energy spectrum of $LaBr_3(Ce)$ with ^{22}Na and ^{60}Co sources present. Histogram was derived from physics-based model fitting from experiments to be described, using a fast amplifier and coincidence mode.

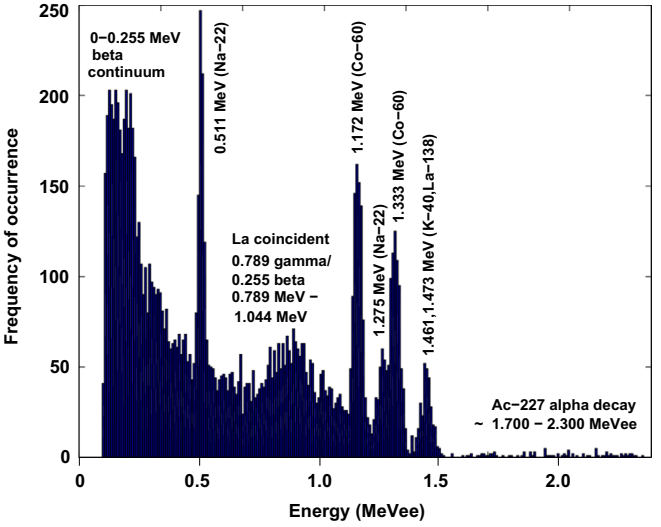

Fig. 2. Energy spectrum of $LaCl_3(Ce)$ with ^{22}Na and ^{60}Co sources present. Histogram was derived from physics-based model fitting from experiments to be described, using a fast amplifier and coincidence mode.

this could be very useful in applications involving mixed gamma ray/fast neutron fields and would give $LaCl_3$ a distinct advantage over $LaBr_3$ in such applications, even though most of the scintillation properties of $LaCl_3$ are slightly inferior to those of $LaBr_3$.

As a first step towards carrying out gamma ray/fast neutron discrimination experiments, methods to analyse and discriminate pulse shapes were evaluated. There are numerous methods, some suited to analogue signals or real-time analysis and others to digitized signals. Two methods of extracting PSD parameters and energy spectra were selected from the latter and their performances in discriminating alpha and electron-induced responses were compared. The first, gated integration, has been commonly used for PSD in organic scintillators [7], and has also been used for PSD in lanthanum halide scintillators [4–6]. It is fairly simple to implement for digital waveforms and has been shown to perform well. The second method, physics-based model fitting, has not been used before for this application. However, model fitting to empirical templates has been used infrequently [6,7]. There are few studies comparing the two methods for lanthanum halide scintillators. Zeng et al. [6] found that the "charge comparison" method (gated integration) performed slightly better for PSD of digitized $LaBr_3(Ce)$ pulses than a template-based model fitting method, which in turn was better than the mean time method. The present study proposes a parametric physics-based description of pulse shape as the model for nonlinear least squares fitting. Although more complicated to implement than the gated integrator, it will be seen that the model fitting method yields higher resolution energy spectra for $LaBr_3(Ce)$ and $LaCl_3(Ce)$ and better PSD for $LaCl_3(Ce)$. The study shows no PSD ability for $LaBr_3(Ce)$ for either method, in agreement with [4] and in contrast with [5,6].

The relevant theory of pulse shapes is discussed in Section 2. The experiments to collect pulse waveforms for analysis by the two methods are described in Section 3. Sections 4 and 5 present results of PSD and energy spectral estimation from the two methods. The results are discussed in Section 6 and conclusions and future work are described in Section 7.

2. Pulse shape theory

Discriminating incident gamma rays from incident neutrons and internal alpha activity requires analysing the shape of individual pulse waveforms to separate electron-induced excitation and ionization from that due to the heavy charged particles.

The scintillation decay time constants of $LaBr_3(Ce)$ and $LaCl_3(Ce)$ as a function of temperature are found in a number of published articles [8]. Assuming excitation at time $t=0$, the light emission intensity of $LaBr_3(Ce)$ can be accurately modelled as a single fast component exponential decay, whose intensity I is

$$I(t; A, \lambda) = A\lambda e^{-\lambda t} \tag{1}$$

where t is the time, $A\lambda$ is the intensity at $t=0$ and the decay constant $\lambda = 1/\tau$ is the inverse of the time constant (mean lifetime) τ. At room temperature, τ is approximately 15–26 ns. (The values for both $LaBr_3(Ce)$ and $LaCl_3(Ce)$ are a function of cerium concentration, as well as temperature.) The light emission intensity of $LaCl_3(Ce)$ is well modelled by a two component exponential decay:

$$I(t; A_1, \lambda_1, A_2, \lambda_2) = A_1\lambda_1 e^{-\lambda_1 t} + A_2\lambda_2 e^{-\lambda_2 t} \tag{2}$$

where the subscript 1 refers to the fast component and subscript 2 refers to the slow component. At 20 °C, τ_1 is approximately 26 ns and τ_2 is approximately 550 ns. The ratio of A_2/A_1 is approximately 0.25 for gamma rays at 20 °C [8].

The activation centres of the scintillator are not populated instantaneously at $t=0$ and so there is an exponential growth component as well as the decay component. For $LaBr_3(Ce)$ and $LaCl_3(Ce)$, the growth time constant is roughly between 0.7 and 3.5 ns [9]. The combination of electrical resistance and parasitic capacitance of the photomultiplier contributes an additional exponential growth component. Assuming an effective combined growth component time constant, $\tau_0 = 1/\lambda_0$, the scintillator/photomultiplier voltage response can be represented by exponential growth/decay models:

$$V(t; A, \lambda_0, \lambda) = A\lambda_0\lambda/(\lambda - \lambda_0)(e^{-\lambda_0 t} - e^{-\lambda t}) \tag{3}$$

$$\begin{aligned} V(t; \lambda_0, A_1, \lambda_1, A_2, \lambda_2) &= A_1\lambda_0\lambda_1/(\lambda_1 - \lambda_0)(e^{-\lambda_0 t} - e^{-\lambda_1 t}) \\ &\quad + A_2\lambda_0\lambda_2/(\lambda_2 - \lambda_0)(e^{-\lambda_0 t} - e^{-\lambda_2 t}) \end{aligned} \tag{4}$$

However, τ_0 is usually sufficiently small with respect to the effective time constant of the electronics which follow, so that the growth component can be neglected. In that case the scintillator/