Available online at www.sciencedirect.com
SCIENCE DIRECT®
Nuclear Instruments and Methods in Physics Research A 540 (2005) 205–208
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Pulse-shape discrimination of La halide scintillators

C. Hoel[a], L.G. Sobotka[a,*], K.S. Shah[b], J. Glodo[b]

[a]*Department of Chemistry, Washington University, Campus Box 1134, St. Louis, MO 63130, USA*
[b]*Radiation Monitoring Devices, Watertown, MA 02472, USA*

Received 27 July 2004; received in revised form 3 December 2004; accepted 6 December 2004

Abstract

The bright scintillators $LaCl_3$:Ce and $LaBr_3$:Ce were tested for α–γ pulse-shape discrimination (PSD). PSD is demonstrated for $LaCl_3$:Ce but not for $LaBr_3$:Ce.

PACS: 29.40.Mc

Keywords: Inorganic scintillators; Pulse-shape discrimination

The La halide scintillators ($LaCl_3$:Ce and $LaBr_3$:Ce) are extremely bright, fast, and hygroscopic. Due to the brightness and time response of these scintillators, they have the potential to serve as detectors of intermediate resolution between other scintillators and solid-state detectors. With resolution of 3% for 662 keV photons, 2–3 times better than NaI(Tl), they are likely to find many applications. One example is in fast-beam spectroscopy, where Doppler broadening prevents the attainment of the intrinsic resolution of solid-state detectors. Another application is for intermediate to high-energy charged-particle detection when sufficient stopping cannot be achieved with solid-state detectors. In this second case, the ability to separate e^-'s, p's, and α-particles, is essential.

While the gross light output, energy resolution and pulse shape for incident photons, has been studied by two groups [1,2], nothing has been reported to date on the pulse-shape discrimination (PSD) capabilities of these relatively new scintillators. This work partially fills this void.

The scintillators were grown by the Bridgeman method (by RMD) with 0.5%Ce. The $LaCl_3$ crystal was $8\,mm \varnothing \times 3\,mm$ while the $LaBr_3$ crystal was $7\,mm \times 7\,mm \times 2\,mm$. To prevent water from being absorbed, a polymer coating was spun onto what would become the radiation exposed surface [3]. We used polyethylene co-vinyl acetate (18% vinyl acetate) dissolved in toluene for this purpose, a choice motivated by the

*Corresponding author. Fax: +1 314 935 6184.
E-mail address: lgs@wuchem.wustl.edu (L.G. Sobotka).

0168-9002/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2004.12.029

requirement of using a hydrophobic polymer and a non-polar solvent. The thickness of the polymer coating was estimated to be 1.0 mg/cm^2 by capacitive and mass measurements of foils spun on phantom detectors. The thickness of this spun layer, and its non-uniformity, strongly affects the alpha energy resolution. The relative energy resolution will improve with increasing α-particle energy, as the fractional (as well as the total) energy dropped in the covering dead layer decreases.

The scintillator was attached with a high viscosity optical grease to a Hamamatsu R7400 compact metal-channel PMT and the remaining sides painted with TiO epoxy paint. The whole assembly was covered in 250 μg/cm^2 aluminized mylar. All testing was done in a vacuum chamber.

Time spectra where collected by signal averaging of 256 events out to 950 ns after triggering. These average traces, area normalized, are shown in Fig. 1 for $LaCl_3$:Ce for α-particles from ^{249}Cf and γ′s and β′s from ^{232}U (with a thick epoxy coating.). The light output for α-particles is suppressed, relative to that for γ-rays, in the 10 to 65 ns window. Conversely, the tail is enhanced for α-particles. The same trend is observed in $LaBr_3$:Ce however the magnitudes of the slow components are far less.

The time dependence of the light output for both new scintillators and CsI:Tl, for both

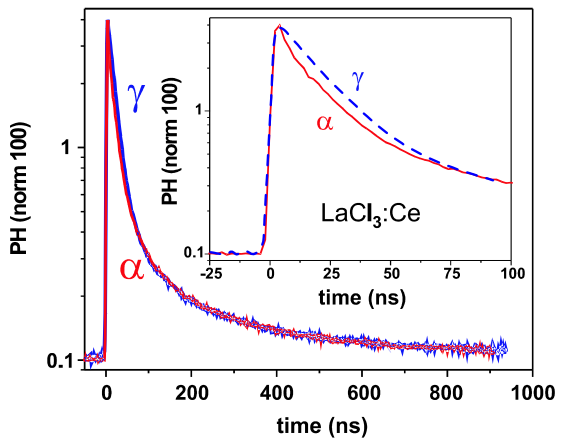

Fig. 1. Normalized light output from $LaCl_3$:Ce from α-particles and γ-rays (dashed in inset figure). The pulse-height spectra are normalized to the same area.

γ/e$^-$and α-particles, were fit with a multi-component exponential form with offset, pulse height $= \sum_{i=1}^{3} A_i e^{-t/\tau_i} + C$. The mean decay times τ_i were free parameters for the fits for the γ/e$^-$ data and fixed to the same values when fitting the α-particle data. The difference in pulse shape required decreasing (increasing) the amplitude of the fast (intermediate) lifetime component. This trend is opposite to that for the most common inorganic scintillator used for charged-particle discrimination, CsI(Tl), for example see [4,5]. If a figure of merit is constructed from the area difference in fast component (or the sum of the areas of the intermediate and slow component), the PSD of $LaCl_3$:Ce is about half that of CsI:Tl. Such a figure of merit (which ignores noise) is larger for $LaBr_3$:Ce than $LaCl_3$:Ce, however $LaBr_3$:Ce suffers from reduced absolute flux of delayed light (Table 1).

The particle time resolution is studied using α–γ (start-stop) from ^{249}Cf. The spectrum is shown in Fig. 2. A 25 ∅ mm × 40 mm CsF scintillator was used to detect the γ-ray (stop) depopulating the 388 keV level, fed by the α-decay, with a $t_{1/2} =$ 0.450 ns [6]. Fitting our data with an exponential convoluted with a Gaussian, reproduced the literature decay constant and provided a value of the coincidence time resolution of $\sigma = 0.30 \pm$ 0.02 ns. We estimate the single-ended time resolution of the CsF (to 380 keV photons) to be ∼ 200 ps, thus σ_{LaBr_3} (5.8 MeV α′s) ∼ 250 ps. This is similar to the value obtained for photons [2].

The quality of this timing is such that light charged particle (p,d,t, and α′s) mass identification, even with very modest flight distances

Table 1
Areas and time constants of La-based scintillators. The values in parentheses are fixed

Material	Source	1:$\frac{A}{\tau}$:τ(ns)	2:$\frac{A}{\tau}$:τ(ns)	3:$\frac{A}{\tau}$:τ(ns)
$LaCl_3$	γ, e$^-$	0.56,18.9	0.22,107	0.022,515
	α	0.47(18.9)	0.28(107)	0.25(515)
$LaBr_3$	γ, e$^-$	0.97,26	0.03,159	0.002,565
	α	0.81(26)	0.15(159)	0.04(565)
CsI:Tl	γ, e$^-$	0.66,400	0.33,1200	0.01,8500
	α	0.83(400)	0.17(1200)	0.008(8500)

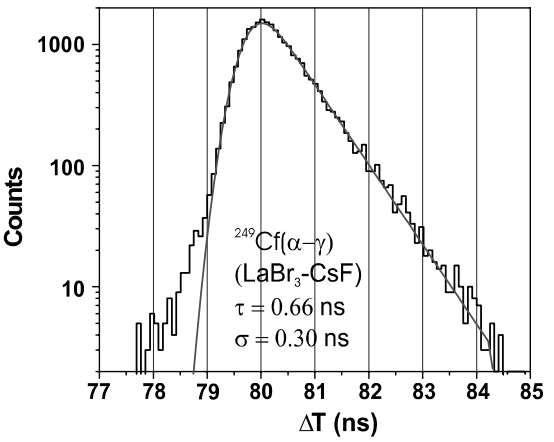

Fig. 2. Alpha–gamma time-to-amplitude spectrum and fit.

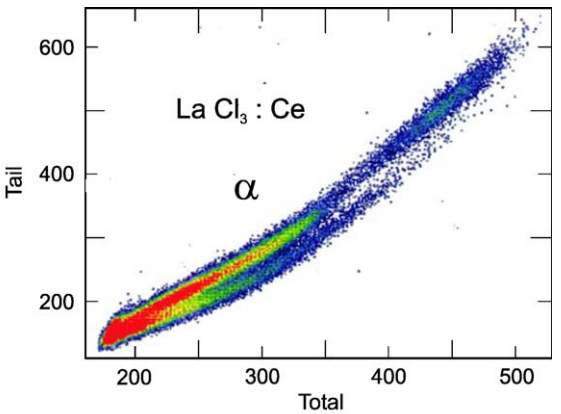

Fig. 3. Standard PSD plot of total (abscissa) and partial (ordinate) integration of the pulses produced by $LaCl_3$:Ce exposed to a ^{232}U source. The locus for α-particles shows two groups, one at lower energy composed of many closely spaced lines and a higher energy peak from the 8.8 MeV decay of ^{212}Po. The e^- ridge is below that for α-particles.

(10 cm), can be done for evaporation energies if a high-quality start is available.

Implementation of PSD was done using the standard scheme of partial integration of the current pulse. Fig. 3 shows the results for $LaCl_3$:Ce where the abscissa is the total integral with a 1000 ns wide gate, while the ordinate is the integral from 75 ns after prompt out to 1000 ns. The α-particle locus is above that for e^-s in this representation. For this study, α′s and γ′s come from ^{232}U decay. The α-particle locus is readily identified as the one with excess (relative) tail and the separated peak corresponding to the 8.8 MeV α-particle (approximately 8.0 MeV after the energy loss in the polymer coating) from the decay of ^{212}Po. The end of the γ-line is the Compton edge of the final decay step of the decay chain, $^{208}\mathrm{Tl} \xrightarrow{\beta} {}^{208}\mathrm{Pb}^* \rightarrow {}^{208}\mathrm{Pb}$ (gd.st.). Using these energies and the appropriate channel numbers, we deduce an approximate quenching factor of 3 for the more highly ionizing α-particles.

A separate study using a FRONT gate (10–75 ns after prompt) rather than a TAIL gate yielded equal quality PSD. While the PSD for $LaCl_3$:Ce is adequate to separate α-particles and γ-rays down to low energies, it is inadequate to separate p's from d's and t's. A higher concentration of Ce might improve the particle identification quality. Our attempts to discriminate between α-particles and recoil e^-s in $LaBr_3$:Ce failed. We attribute this to the small amplitude (and thus poor signal-to-noise ratio) of light associated with the TAIL.

La halide scintillators have been tested for pulse-shape discrimination. The amplitude of the component with the shortest decay constant is suppressed with increasing dE/dX. The PSD for $LaCl_3$:Ce is modest (about $\frac{1}{2}$ that of the commonly used scintillator CsI:Tl) and of potential use while that for $LaBr_3$:Ce is too small to be useful. The PSD of $LaCl_3$:Ce, coupled with its high light output and excellent time resolution, make it a potential choice for high-rate charged-particle detectors. The major drawback of this material is that it is far more hygroscopic than CsI:Tl (or NaI:Tl.) The simple technique, used in the present study, of spinning a polymer coating on the entrance face may provide a means for using these scintillators when thin entrance windows are required.

This work was supported in part by the U.S. Department of Energy, Division of Nuclear Physics under Grant no. DE-FG02-87ER-40316.

References

[1] E.V.D. van Loef, P. Dorenbos, C.W.E. van Eijk, K. Krämer, H.U. Güdel, Appl. Phys. Lett. 79 (2001) 1573.

[2] K.S. Shah, J. Glodo, M. Klugerman, W.W. Moses, S.E. Derenzo, M.J. Weber, IEEE Trans. Nucl. Sci. (2003).
[3] E. Norbeck, T.P. Dubbs, L.G. Sobotka, Nucl. Instr. and Meth. A 262 (1987) 546.
[4] D.W. Stracener, D.G. Sarantites, L.G. Sobotka, J. Elson, J.T. Hood, Z. Majka, V. Abenante, A. Chbihi, D.C. Hensley, Nucl. Instr. and Meth. A 294 (1990) 485.
[5] D.G. Sarantites, P.-F. Hua, M. Devlin, L.G. Sobotka, J. Elson, J.T. Hood, D.R. LaFosse, J.E. Sarantites, M.R. Maier, Nucl. Instr. and Meth. A 381 (1996) 418.
[6] R.B. Firestone, V.S. Shirley, C.M. Baglin, S.Y.F. Chu, J. Zipkin, Table of Isotopes, eighth ed., Wiley, New York, 1996.