Non-Proportionality and Energy Resolution of a $LaCl_3$:10% Ce^{3+} Scintillation Crystal

E. V. D. van Loef, W. Mengesha, *Member, IEEE*, J. D. Valentine, *Member, IEEE*, P. Dorenbos, and C. W. E. van Eijk, *Member, IEEE*

***Abstract*—The electron and photon response of an aluminum canned $LaCl_3$:10% Ce^{3+} crystal were measured using the Compton Coincidence Technique (CCT). The $LaCl_3$:10% Ce^{3+} electron response increases from 7 keV to 30 keV by about 10%. Above 30 keV, the electron response levels, i.e., it is flat within 5%. Monte Carlo N Particle code (MCNP4C) was used in the photon response calculation. The calculated theoretical photon response is in good agreement with the measured photon response. An energy resolution (full width at half maximum over the peak position) of $4.2 \pm 0.5\%$ was observed for the 662 keV full absorption peak. The energy resolution as function of photon energy exhibits a linear relationship with the inverse square root of the energy. The step like curves of NaI(Tl) with a semi plateau in the energy range between 100 and 500 keV have not been observed for $LaCl_3$:10% Ce^{3+}.**

***Index Terms*—Energy resolution, $LaCl_3$:Ce^{3+}, non-proportionality, scintillation.**

I. Introduction

When evaluating a scintillator for its applicability in gamma-ray spectroscopy and dedicated detector systems, light yield, scintillation decay time, and energy resolution are considered to be critical properties. Also the density of the material and production costs play an important role. With respect to scintillation light yield, it is desirable that it is proportional to the energy of the gamma to be detected. Deviations, which are known as nonproportionality (or nonlinearity) in the scintillation response (or efficiency), may result in difficulties in the determination of the energy and energy resolution of the detected radiation.

The phenomenon of nonproportionality response and its relation with energy resolution has been studied in detail for many classical scintillators, especially for NaI(Tl) [1]–[10]. For NaI(Tl) and most other alkali halides, the scintillation response peaks between 10 and 15 keV and decreases as the photon energy is increased. Finally, the scintillation response levels at higher energy [11]. Contrary to alkali halides, oxide based scintillating materials in general show an increasing scintillation response with increasing photon energy, which levels at higher energies [11]–[15].

In principle, scintillation light yield nonproportionality can be characterized as a function of either photon or electron energy. While the scintillation response as a function of photon energy (photon response) is in general easier to measure and is more useful as an indication of scintillator quality to the customer, the scintillation response as function of electron energy (electron response) is more fundamental. Meaning that, in principle, the photon response can be calculated from the measured electron response using dedicated Monte Carlo codes. Obviously, it is preferable to measure both the photon response and the electron response of the scintillator in question.

A technique for measuring the electron response of scintillators is the so-called Compton Coincidence Technique (CCT) [16], [17]. In short, this method measures the light yield generated in a scintillator when a Compton scattered electron is absorbed. The Compton scattered gamma ray is detected in a secondary detector, such that the energy of the scattered gamma ray and thus the energy of the Compton electron is known. When the scatter angle is varied, the energy of the Compton electron changes as well. Accordingly, the electron response for that particular scintillator is obtained.

In this paper, we present the scintillation response and energy resolution of aluminum canned $LaCl_3$:10% Ce^{3+} crystal. Both the electron and photon response are reported. The calculated theoretical photon response and the energy resolution as a function of photon energy are presented as well.

II. Experimental Technique

An aluminum canned $LaCl_3$:10% Ce^{3+} scintillator, manufactured by Saint-Gobain, of 16 mm diameter and 19 mm height was mounted on a Philips XP2020Q photomultiplier tube (PMT). It was used in conjunction with a high purity germanium (HPGe) detector in CCT. Details on CCT are given in [16] and [17]. The electron response of $LaCl_3$:10% Ce^{3+} was measured from 7 to 433 keV by varying the scatter angle. In order to obtain the photon response curve for $LaCl_3$:10% Ce^{3+} we used ^{241}Am, ^{57}Co, ^{133}Ba, ^{137}Cs, and ^{22}Na γ-ray sources to excite the scintillator at energies between 59 and 1.274 MeV and a variable X-ray source from Amersham (AMC.2084) to excite the scintillator at energies between 17.4 and 59 keV. A Hamamatsu R1791 PMT was used to obtain the pulse height spectra for energy resolution measurements.

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E. V. D. van Loef, P. Dorenbos, and C. W. E. van Eijk are with the Radiation Technology Group, Interfaculty Reactor Institute of Delft University of Technology, 2629JB Delft, The Netherlands (e-mail: loef@iri.tudelft.nl; dorenbos@iri.tudelft.nl; vaneijk@iri.tudelft.nl).

W. Mengesha is with the Neely Nuclear Research Center, Georgia Institute of Technology, Atlanta, GA 30332 USA (e-mail: gt7875a@prism.gatech.edu).

J. D. Valentine is with the Lawrence Livermore National Laboratory, Livermore, CA 94550-9234 USA (e-mail: valentine12@llnl.gov).

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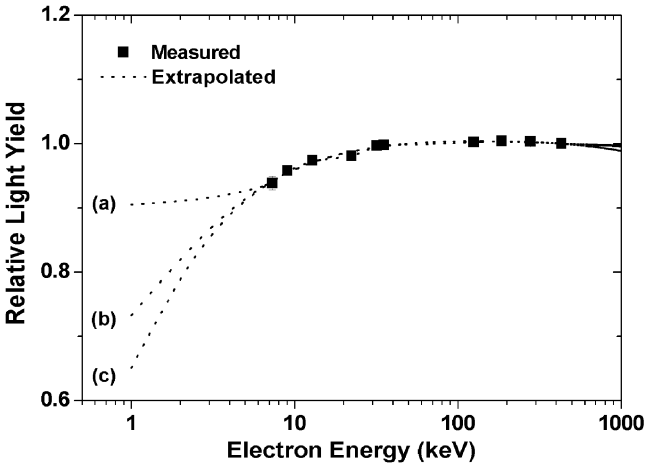

Fig. 1. Electron response of $LaCl_3$:10% Ce^{3+}. The smooth functions are the extrapolated curve fits of the electron response with relative light yield (a) 0.9; (b) 0.5; and (c) 0.0 at electron energy 0.0 keV.

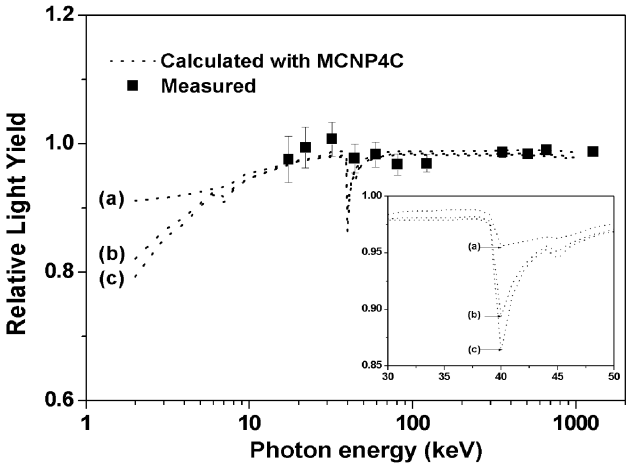

Fig. 2. Photon response of $LaCl_3$:10% Ce^{3+}. The dotted curves are the calculated photon responses according to the electron response with relative light yield (a) 0.9; (b) 0.5; and (c) 0.0 at electron energy 0.0 keV. The inset shows part of the calculated photon responses, between 30 and 50 keV, on a linear energy scale.

III. Theoretical Photon Response Calculation

The theoretical photon response R_γ was calculated by discrete convolution of the measured electron response $R_{e,i}$ with the primary electron energy spectrum $\Psi_{i,n}$ obtained as a result of N full-energy gamma-ray absorption events in the scintillator. A total of k primary electron energies are produced for each full gamma energy absorption event, where k may vary from event to event

$$R_\gamma = \frac{1}{N}\sum_{n=1}^{N}\sum_{i=1}^{k}\Psi_{i,n}R_{e,i}. \tag{1}$$

The MCNP4C code [18] was used to sample to the primary electron spectrum, with N of the order of 10^5 events. All interactions (Compton scatter and photoelectric absorption for 1 keV $< E_\gamma \leq$ 1000 keV) for each simulated photon were recorded. Since each event is terminated by a photoelectric absorption, a cascade of X-rays and Auger electrons is also produced. The cascade sequence associated with the K, L, and M shells of the lanthanum, and to a lesser degree chlorine, was also simulated with MCNP4C.

IV. Results and Discussion

The electron response of $LaCl_3$:10% Ce^{3+} is shown in Fig. 1. The relative light yield as function of electron energy increases from 7 to 30 keV by about 10%. Above 30 keV, the electron response levels, i.e., it is flat within 5%. Here, relative light yield is defined as the light yield relative to that at 662 keV. Accurate CCT measurements below 7 keV electron energy could not be achieved due to interfering PMT thermal noise. The calculated photon response based on (1) requires a knowledge of the electron response at discrete energies ranging from the lowest energy Auger electron from the cascade sequence to the largest photon energy (typically 1 MeV). Consequently, both low and high-energy extrapolations of the measured electron response, as well as a smooth fit of the data are required. These extrapolations are shown in Fig. 1 as well.

The $LaCl_3$:10% Ce^{3+} photon response calculated by (1) using the measured electron response is shown in Fig. 2. Measured photon response data is shown as well. There is good agreement between the calculated and measured photon response; the observed deviation between calculation and measurement is less than 5%. The general shape of the $LaCl_3$:10% Ce^{3+} photon response, as seen in Fig. 2, is very similar to that of K_2LaCl_5:10% Ce^{3+} [11]. For K_2LaCl_5:10% Ce^{3+}, the photon response gradually increases by 15%–20% between 6 and 20 keV. For energies larger than 30 keV the photon response of K_2LaCl_5:10% Ce^{3+} levels, i.e., is flat within 5%. Also for $YAlO_3$:Ce a constant light yield response from 14 keV to 1.3 MeV was observed [19].

The intensity of the calculated photon response at the K edge of La (38 keV) seems to depend critically on the behavior of the electron response between electron energies 0.0 and 10 keV. If the electron response levels at a relative light yield of 0.9 at $E_e = 0$ keV: (a) the calculated photon response decreases by about 3% from 30 to 40 keV. However, if the electron response levels at a relative light yield of 0.5 at $E_e = 0$ keV, (b) the calculated photon response decreases by about 7% from 30 to 40 keV and, if the electron response monotonically decreases to zero at electron energy $E_e = 0$ keV, (c) the calculated photon response decreases by about 12% from 30 to 40 keV. Apparently, the contribution of (low energy) Auger electrons to the calculated photon response is significant, especially near the K edge. This becomes clear if we compare the primary electron spectra for γ-ray energies E_γ of 30, 40, and 50 keV, shown in Fig. 3. Apparently, at 40 keV (just above the K edge of La) electrons are produced with a relatively low energy, i.e., $E_e < 10$ keV, whereas for γ-ray energies of 30 and 50 keV, electrons are produced with a relatively high energy, i.e., $E_e > 10$ keV.

Fig. 4 shows the energy resolution as function of photon energy between 17.4 keV and 1.274 MeV. Also shown is the PMT resolution calculated from the number of photoelectrons, and

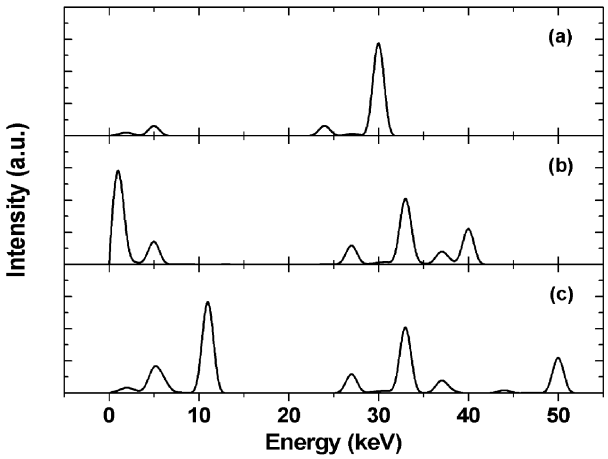

Fig. 3. Primary electron spectra for γ-ray energies E_γ of (a) 30 keV; (b) 40 keV; and (c) 50 keV.

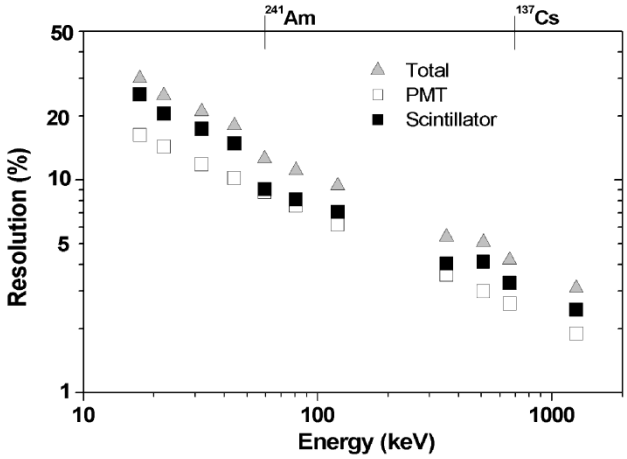

Fig. 4. Energy resolution as function of photon energy of $LaCl_3$:10% Ce^{3+}.

the scintillator resolution of the crystal. In principle, the energy resolution, $\Delta E/E$, of a scintillator detector can be written as

$$(\Delta E/E)^2 = R_\delta^2 + R_{\rm np}^2 + R_{\rm inh}^2 + R_p^2 + R_M^2 \qquad (2)$$

where R_δ is the contribution of δ-rays to the energy resolution, $R_{\rm np}$ the contribution of the nonproportional response of the scintillator, $R_{\rm inh}$ is connected with inhomogeneities in the crystal, which can cause local fluctuations in the scintillation light output, R_p the transfer resolution and R_M the contribution of the PMT to the resolution. The combined effects of $R_\delta, R_{\rm np}, R_{\rm inh}$, and R_p are usually referred to as scintillator resolution R_S and is defined by

$$R_S^2 = R_\delta^2 + R_{\rm np}^2 + R_{\rm inh}^2 + R_p^2. \qquad (3)$$

For an ideal scintillator, the four $R_\delta, R_{\rm np}, R_{\rm inh}$, and R_p resolutions vanish and $\Delta E/E = R_M$. With the R1791 PMT, an energy resolution R of $4.2 \pm 0.5\%$ (FWHM over peak position) for the 662 keV full absorption peak has been observed.

In an earlier paper [20], we reported an energy resolution of $3.3 \pm 0.3\%$ for a $LaCl_3$:10% Ce^{3+} crystal of size $\oslash 8$ mm $\times$ 5 mm. This energy resolution is significantly better than the energy resolution reported in this paper. Interestingly, both crystals exhibit a linear relationship between resolution and the inverse square root of the energy. The step like curves of NaI(Tl) with a semi plateau in the energy range between 100 and 500 keV [21] have not been observed for $LaCl_3$:Ce^{3+}. Since the photoelectron yields are relatively large for the $LaCl_3$:Ce^{3+} crystals, the contribution of R_M to the total resolution is small, especially at energies above 200 keV. Consequently, the observed energy resolution is mainly due to the intrinsic resolution.

What is the origin of this intrinsic resolution? Above 200 keV, the curve of NaI(Tl) is characterized by a bell-shaped contour due to the contribution of multiple Compton scattering to the full energy peak. For $LaCl_3$:Ce^{3+} we did not observe such a contribution, which is in agreement with the *proportional* photon response of $LaCl_3$:10% Ce^{3+} between 40 keV and 1.274 MeV. Consequently, the contribution of multiple Compton scattering and the contribution of the nonproportional response of $LaCl_3$:10% Ce^{3+}, $R_{\rm np}$, to the intrinsic resolution are probably negligible. Nevertheless, the scattering of electrons produced by γ-rays, commonly referred to as δ-rays, may result in an additional spreading of the energy resolution. According to Iredale [22], δ-rays of energy >1 keV add approximately 3.2% to the energy resolution of NaI(Tl) for 0.66 MeV electrons. For $LaCl_3$:10% Ce^{3+}, we found that on average, 10% of the total energy deposited by electrons is dissipated by δ-rays with mean energy 1 keV. Since the contribution of δ-rays to the total energy resolution may vary with electron energy [22], we expect that R_δ significantly contributes to the intrinsic resolution.

A larger R_S for $E_\gamma < 662$ keV may also be due to an increase in $R_{\rm inh}$. For a 662-keV photon, the absorption length in $LaCl_3$:10% Ce^{3+} is 3.4 cm. For this energy, the γ-rays are uniformly absorbed in the crystal. For lower energies, the absorption length is considerably shorter: at 60 keV, 0.49 mm, and at 17.4 keV, 0.084 mm. These low energy photons are only absorbed in the surface layer of the crystal. If the scintillation or the light collection properties at the surface are somewhat different from the bulk, it will result in a larger dispersion of the energy resolution and $R_{\rm inh}$ increases.

Finally, variations in R_p are expected to make only a small contribution to the observed energy resolution. In this paper it has been assumed that it is not dependent on energy.

V. Conclusion

A proportional response to electron and γ-rays is observed for a canned $LaCl_3$:10% Ce^{3+} crystal in the energy region between 7 and 440 keV and 40 keV and 1.274 MeV, respectively. The electron response determines to a large extent the shape of the photon response. Of crucial importance is the shape of the electron response at $E_e < 10$ keV: the number of Auger electrons produced in that region determines the magnitude of the relative photon light yield just above the K edge (of lanthanum). The energy resolution of this crystal is worse than expected, due to surface effects and δ-ray energy fluctuations. A better encasement of the crystal might improve the energy resolution at $E_\gamma < 100$ keV.

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