Study on Fast Luminescence Component Induced by Gamma-rays in Ce Doped LiCaAlF₆ Scintillators

Kenichi WATANABE^a, Yoshiyuki KONDO^a, Atsushi YAMAZAKI^a, Akira URITANI^a, Tetsuo IGUCHI^a,
Noriaki KAWAGUCHI^b, Kentaro FUKUDA^b, Sumito ISHIZU^b, Takayuki YANAGIDA^c,
Yutaka FUJIMOTO^c, Akira YOSHIKAWA^d

^aNagoya University, Furo-cho, Chikusa-ku, Nagoya, 464-8603, Japan

^bTOKUYAMA Corp, 1-1, Mikage-cho, Shunan, Yamaguchi, 745-8648, Japn.

^cKyusyu Institute of Technology, 2-4 Hibikino, Wakamatsu-ku, Kitakyushu-shi, Fukuoka, 808-0196, Japan.

^dTohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, 980-8577, Japan

Corresponding author: Kenichi Watanabe, Nagoya University

Tel: +81-52-789-4513, Fax.: +81-52-789-3843

E-mail: k-watanabe@nucl.nagoya-u.ac.jp

Abstract

We discuss the origin of the fast luminescence component induced by fast electrons generated in gamma-ray interactions in Ce doped LiCaAlF₆ scintillators. Although the slow luminescence component induced by Ce³⁺ emissions depends on the Ce concentration in the LiCaAlF₆ scintillator, the fast component is independent. The fast component is suggested to be generated in the host matrix of the LiCaAlF₆ crystal. From quantitative considerations based on Frank-Tamm equation, which shows the light yield of the Cherenkov radiation, the Cherenkov radiation was determined as the origin of the fast component. We, additionally, found that the slow rise time of main Ce³⁺ emissions in the Ce:LiCaAlF₆ scintillator plays an important role to perform the pulse shape discrimination.

KEYWORDS: He-3 alternatives, neutron scintillator, pulse shape discrimination, Cherenkov radiation

1. Introduction

Neutron detection techniques play an important role in various fields. For thermal neutron detection, a He-3 proportional counter has been the gold standard detector. Within the last few years, the He-3 gas shortage has been a severe problem for neutron detection applications. Therefore, alternatives to the conventional He-3 neutron detectors have been required. Inorganic scintillators containing lithium or boron have attracted much attention for thermal neutron detection. As one of the promising candidates, Ce doped LiCaAlF_6 scintillators have been developed (Yoshikawa et al., 2009, Kawaguchi et al., 2011 and Iwanowska et al., 2011). The Ce: LiCaAlF_6 scintillator has excellent properties for neutron detection such as significant light yield, fast response, high transparency, light composition and no hygroscopicity. Especially, since Ce: LiCaAlF_6 scintillators have a high alpha to beta ratio, which is defined as the ratio of light outputs per unit energy for alpha particles and electrons, they can be promising candidates from viewpoint of the gamma-ray rejection. In addition, Ce: LiCaAlF_6 scintillators were reported to distinguish neutron and gamma-ray events by the pulse shape discrimination technique (Yamazaki et al., 2011). For only gamma-ray events, a scintillation pulse shape has a quite fast component with the decay time less than a few nanoseconds. On the other hand, for neutron events, it has only a relatively slow component with the decay time of a few ten nanoseconds, corresponding to the emission of Ce^{3+} ions. This fast component can be applied to discriminate neutron and gamma-ray events. Although the origin of this fast component was discussed from the viewpoint of optical physics, the origin has not been identified (Koshimizu et al., 2013). In this paper, we discuss the fast luminescence component induced by gamma-rays in Ce: LiCaAlF_6 scintillators through the experiments using the digital signal processing technique, which can quantitatively separate a scintillation signal pulse into the fast and slow luminescence components.

2. Experimental Procedures

In order to perform the pulse shape discrimination, we apply the digital signal processing technique. In the basic experiments, the following experimental setup was used to acquire signal data. The

Ce:LiCaAlF₆ scintillator was optically connected to a photomultiplier tube (PMT, Hamamatsu, R7600U-200). The output signal from the PMT was directly digitized with the fast digitizer (Agilent, U1071A, 8bit, 1GHz, 2GS/s). The digitized signal pulse shapes were processed off-line with PC.

Figure 1 shows typical signal pulse shapes induced by a neutron and gamma-ray in Ce:LiCaAlF₆ scintillator. The Ce:LiCaAlF₆ scintillator has a quite fast component about a few nanoseconds only when excited by fast electrons produced by gamma-rays. In order to separate quantitatively the scintillation signal into the fast and slow components, the digitized data was integrated in two regions. The first region is defined as the period of 4 nanoseconds from signal trigger timing and shows the fast luminescence component. The second region covers the whole signal pulse and shows the slow luminescence component.

Figure 2 shows the two dimensional histogram of the fast and slow components obtained from a Ce:LiCaAlF₆ scintillator when irradiated with a moderated Cf neutron source. The horizontal axis is also shown in the unit of the electron equivalent energy. The value on this axis is calibrated with the Compton edge obtained by using a Cs-137 mono-energy gamma-ray source. In this figure, we can find two groups, corresponding to gamma-ray and neutron events. An island group with low fast component and constant slow luminescence corresponds to the neutron event group. On the other hand, the band group with the slope corresponds to the gamma-ray event group. This group has larger fast component than the neutron one. In this group, the fast component has positive correlation with the slow luminescence. The fast luminescence component increases with the deposition energy into the scintillator in gamma-ray induced events. We can distinguish neutron and gamma-ray events in this two dimensional histogram. By using these procedures, we can determine quantitatively the contributions of fast and slow luminescence component.

3. Results and Discussion

3.1 Ce Content Dependence

In order to discuss the origin of the fast luminescence, the Ce doping concentration dependence of

the luminescence property was measured. LiCaAlF_6 scintillators with Ce concentrations of 2%, 3% and 4% in raw material composition were prepared. We irradiated Co-60 gamma rays into the specimens to measure the relationship between the fast and slow components. For each LiCaAlF_6 scintillator sample, the digital signal processing was applied and the fast and slow components were extracted separately. **Figure 3** shows the pulse height spectrum of the slow component obtained from Ce:LiCaAlF_6 scintillators with various Ce concentrations. These spectra are created by accumulating the two dimensional histogram shown in Fig. 2 in the horizontal direction and correspond to the normal pulse height spectra. In these spectra, only the Compton continuum can be shown but no photoelectric peak can be found, because Ce:LiCaAlF_6 scintillators have no heavy element and they have quite small photoelectric absorption probability. Since the light yield of Ce:LiCaAlF_6 scintillators is not so high, the energy resolution in the pulse height spectra is relatively low. Consequently, two Compton edges corresponding to 1.17 and 1.33 MeV gamma-ray from a Co-60 source cannot be observed clearly. However, we can confirm that the pulse height of the Compton edges increases with the Ce concentration. This is because the slow luminescence component corresponds to emissions in the Ce^{3+} site. **Figure 4** shows the pulse height spectra of the fast component obtained from Ce:LiCaAlF_6 scintillators with various Ce concentrations. These spectra are created by accumulating Fig. 2 in the vertical direction. In contrast to the case of the slow component, these spectra have no dependence on the Ce concentration. This indicates that the fast luminescence component is independent of the Ce^{3+} site and is generated in the host matrix of the LiCaAlF_6 crystal.

3.2 Cherenkov Radiation

One of the candidates for the origin of the host matrix emission is Cherenkov radiation. Cherenkov radiations are emitted when a charged particle passes through a medium with the speed greater than the velocity of light in that medium. Therefore, electrons with the energy more than the threshold, which is determined by the refractive index of the medium, can emit this type of radiation in all materials. Since the Ce:LiCaAlF_6 scintillator has the refractive index of 1.4, the threshold electron energy is 220 keV. If the fast

component is based on the Cherenkov radiation, it cannot be found below this energy. In Fig. 2, the fast component for low energy deposition events has near zero values. In order to discuss more quantitatively, the light yield of Cherenkov radiation is calculated with the Frank-Tamm equation as given by

$$\frac{d^2 N_p}{dL d\lambda} = \frac{2\pi\alpha Z^2}{\lambda} \left(1 - \frac{1}{n^2 \beta^2} \right) \quad (1),$$

where N_p is the number of emitted photons; fine structure constant, α ; charge of a particle, Z ; refractive index, n ; relative velocity (v/c), β ; wavelength, λ and L is the path length of a particle (Tamm, 1939). The calculated light yield is fitted to the result in Fig. 2. **Figure 5** shows the fitted curve for two dimensional histogram of the fast and slow components. A dashed line shows only Ce^{3+} emission yield without Cherenkov radiations. A solid line shows the total yield of Ce^{3+} emission and Cherenkov radiations. The experimental results can be well explained by the Cherenkov radiation yield calculated with the Frank-Tamm equation. Events in a circle in Fig. 5 are considered to be events induced by electrons passing through both of the scintillator and the window glass of the PMT. These events have higher fast component than events induced by electrons passing through only the scintillator, corresponding to events on the solid line in Fig. 5. This increment is due to the Cherenkov radiation caused in the window glass of the PMT. We, additionally, confirmed that the fraction of this component decreased in the case of a larger Ce:LiCaAlF_6 scintillator. From these results, we conclude that the fast component induced in gamma-ray events is based on the Cherenkov radiation by energetic electrons generated in gamma-ray interactions.

3.3 Rise Time Property

Since the Cherenkov radiation can be emitted from every media, every Li containing scintillators can perform the pulse shape discrimination using the fast component. However, a Li glass scintillator, which is one of the conventional neutron scintillators, cannot effectively perform the pulse shape discrimination between neutron and gamma-ray events. **Figure 6** shows typical pulse shapes induced by a neutron obtained from a Li glass and a Ce:LiCaAlF_6 scintillator. As you can see, the Ce:LiCaAlF_6 scintillator has

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4 relatively slow rise time in the Ce^{3+} emission. Since the Li glass scintillator has quite fast rise time in main
5 emission component, the Cherenkov radiations can be obscured by the fast rise time component. The slow
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7 rise time in the main emission component of the $\text{Ce}:\text{LiCaAlF}_6$ scintillator plays an important role in order to
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9 perform the pulse height discrimination because the light yield of the Cherenkov radiation is not so high
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11 compared with the main Ce^{3+} emission as shown in Fig. 1.
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16 Finally, we consider the mechanism of the slow rise time in the $\text{Ce}:\text{LiCaAlF}_6$ scintillator. The
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18 dependence of the rise time on the Ce concentration is shown in **Fig. 7**. The rise time decreases with the Ce
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20 concentration. We can, additionally, confirm that the rise time is proportional to one-third power of the Ce
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22 concentration. This means that the rise time is proportional to the interatomic distance of Ce^{3+} ions. This
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24 suggests that excited electrons by an ionizing radiation are trapped at trapping sites before they reach to the
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26 Ce^{3+} site and de-excite with scintillation photon emissions. Further studies, such as investigation of these
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28 trapping site, are required to determine the mechanism of the slow rise time.
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35 4. Conclusion

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57 process of radiation excited electrons.
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Acknowledgement

This study is the result of “Development of an alternative to He-3 neutron detectors for homeland security and nuclear material safeguards” carried out under the Adaptable and Seamless Technology transfer Program through targetdrive R&D by Japan Science and Technology Agency.

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Figure Captions

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Figure 2 Two dimensional histogram of the fast and slow components in Ce:LiCaAlF₆ scintillation emissions. The scintillator was irradiated with a moderated Cf neutron source.

Figure 3 Pulse height spectra of the slow component obtained from Ce:LiCaAlF₆ scintillators with various Ce contents when irradiating Co-60 gamma rays.

Figure 4 Pulse height spectra of the fast component obtained from Ce:LiCaAlF₆ scintillators with various Ce contents when irradiating Co-60 gamma rays.

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Figure 6 Typical pulse shapes induced by a neutron obtained from a) Li glass and b) Ce:LiCaAlF₆ scintillator.

Figure 7 Ce content dependence of the rise time in Ce:LiCaAlF₆ scintillation emission induced by neutron absorptions. The solid line shows the fitted line to a function proportional to one third power of the Ce concentration.

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