Available online at www.sciencedirect.com
ScienceDirect
Radiation Measurements 42 (2007) 1351 – 1354
Radiation Measurements
www.elsevier.com/locate/radmeas

The role of CeF_3 in $LaCl_3$ scintillation crystal

Yu Pei, Xiaofeng Chen, Dongmin Yao, Guohao Ren*

Shanghai Institute of Ceramics, Chinese Academy of Sciences, No. 215, Chengbei Road, Jiading District, Shanghai 201800, China

Received 6 June 2006; received in revised form 25 December 2006; accepted 5 June 2007

Abstract

The properties of $LaCl_3$:CeF_3 crystal are investigated and its hydroscopic nature has a much more decrease than that of $LaCl_3$:$CeCl_3$ crystal. The scintillation properties of $LaCl_3$:CeF_3 maintain as excellent as that of $LaCl_3$:$CeCl_3$. The $LaCl_3$:CeF_3 crystal has a good optical transmittance without obvious optical absorption bands between 322 and 800 nm, and has the same absorption edge as the $LaCl_3$:$CeCl_3$ crystal (about 315 nm). Its emission spectra, decay time and light yield are the same as the $LaCl_3$:$CeCl_3$.

PACS: 78.55.Hx; 78.70.En

Keywords: $LaCl_3$; Scintillator; Hydroscopy

1. Introduction

Within the last few years, the scintillation properties and optical properties of $LaCl_3$:$CeCl_3$ were reported by a number of investigators. It is a very promising scintillation crystal in medical image and nuclear physics because of its excellent energy resolution, high light yield and fast decay time (Krämer et al., 1998; Guillot-Noël et al., 1999; van Loef et al., 2000, 2001; Shah et al., 2003; Allier et al., 2002; van Eijk et al., 2001). However, it also has some disadvantages, especially hygroscopy, which seriously discourages its application. In this paper, we report our initial investigation about depressing its hygroscopy by doping CeF_3 as an activator. It is discovered that hygroscopy of $LaCl_3$:CeF_3 crystal has a much more decrease than that of $LaCl_3$:$CeCl_3$ crystal. Meanwhile, its scintillation properties are almost as excellent as $LaCl_3$:$CeCl_3$.

2. Experiments

The crystal is grown by Modified Bridgman Method. The detailed process about the growth can be found in Pei et al. (2005). Some small pieces from the ingot of the $LaCl_3$:CeF_3 are used to observe their hydroscopic behavior, shown in Fig. 1. $LaCl_3$:CeF_3 crystals are characterized by photoluminescence (PL), X-ray excited luminescence (XEL), γ-ray pulse height spectrum, and decay time. During the measurement, the crystals are sealed in a dry container with an optical quartz window.

3. Results and discussions

3.1. The decrease of the hydration ability

The processes of hydration are shown in Fig. 1. When $LaCl_3$:CeF_3 and $LaCl_3$:$CeCl_3$ crystals are placed in the same humid environment at the same time (humidity of 70% and temperature at 295 K). It can be observed that the surface of $LaCl_3$:$CeCl_3$ crystal becomes moist at once, and its surface is surrounded by clear H_2O solution after 2 h, and then it completely turns into solution after 5 h. Whereas, the surface of $LaCl_3$:CeF_3 crystal just becomes moist after 5 h. Although this observation is a little superficial, it is still able to reflect its nature. However, when the CeF_3 is doped into $LaCl_3$ as an activator, the hydration ability of the crystal decreased significantly compared with that of $LaCl_3$:$CeCl_3$. This phenomenon can be explained as follows. The F^- ions have much larger electronegative and less ion radius than that of Cl^- ions, so the

* Corresponding author. Tel.: +86 21 69987740.
E-mail address: rgh@mail.sic.ac.cn (G. Ren).

1350-4487/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.radmeas.2007.06.011

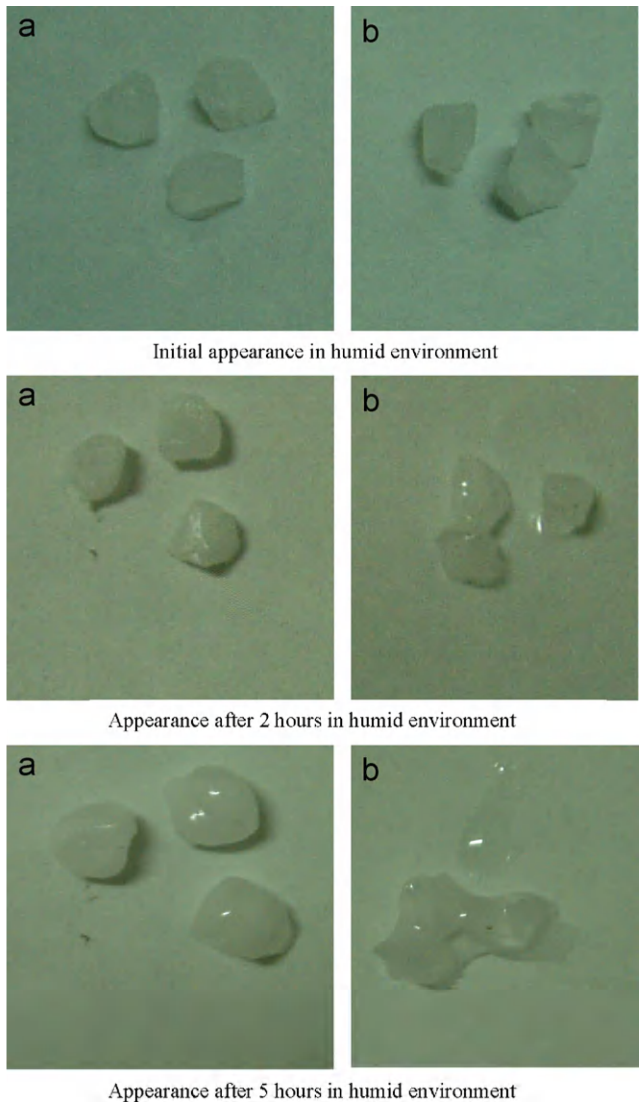

Fig. 1. The comparison of the hydration ability of the $LaCl_3$:0.5 at.%CeF_3 (a) and $LaCl_3$:0.5 at.%$CeCl_3$ (b) (Humidity of 70%, temperature at 295 K).

chemical bond between F^- and La^{3+} will be stronger than that between Cl^- and La^{3+}. Therefore, the chemical bond between F^- and La^{3+} is not easy to be destroyed by H_2O molecule. As a consequence, the hydration ability of $LaCl_3$ crystal largely decreased.

3.2. Transmittance

Transmission spectra are recorded by Shimadzu UV-2501 spectrophotometer. Fig. 2 shows the optical transmission spectra of $LaCl_3$:CeF_3 and $LaCl_3$:$CeCl_3$ crystals with the thickness of 4 mm. $LaCl_3$:CeF_3 crystals have almost the same absorption edge as $LaCl_3$:$CeCl_3$ (about 315 nm). No obvious absorption bands are found in the transmission spectrum between 320 and 800 nm, which indicates that dopant of CeF_3 do not induce any color center in $LaCl_3$ crystal.

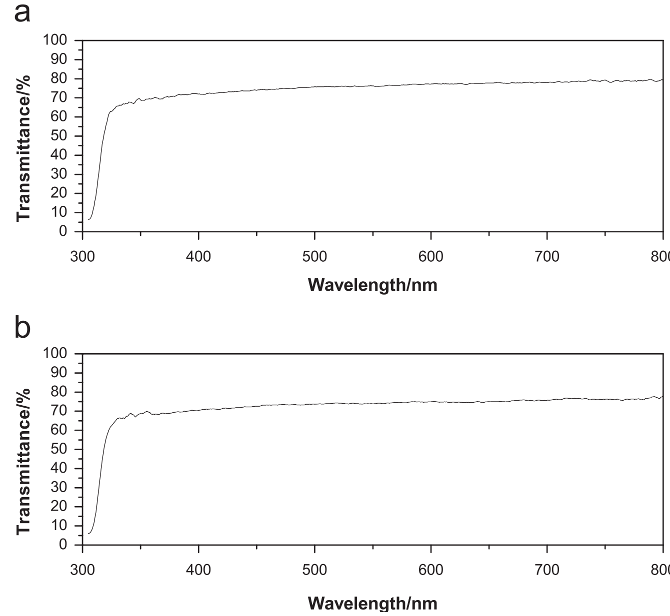

Fig. 2. Optical transmittance as a function of wavelength for $LaCl_3$: 0.5 at.%CeF_3 (a) and $LaCl_3$:0.5 at.%$CeCl_3$ (b) crystal (4 mm).

3.3. Photoluminescence and X-ray excited luminescence

Photoluminescence (PL) of $LaCl_3$:0.5 at.%CeF_3 and $LaCl_3$: 0.5 at.%$CeCl_3$ are measured by FLS920 fluorescence spectrophotometer at room temperature (20 °C) as shown in Fig. 3. The 0.2 nm slit of emission and excitation is chosen in the measurement. The spectra are normalized. It can be seen in Fig. 3(a), the maximum emission peak of $LaCl_3$:CeF_3 locates at 332 nm and maximum excitation peak at 306 nm. The whole shape and peak of the spectrum are almost same as the $LaCl_3$:$CeCl_3$, just as shown in Fig. 3(b). The band of self-trapped excitons (STE) luminescence at about 405 nm is not present under optical excitation, but under X-ray excitation, this band can be clearly seen, as shown in Fig. 4(a).

X-ray excited luminescence (XEL) is measured by FLS920 fluorescence spectrophotometer at room temperature (20 °C). The Fig. 4 illustrates the shape and peak position of XEL of $LaCl_3$:CeF_3 and $LaCl_3$:$CeCl_3$. Each curve is normalized.

Each curve could be fitted with three Gauss-shaped bands—peak1, peak2 and peak3. Black lines are the measurement curves, and red and green lines are the Gauss fitted curves. Peak1 and peak2 (at about 332 and 352 nm, respectively) are attributed to the two emission of Ce^{3+} from 5d to the two sub-level ground states of 4f ($^2F_{5/2}$ and $^2F_{7/2}$), and peak3 (at about 405 nm) attributed to STEs emission (Guillot-Noël et al., 1999). According to Fig. 4(a) and (b), it can be seen that $LaCl_3$:CeF_3 have the same spectral shape and peaks as the $LaCl_3$:$CeCl_3$.

3.4. γ-ray pulse height spectrum

The pulse height spectrum of $LaCl_3$:0.5 at.%CeF_3 crystal with the dimension of $\Phi 8\,mm \times 4\,mm$ for 662 keV gamma

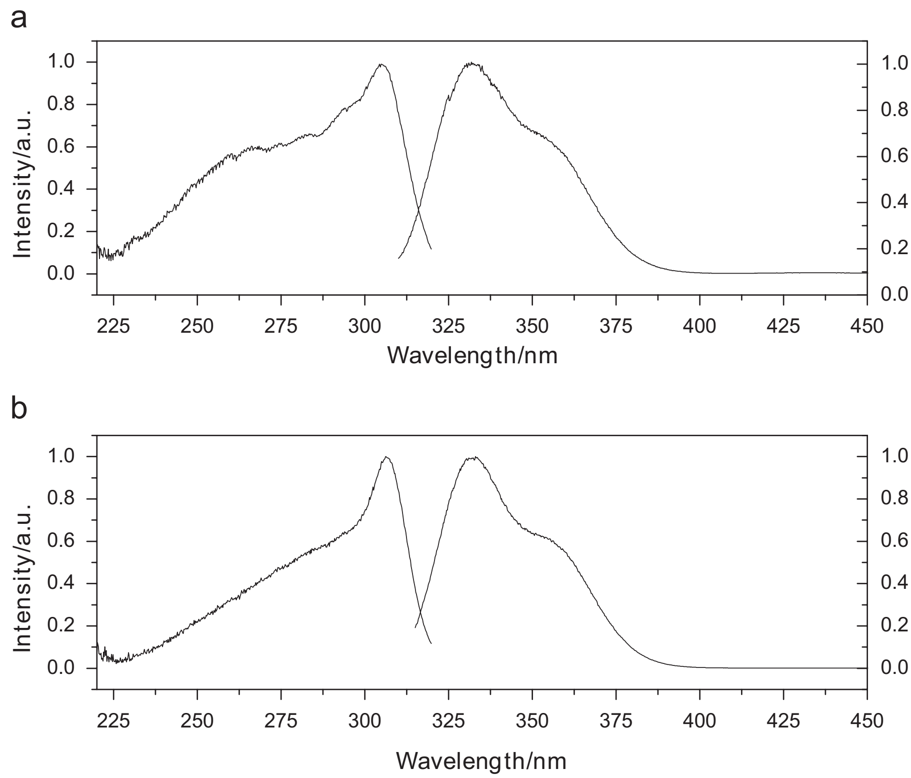

Fig. 3. Emission and excitation spectra of $LaCl_3$:0.5 at.%CeF_3 (a) and $LaCl_3$:0.5 at.%$CeCl_3$ crystals.

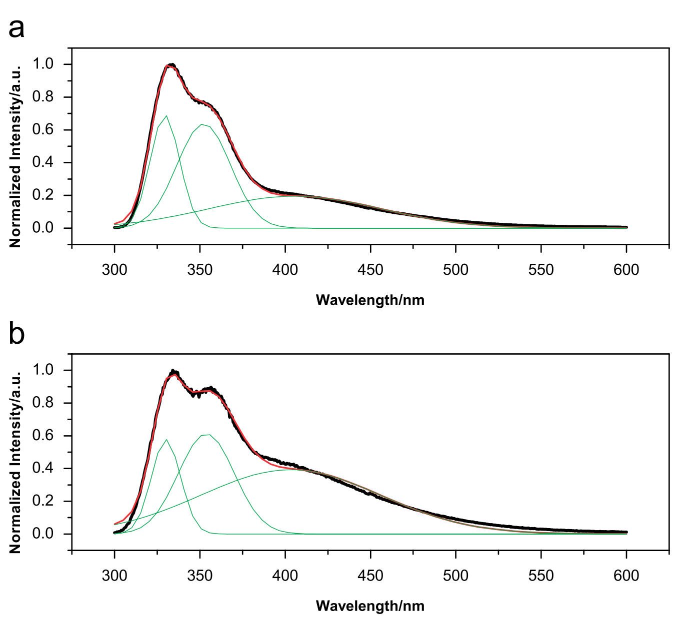

Fig. 4. X-ray excited luminescence of $LaCl_3$:0.5 at.%CeF_3 (a), $LaCl_3$:0.5 at.%$CeCl_3$ (b).

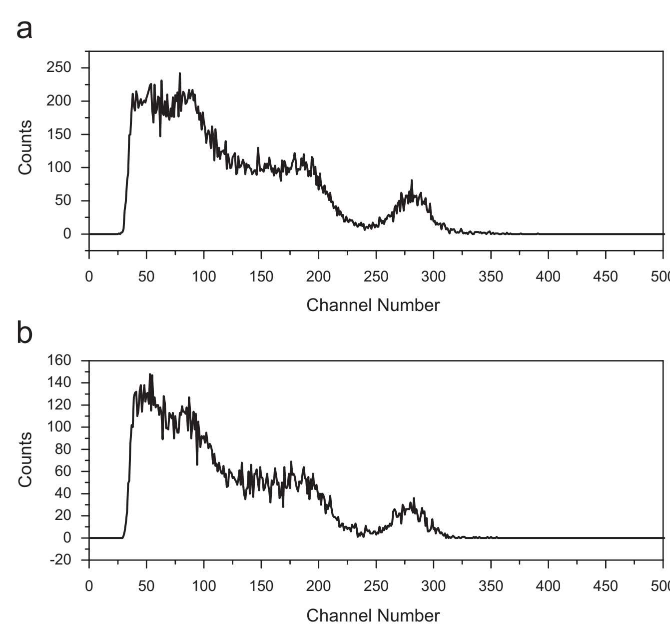

Fig. 5. Pulse height spectra of ^{137}Cs γ-quanta of $LaCl_3$:0.5 at.%CeF_3 (a) and $LaCl_3$:0.5 at.%$CeCl_3$ (b) crystals coupled to a Hamamatsu R2059 PMT.

ray from ^{137}Cs is shown in Fig. 5, obtained with a Hamamatsu R2059 photomultiplier tube (PMT). Meanwhile, the pulse height spectrum of $LaCl_3$:0.5 at.%$CeCl_3$ crystal with the same dimension is also shown in Fig. 5. It can be seen that the channel number of the $LaCl_3$:0.5 at.%CeF_3 crystal is almost the same as that of the $LaCl_3$:0.5 at.%$CeCl_3$. However, it should be able to be further enhanced by the improvement of the crystal qualities and the Ce^{3+} concentration.

3.5. Photoluminescence decay

The photoluminescence decay curves are measured by the single photon counting method with FLS920 fluorescence

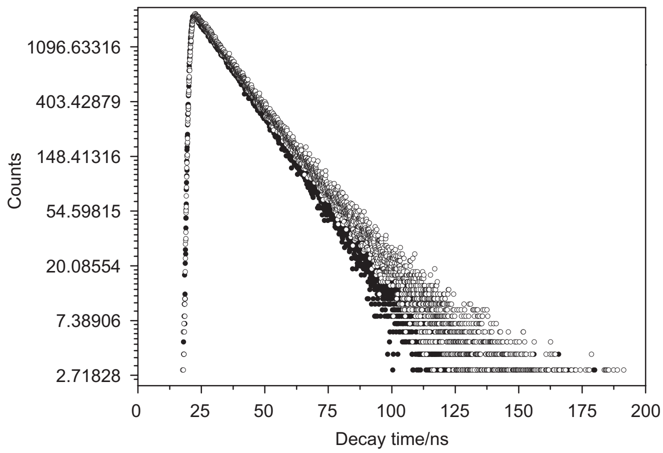

Fig. 6. The photoluminescence decay time curves of $LaCl_3$:0.5 at.%CeF_3 and $LaCl_3$:0.5 at.%$CeCl_3$.

spectrophotometer. Fluorescence decay times are obtained from the decay curves using common deconvolution procedure. The decay time at different wavelength can be measured by placing monochromator between the crystal and the photomultiplier.

The results of the $LaCl_3$:0.5 at.%CeF_3 and $LaCl_3$:0.5 at. %$CeCl_3$ crystals are shown in Fig. 6. The emission wavelength is fixed at 332 nm, excitation wavelength at 306 nm. As can be seen in the scintillation decay curves, both $LaCl_3$:CeF_3 and $LaCl_3$:$CeCl_3$ obviously have only one single exponential decay constant, and the decay time is almost same (about 15 ns), which is attributed to direct excitation of Ce^{3+} ion.

4. Conclusions

CeF_3 is a good activator in the preparation of $LaCl_3$ scintillation crystals. The introduction of the F^- ions into $LaCl_3$ crystal, not only can reduce its hygroscopy, which is very beneficial for the practical application in medical image and nuclear physics. Meanwhile, the introduction of the CeF_3 does not cause the deterioration of the scintillation properties compared with $LaCl_3$:$CeCl_3$ crystal. The crystal has a good optical transmittance between 323 and 800 nm without any obvious optical absorption band. Both $LaCl_3$:CeF_3 and $LaCl_3$:$CeCl_3$ crystal have the same Ce^{3+} absorption edge (315 nm) and the similar spectra structure. The $LaCl_3$:CeF_3 crystal has the same emission spectra as the $LaCl_3$:$CeCl_3$ crystal. In addition, the decay time and light yield of $LaCl_3$:CeF_3 sample also almost keep the same as $LaCl_3$:$CeCl_3$ (about 15 ns). However, the quality and dimension of the crystal need further improvement, and its scintillation properties need further study.

References

Allier, C.P., et al., 2002. Nucl. Instrum. Methods A 485, 547.
Guillot-Noël, O., et al., 1999. J. Luminescence 85, 21.
Krämer, K.W., Gudel, H.U., Schwartz, R.N., 1998. J. Alloys Compd. 275–277, 191.
Pei, Y., et al., 2005. J. Crystal Growth 279, 390.
Shah, K.S., et al., 2003. Nucl. Instrum. Methods A 505 (10), 76.
van Eijk, C.W.E., et al., 2001. Nucl. Instrum. Methods A 471, 244.
van Loef, E.V.D., et al., 2000. Appl. Phys. Lett. 77 (10), 1467.
van Loef, E.V.D., et al., 2001. IEEE Trans. Nucl. Sci. 48 (3), 341.