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Directionally solidification of Ce:$LaCl_3$/$AECl_2$ (AE=Mg, Ca, Sr, Ba) eutectic scintillators

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Abstract

In this study, Ce doped $LaCl_3$/$AECl_2$ (AE=Mg,Ca,Sr) system eutectics were explored. Ce doped $LaCl_3$/ $AECl_2$ (AE=Mg,Ca,Sr) eutectics were grown by the BZ method in the quartz ampoule. In the case of AE=Mg,Ca, eutectic with PSSFs structures were observed. In the case of AE = Sr sample, solid state of $La_{0.708}Sr_{0.292}Cl_{2.708}$ was observed. The eutectics showed optical transparency like bundle optical fibers. The grown eutectic structure showed mixture of rods and plates shape. This mixed structure was aligned with length of around 300-400 □m. The Ce:$LaCl_3$/$CaCl_2$ eutectic shows 345 nm emission ascribed to Ce^{3+} 4f-5d transition under X-ray excitation.

1. Introduction

Scintillators coupled with photodetectors were widely used in radiation imaging applications such medical imaging, security, high energy physics, astrophysics, oil well logging, etc. In the X-ray imaging applications, radiation imaging sensors were composed of photodetector arrays and indirect flat panel detector (FPD) coupled with a scintillator plate such as Tl:CsI. Improvement in the spatial resolution is required in this application. However, CsI:Tl columnar grown Tl:CsI reduce the spatial resolution, because light scattering in the micro-meter size Tl:CsI fiber crystals. Light diffusion through scintillatior materials on photodetector degrade resolution of radiation imaging sensors and limit the sensitivity. Recently, pixilated photodetector arrays has been improved to achieve a micrometer scale special resolution. However pixel size of scintillator arrays and the light diffusion limit the spatial resolution.

Corrently, submicron-diameter phase-separated scintillator fibers (PSSFs) were reported and they have both characteristics of optical fiber and a radiation-to-light conversion. The PSSFs were realized by a directionally solidified eutectic (DSE) growth in previous research [1]. In PSSFs, the emitted scintillation is confined and transported along the eutectic structure by a total reflection mode, so that light diffusion can be reduced and high-resolution imaging can be achieved(Fig.1). Up to now, research on PSSFs such GAP/α-Al_2O_3[2], $SrHfO_3$/Al_2O_3 [3], $Gd_2Si_2O_7$/SiO_2 and LiF/ CaF_2/$LiBaF_3$ have been already reported by our group.

In order to find good combination of eutectic structure with PSSFs, choice of scintillator materials is important. Here, Ce:$LaCl_3$ scintillator has attracted attention due to its high light yield of above 50,000 photons/MeV and fast decay time of 28 ns with enough density of 3.85 g/cm^3 for low energy X-ray detection even it is hygroscopic[4]. In this research, exploration of PSSFs by directional crystal growth method will be reported. In this study, Ce doped $LaCl_3$/$AECl_2$ (AE=Mg,Ca,Sr) system eutectics were explored.

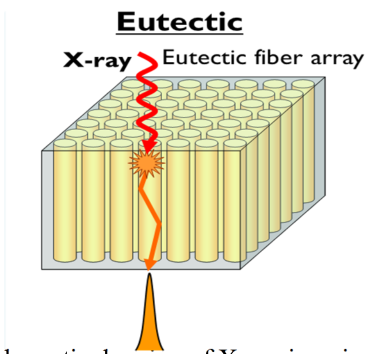

Fig.1 Schematic drawing of X-ray imaging by PSSFs

2. Results

Starting materials were prepared from high-purity fluoride powders of $LaCl_3$, $CeCl_3$, $MgCl_2$, $CaCl_2$ and $SrCl_2$ (4N, produced by APL) according to the molar ratio of $AECl_2$: $LaCl_3$:$CeCl_3$=72 : 27.72 : 0.28 (AE=Mg,Ca,Sr). The ratio was based on the eutectic point of $LaCl_3$/$CaCl_2$ system. $LaCl_3$/$AECl_2$ eutectics were grown at the composition of eutectic point. Crystal growth was performed by the BS method in a quartz ample with 6 mm inner diameter. In the unidirectional solidification processes, $LaCl_3$ and $AECl_2$ phases deposit from the melt on their own formed solid phases.

White rod with 6 mm diameter and 25 mm length was obtained by the BS method. An example of the grown eutectic and 1 mm thick plates after polishing were shown in Fig. 2. The samples showed optical transparency like bundle optical fibers and the background line is visible on the surface through the transparent rods grown in the material in the case of $LaCl_3$/$AECl_2$ (AE=Mg, Ca). The grown eutectic samples were crushed into powder with oil and XRD measurement was performed. In the case of $LaCl_3$/$MgCl_2$ and

$LaCl_3/CaCl_2$, the grown eutectics showed two main phases of hexagonal $LaCl_3$ with rhombohedral $MgCl_2$ and orthorhombic $CaCl_2$. In the case of $LaCl_3/SrCl_2$ sample, Solid solution of cubic $(La_{1-x},Sr_x)Cl_{3-x}$.

The BEI of the grown crystals in vertical and transverse cross-section are shown in Fig. 3. White $LaCl_3$ rods phase was surrounded by black $MgCl_2$ matrix in the case of the Ce:$LaCl_3/MgCl_2$ sample. Rods and plates shape of white $LaCl_3$ phase was surrounded by black $CaCl_2$ matrix in the case of the Ce:$LaCl_3/CaCl_2$ sample. These structures of $LaCl_3$ were aligned with length of around 300-400 □m. In the case of the Ce:$LaCl_3/SrCl_2$, gray phase identified to $La_{0.708}Sr_{0.292}Cl_{2.708}$ according to an EDX results and white phase was $LaCl_3$

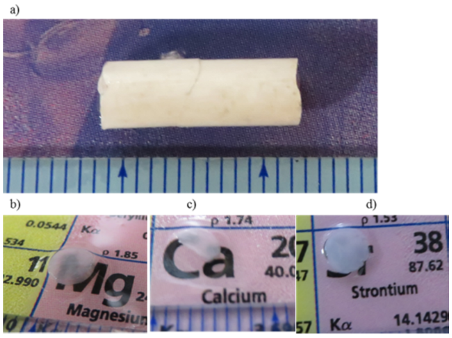

Fig. 2. Photograph of the a) as grown Ce:$LaCl_3/CaCl_2$ eutectic and b) polished Ce:$LaCl_3/MgCl_2$, c) Ce:$LaCl_3/CaCl_2$ and Ce:$LaCl_3/SrCl_2$

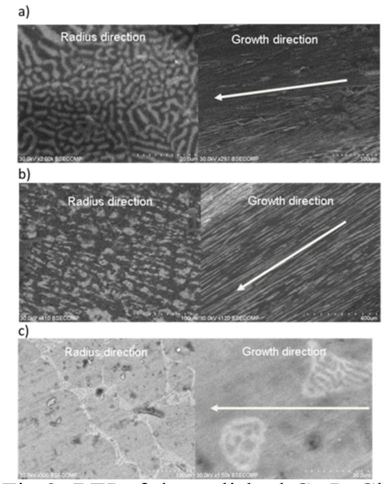

Fig.3. BEI of the polished Ce:$LaCl_3/MgCl_2$, Ce:$LaCl_3/CaCl_2$ and Ce:$LaCl_3/SrCl_2$ samples on the transverse cross-section and vertical cross-section

Radioluminescence spectra of the grown Ce doped $LaCl_3/AECl_2$ (AE=Mg,Ca,Sr) samples measured under X-ray irradiation were shown in Fig. 6. The expected Ce^{3+} 4f5d emission in the rage of 300-450 nm peaking at around 350nm have been observed. These results are good agreement of previous reports on Ce:$LaCl_3$[4]. Emission peak of the AE = Ca sample was shifted to shorter wavelength. It is guess that Ca^{2+} substitution in Ce:$LaCl_3$ causes this peak shift.

The Pulse height spectra of the grown Ce doped $LaCl_3/AECl_2$ (AE=Mg,Ca,Sr) samples excited by 662 keV gamma-ray of ^{137}Cs at room temperature and measured using the PMT are shown in Fig. 7. Light outputs of the AE = Mg and Ca sample were 1,400 around 9,000 photon/MeV comparing with a LYSO standard with light outputs of 27,000 photon/MeV. Energy resolution of the AE = Ca was 17%@662keV. The scintillation decay curves of the AE=Mg,Ca and Sr samples excited by 662 keV gamma-ray are shown in Fig. 8. Scintillation decay time of the AE=Mg,Ca and Sr samples were 20.2ns (28%) 467 ns (72%) , 26.8 ns (26%) 282 ns (74%) and 42.5 ns (32%) 365 ns (68%) , respectively. The obtained decay time in AE=Mg and Ca are comparable to the Ce:$LaCl_3$ single crystal in the previous report[4]. Light output of the Ce:$LaCl_3/CaCl_2$ eutectic can be increased by getting well aligned rod shape eutectic structure and improving optical transparency.

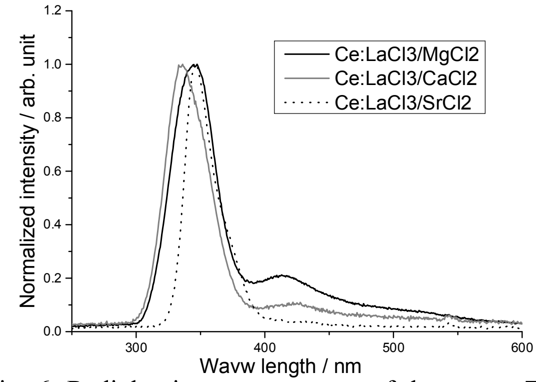

Fig. 6. Radioluminescence spectra of the grown T eutectic measured under X-ray irradiation.

3. Conclusion

Ce doped $LaCl_3/AECl_2$ (AE=Mg,Ca,Sr) eutectics were grown by the BZ method. The eutectics showed optical transparency like bundle optical fibers. The grown eutectic structure showed mixture of rods and plates shape. This mixed structure was aligned with length of around 300-400 μm. In this study, Ce:$LaCl_3/CaCl_2$ eutectic was most promising scintillator for X-ray imaging application using its PSSFs structures because of its higher light output and smaller total reflection anger between the eutectic interface. The light output of Ce:$LaCl_3/CaCl_2$ eutectic can be increased by getting well aligned rod shape eutectic structure and improving optical transparency. Increase of the growth size can improve the homogeneousity of eutectic structure and energy resolution.

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