Extended Abstracts of the 2014 International Conference on Solid State Devices and Materials, Tsukuba, 2014, pp212-213

$LiF/CaF_2/LiBaF_3$ ternary fluoride eutectic scintillator

Kosuke Hishinuma[1], Kei Kamada[2,3], Shunsuke Kurosawa[1,2] , Akihiro Yamaji[1], Jan Pejchal[2,4] ,

Yuui Yokota[2] , Yuji Ohashi[1] ,Akira Yoshikawa[1,2,3]

[1]Institute for Material Research, Tohoku University. 2-1-1 Katahira Aoba-ku, Sendai, Miyagi, 980-8577, Japan
E-mail: kosuke123@imr.tohoku.ac.jp
[2]New Industry Creation Hatchery Center, Tohoku University. 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai, Miyagi, 980-8579, Japan
[3] C&A corporation.T-Biz, 6-6-10 Aoba, Aramaki, Aoba-ku, Sendai, Miyagi, 980-8579, Japan

[4]Institute of Physic AS CR, Cukrovarnicka 10, 16253 Prague 6, Czech Republic

Abstract

$LiF/CaF_2/LiBaF_3$ eutectic scintillators were grown by the m-PD method. In the solidified eutectic the phases were uniformly distributed in the transverse direction and slightly aligned along the growth direction. For the Eu-doped samples, the expected emission peak observed at 420 nm was ascribed to Eu^{2+} 5d-4f transition from $Eu:CaF_2$ under X-ray excitation. The Li concentration in the $LiF/CaF_2/LiBaF_3$ eutectic is around 67.9 mol%. This is the advantage for neutron detection because ^6Li has high neutron capture cross-section. Due to the expected high sensitivity, the grown eutectic scintillator is promising candidate for application in neutron imaging such as nondestructive inspection, etc.

In our presentation, relation between chemical composition of starting materials, growth rate and volume ratio of eutectic will be discussed. Furthermore, other scintillation properties will be reported.

1. Introduction

Inorganic scintillators have been playing a major role in many fields of radiation detection, including medical imaging, security, astrophysics, and oil well logging. In these applications, scintillators for thermal neutron detection have recently attracted much attention due to depletion of the resources of ^3He gas. Up to now, most of the thermal neutron detectors are the ^3He gas counters with high thermal neutron cross section and low background gamma-ray sensitivity. However, the recent demand for ^3He highly exceeds its supply because demand for neutron detectors for security and oil well logging applications has been increased. This huge discrepancy between the demand and supply motivates us to develop novel thermal neutron scintillators which would replace the contemporary ^3He based systems.

In this decade, some novel inorganic scintillators for neutron detection have been reported. Among them, $LiCaAlF_6$ based scintillator is the most promising candidate [1,2]. $LiCaAlF_6$ contains Li in the host lattice and has the pulse shape discrimination ability to distinguish between neutrons and gamma-rays. As it is single crystal, Li content is limited by the chemical formula and cannot be increased. Thus, if we need higher neutron detection efficiency, we have to increase the Li concentration in another way.

In this study, as a candidate for novel neutron detectors, Eu doped $LiF/CaF_2/LiBaF_3$ eutectic scintillators were developed. The most important advantage of this eutectic scintillator is the Li content which is higher than that of the conventional neutron scintillators like Li-glass, Eu:LiI, or LiFIZnS [3]. In addition, here we propose submicron-diameter phase-separated scintillator fibers (PSSFs) with both the properties of an optical fiber and a capability of neutron-to-light conversion. The PSSFs were fabricated as a directionally solidified eutectic (DSE) system. The DSE systems have been discovered for various material systems for many applications [4-6]. In PSSFs, the light emitted from the scintillator fibers is confined and transported along the fiber direction by a total reflection mode as shown in Fig.1(a), so that high-resolution neutron imaging can be achieved.

2. Experimental Procedures

The starting material was prepared from LiF, CaF_2, BaF_2 and EuF_3 powders (4N). $LiF/CaF_2/LiBaF_3$ ternary eutectics were grown at the composition of the eutectic point. Crystal growth was performed by the micro-pulling-down (μ-PD) method with the tight -vacuum chamber. The starting composition was $67.89\%LiF/16.14\%CaF_2/15.81\%BaF_2/0.16\%EuF_3$.
Mixed powder was set in a graphite crucible with a 3 mm die which has a 2 mm hole in the center. The crucible was heated by the high- frequency induction coil in Ar/CF_4 gas mixture after baking process under high vacuum

(~10^{-4} Pa). The melt in the crucible was pulled down using Platinum wire as the seed at 1.0 mm/min. The detail of crystal growth by the μ-PD method was described in Ref. [7]. Circular samples were obtained from the grown crystal. The cut surface was optically polished and the eutectic phase structure was observed by back scattered electron image (BEI) using Hitachi S3400N scanning electron microscope. The phases in the eutectics were determined by XRD analysis. The radioluminescence spectra were investigated under x-ray excitation.

3. Results.

The $LiF/CaF_2/LiBaF_3$ eutectics were prepared by the μ-PD method. The rod-shaped sample with a length of 48mm and a diameter of 2 mm was prepared as shown in Fig.2 (a). The BEI image of transverse cross-section of the eutectic is shown in Fig.2(b). The three phases seem to be uniformly dispersed. The BEI of vertical cross-section is shown in Fig.2(c). Mainly black phase and white phase were well aligned along the growth direction. These phases were determined as follows White:LiF, Gray:CaF_2 and Black:$LiBaF_3$.The phases were determined using EDX and XRD pattern shown in Fig.3. Radioluminescence spectrum measured under x-ray excitation is shown in Figure 4. The expected emission peak observed at 420 nm ascribed to Eu^{2+} 5d-4f transition from Eu:CaF_2. That is good agreement with Ref.[8]. The emission peak at 360nm has been tentatively ascribed to the emission of $LiBaF_3$ host.

4. Conclusion

We have fabricated the $LiF/CaF_2/LiBaF_3$ eutectic scintillator by directional solidification. Li concentration in the $LiF/CaF_2/LiBaF_3$ eutectic is around 67.9 mol%. This is the advantage for neutron detection because ^6Li has high neutron capture cross-section. Our PSSFs system is promising for neutron imaging application such as detector for neutron diffraction, nondestructive inspection, etc. In the eutectic each phase was uniformly dispersed in transverse direction and slightly aligned along the growth direction.

In the presentation, relation between chemical composition of starting materials, growth rate and volume ratio of eutectic will be discussed. Furthermore other scintillation properties will be reported.

References

[1] A. Yoshikawa, N. Kawaguchi, et. al., IEEE Trans. Nucl. Sci., 56 (2009) 3796.

[2] N. Kawaguchi, A. Yoshikawa, et. al., Nucl. Instrum. Methods. A, 652 (2011) 351.

[3] Carel W. E. van Eijk IEEE Trans. Nucl. Sci., 59(5)(2012)2242.

[4] A. Yoshikawa , et. al., J. Cryst. Growth 218(2000)67.

[5] J. Llorca, et. al., Prog. Mater Sci. 51, (2006) 711.

[6] Y. Ohashi, A. Yoshikawa, et. al., App. Phy. Lett. 102, (2013) 05197.

[7] A. Yoshikawa, et. al., Opt. Mater. 30 (2007) 6.

[8] B. Moine, et. al., J. Lumin., 50 (1991) 31-38.

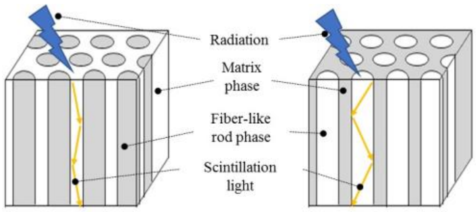

Fig. 1. Schematic view of PSSFs for light-guide system.

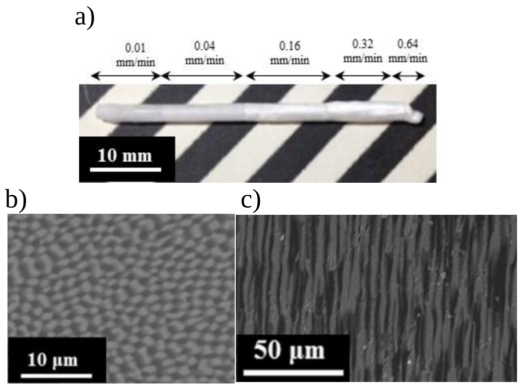

Fig. 2. Structure of $LiF/CaF_2/LiBaF_3$ ternary eutectic scintillator, a) photograph of $LiF/CaF_2/LiBaF_3$ ternary eutectic scintillator fabricated by micro-pulling down method,,b)BEI of transverse cross-section of the polished crystal, c)BEI of vertical cross-section along the growth direction of the polished crystal.

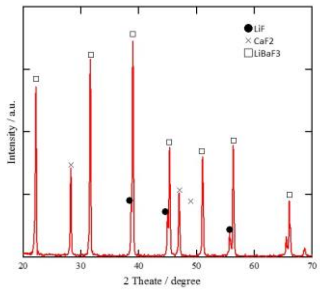

Fig. 3. The powder XRD pattern of the grown eutectic

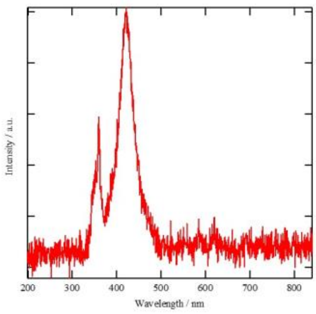

Fig. 4. Radio-luminescence spectra under X-ray.