

## Evaluation of Ce<sup>3+</sup> and alkali metal ions Co-doped LiSrAlF<sub>6</sub> crystalline scintillators



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### HIGHLIGHTS

- ▶ Ce-doped and alkali metal co-doped LiSAF crystals were grown by  $\mu$ -PD method.
- ▶ Alkali metal co-doped crystals showed higher light yield than Ce only doped crystal.
- ▶ Decay time of alkali metal co-doped LiSAF were longer than that of Ce only doped one.

### ARTICLE INFO

#### Article history:

Received 16 October 2012

Received in revised form

5 December 2012

Accepted 18 December 2012

#### Keywords:

LiSrAlF<sub>6</sub>

Thermal neutron

Ce<sup>3+</sup> 5d-4f transition

### ABSTRACT

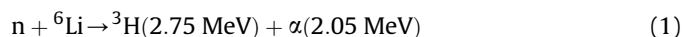
High scintillation efficiency of Eu-doped LiSrAlF<sub>6</sub> (LiSAF) and LiCaAlF<sub>6</sub> (LiCAF) codoped with alkali metal ions has been reported in our recent studies. Thus in this paper, we demonstrated the scintillation properties of 1% Ce-doped LiSAF crystals with 1% alkali metal ions co-doping to increase the light yield and understand the scintillation mechanism. The crystals showed intense emission band corresponding to the 5d-4f transition of Ce<sup>3+</sup>, and their light yields under thermal neutron excitation were higher than that of the Ce only doped crystal. Especially, the light yield of Ce–Na co-doped crystal exceeded about two times that of Ce only doped one.

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### 1. Introduction

Recently, the study of the alternative thermal neutron detection techniques have been started because the supplies of <sup>3</sup>He gas have been diminished due to its increased demand in the world (Kouzes et al., 2010). The techniques are used in a wide variety fields such as homeland security, astrophysics, oil well logging and so on. In the search for new scintillators, various scintillation properties are generally required, for example, high light yield, fast response, low hygroscopicity and linearity of the scintillation response with respect to the incident radiation energy. In the case of thermal neutron scintillator, the large capture cross-section for

thermal neutron is important parameter for the host scintillator matrix. Therefore, we have studied <sup>6</sup>Li-containing inorganic crystal as the host material because <sup>6</sup>Li isotope may capture thermal neutrons and convert them into ionizing particles according to the reaction (1).



The two charged particles produced in the reaction have a total kinetic energy of 4.8 MeV and scintillation light is produced along their ionization tracks. Additionally, low effective atomic number ( $Z_{\text{eff}}$ ) and low density ( $\rho$ ) are desired to decrease the sensitivity to  $\gamma$ -ray background.

Unfortunately previously developed scintillation materials still have not met all the above requirements, thus our group developed Eu- and Ce-doped LiCAF (Yanagida et al., 2011a; Yokota et al., 2011) and LiSAF (Yamaji et al., 2011, 2012; Yanagida et al., 2011b)

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crystalline scintillators. They showed good scintillation performance and no hygroscopicity. Moreover, in recent study, we succeeded to improve the light yield by co-doping with alkali metal ions, especially, the Na co-doped Eu:LiCAF outperformed the Eu:LiCAF by about 30% with its light yield of  $40,000 \pm 4000$  ph/n (Yanagida et al., 2011c). In the case of the Ce doped ones, though they showed fast response assigned to 5d–4f transition of  $Ce^{3+}$  ion, their light outputs are insufficient (about 3000 ph/n) (Yanagida et al., 2011b) for use in actual applications.

In this work, Ce-alkali metal (Na, K, Rb and Cs) co-doped LiSAF crystalline scintillators were grown and investigated to understand the effect of co-dopant on scintillation properties. We evaluated (1) the phase structure and chemical composition of the crystals by XRD, (2) the optical properties by absorption spectra and photoluminescence decay time, and finally, (3) the scintillation properties by radioluminescence spectra and pulse height spectra. From these experiments, we determined the suitable co-dopant for Ce doped LiSAF crystalline scintillator.

## 2. Experiments

Crystal samples with Ce 1 mol% and alkali metal 1 mol% co-doped LiSAF were grown by the Micro-Pulling Down ( $\mu$ -PD) method with an RF heating system (Yoshikawa et al., 2004). Mixtures of 4N LiF (95%  $^6LiF$ ),  $SrF_2$ ,  $AlF_3$ , NaF, KF, RbF and CsF powders produced by Stella Chemifa Corporation Co. Ltd. were used in the stoichiometric ratio as starting materials. After the starting powders were put into the carbon crucible, the chamber was evacuated down to  $10^{-4}$  Pa and heated to about 300 °C for baking out for several hours. Then, the atmosphere was changed in the crucible with the high purity Ar +  $CF_4$  gas (= 9:1) and finally, the crucible was heated up to the melting point of the LiSAF. A thin Pt wire was used as the seed crystal and the pulling speed was 5–6 mm/h.

The grown crystals were cut to the plates of 1 mm thickness and then polished to evaluate the optical and scintillation properties. To identify the phase of the grown crystals, powder X-ray diffraction (XRD) was carried out using RINT-2000 (RIGAKU). The X-ray source was  $CuK\alpha$  (accelerating voltage: 40 kV, beam current: 40 mA).

Transmittance spectra and reflectivity were measured with spectrophotometer (V-550, JASCO and UV-2550, Shimadzu, respectively) in the 190–900 nm wavelength range. The tungsten lamp (370–900 nm) and the deuterium lamp (190–370 nm) were used as a light source. The results obtained from the spectra were used to calculate the absorption coefficient. To study the luminescence properties of the crystals, photoluminescence decay times were measured by a modified spectrofluorimeter (FLS920 Edinburgh Instruments). A hydrogen nanosecond flashlamp was used for the photoluminescence decay measurements as an excitation source using the method of time-correlated single photon counting. Using the detection part of the same spectrofluorimeter, radioluminescence spectra under  $^{241}Am$   $\alpha$ -ray were measured. The  $\alpha$ -ray excitation simulates the  $^6Li(n, \alpha)^3H$  reaction where charged particles excite the scintillation material with high Q-value of 4.8 MeV. The scintillation light yields were evaluated with Photo Multiplier Tube (PMT, R8778, Hamamatsu) with which they were coupled with an optical grease (Krytox16350, DuPont). The crystals were covered with Teflon tapes and the  $^{252}Cf$  ( $<3.7$  MBq) radioisotope was used as the neutron source. The signals were directly fed into a shaping amplifier (ORTEC 572) and a multichannel analyzer (MCA, Amptek 8000A). The PMT was surrounded by 43 mm thick polyethylene layer serving as the moderator and reflector. In addition, Pb block of 50 mm thickness was set between the crystal and the  $^{252}Cf$  source to reduce gamma-ray background. In the measurements, a Li-glass scintillator GS20 that has light yield of 6000 ph/n (Van Eijk, 2004)

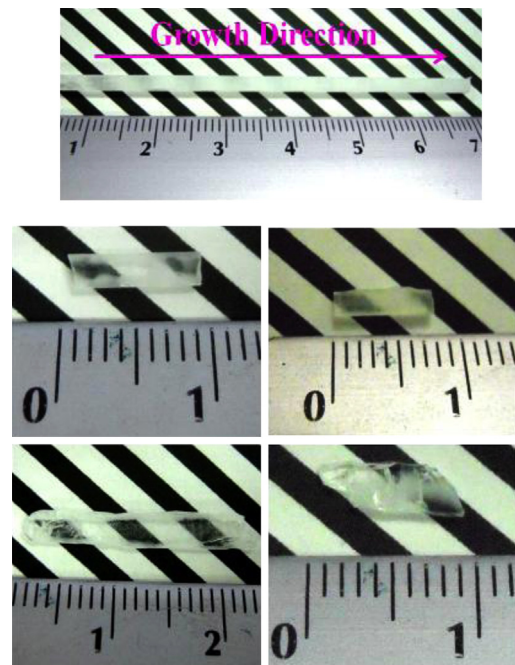


Fig. 1. As grown of Ce–K co-doped LiSAF (top) and polished grown crystals. Clockwise from the top left, Ce–Na, Ce–K, Ce–Cs and Ce–Rb co-doped LiSAF crystals.

was used as a reference sample. At the same time, decay time profiles were also recorded by WE7311 digital oscilloscope (Yokogawa).

## 3. Result

The crystals of Ce 1% and alkali metal (Na, K, Rb and Cs) 1% co-doped LiSAF were successfully grown by  $\mu$ -PD as shown in Fig. 1. Despite most of the parts were transparent, some milky parts due to secondary phase were also observed in the crystal. To confirm the phase of the milky parts, the powder XRD patterns of Ce–Na co-doped and non-doped LiSAF crystals were compared in Fig. 2(a). The secondary phase was identified as  $CeF_3$  and then, this is consistent with the results of heavily Ce doped LiSAF (Yamaji et al., 2012). The existence of alkali metal phase or compounds were not detected. Additionally, the lattice constants of the crystals are summarized in Fig. 2(b). According to this result, the  $a$  axis slightly decrease compared to non doped one due to dopant, while the  $c$  axis decreased.

Taking into account transmittance and reflectivity, the absorption coefficient was calculated using Lambert–Beer law, and the results are shown in Fig. 3. All the crystals showed sharp absorption peaks at 270 nm due to 4f–5d transitions of  $Ce^{3+}$  ion. The systematic effect of co-doping on absorption wavelength was not clearly observed. The decay times under 270 nm excitation were determined using a single exponential equation fitting by a deconvolution with instrumental peak, the determined decay times for Ce only, Ce–Na, K, Rb and Cs co-doped crystals were 23, 28, 27, 27 and 24 ns, respectively. Thus, the decay times of the co-doped crystals were comparable or slightly longer than that for the Ce only doped one and these results are consistent with those of Eu-alkali metal co-doped LiSAF.

Then, the radioluminescence spectra of the crystals under  $^{241}Am$   $\alpha$ -ray excitation are shown in Fig. 4. All the crystals showed intense emission band around 280–400 nm and this ultraviolet emission was assigned to the transition from  $Ce^{3+}$  5d state to the 4f ground state levels  $^2F_{5/2}$  and  $^2F_{7/2}$ . The effect of co-doping on the

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