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Growth and optical properties of LiF/LaF_3 eutectic crystals

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Abstract

Neutron imaging devices employing a scintillator can be used in various fields, and eutectic crystals can be suitable for the imaging with a fine position resolution of a few hundred micrometers. Since LiF and LaF_3 have different refractive indexes of 1.41 and 1.64 at 300 nm, respectively, the eutectic crystal is expected to behave as a scintillator with light guiding properties. Thus, the optical properties of Ce-doped LiF/LaF_3 eutectic crystal grown by micro-pulling down method were investigated. The light output of LiF/Ce:LaF_3 eutectic crystal was relatively small. The emission peaks at 300 nm originating from Ce^{3+} of 5d–4f transition were observed under excitation by UV photons and 5.5 MeV alpha rays. Moreover, the photo-luminescence decay time of Ce-doped LiF/LaF_3 eutectic crystal was estimated to be 17 ± 1 ns.

Keywords: Eutectic crystal; Neutron scintillator; LaF_3; LiF

1. Introduction

Neutron detection and imaging devices are (expected to be) used in various fields such as crystallography, homeland security, etc.[1,2] Since ^{3}He has an unusually large cross-section for neutron capture (approximately 5300 barns for thermal neutrons[3]), a ^{3}He-gaseous detector has been used for neutron detection.[3,4] However, the ^{3}He sources are being depleted, and an alternative suitable nuclei are searched for. Then several scintillator crystals containing elements with high-cross-section nuclei, like ^{10}B (∼3800 barns[3]), ^{6}Li (∼940 barns[3]), have been investigated for detection of thermal neutrons.[5–8] In addition, some groups have studied neutron imaging with a scintillator.[9–11]

When ^{6}Li undergoes a nuclear reaction after absorption of a thermal neutron, it disintegrates into an alpha particle and a triton ($^3H_1$) as follows:

$$^6\mathrm{Li} + n \rightarrow {}^3\mathrm{H}_1 + \alpha + 4.78\,\mathrm{MeV} \quad (1)$$

the 4.78-MeV energy, called Q-value, is distributed between the triton (2728 keV) and the alpha particle (2055 keV). Due to these particles, scintillation light is emitted. On the other hand, ^{10}B undergoes the following reactions:

$$^{10}\mathrm{B} + n \rightarrow 7\mathrm{Li}(0.8\,\mathrm{MeV}) + \alpha(1.5\,\mathrm{MeV}) + \gamma(0.5\,\mathrm{MeV}) \quad (2)$$

and

$$^{10}\mathrm{B} + n \rightarrow 7\,\mathrm{Li}(1\,\mathrm{MeV}) + \alpha(1.8\,\mathrm{MeV}) \quad (3)$$

where the branching ratios of (2) and (3) are 94 and 6%, respectively. Although ^{10}B has a larger cross section for a thermal neutron than ^{6}Li, the Q-value and the alpha-ray energies produced by reactions (2) or (3) are twice as small as that of the reaction (1). Moreover, gamma rays can be generated from the (2) as a noise. Thus, signal-noise-ratio for ^{6}Li-containing scintillator is expected to be better than that of ^{10}B-containing scintillator. We have investigated neutron scintillators containing ^{6}Li, such as Ce-doped $LiCaAlF_6$,[8] which had good signal-noise-ratios (alpha–gamma ratio). Since fluoride crystals have generally low melting points with respect to the other crystals such as oxide crystals, they can be produced with a lower cost.

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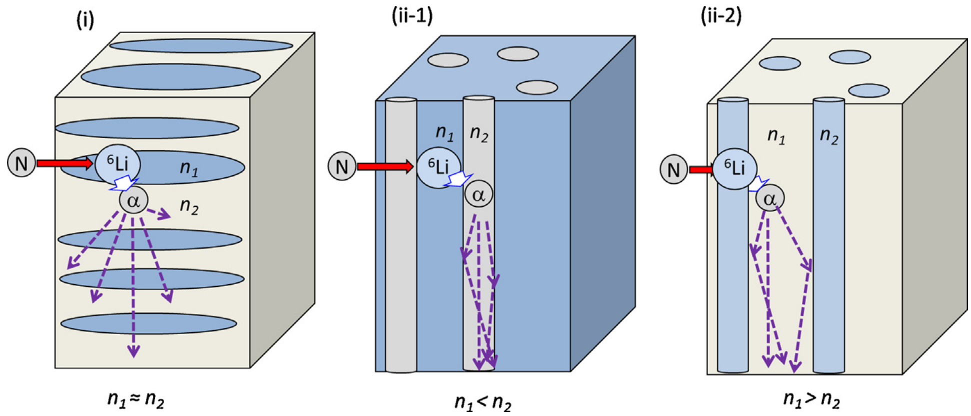

Fig. 1. Schematic view of eutectic scintillation crystals. n_i ($i = 1, 2$) denote the refractive indexes. (i) normal type ($n_1 \approx n_2$), (ii) light-guide type, $n_1 < n_2$ and $n_1 > n_2$ for (ii-1) and (ii-2), respectively. Dotted arrows denote scintillation light.

In order to readout position-sensitive signals (electrons) in the gaseous neutron detector, micro pattern gaseous detectors with an anode pitch of a few hundred micrometers are used.[4] As a result, the gaseous type has a position resolution of a few hundred micrometers. Meanwhile, the scintillation type has worse position resolution, because a scintillation array camera has a pixel pitch of more than ~1 mm. Even if a monolithic crystal is coupled to a fine structure photo detector, like a Charge Coupled Device (CCD) camera, the scintillation light would be spread in the crystal.

Recently, a light-guide scintillator based on eutectic crystals consisting of components with different refractive indexes has been reported for X-ray imaging.[12] This needle-like shape crystal scintillator was grown by micro-pulling (μ-PD) down method. Thus, we decided to study ^{6}Li-containing eutectic crystals for neutron detector in this paper.

Fig. 1 shows a schematic view of position-sensitive eutectic crystals for neutron scintillator: (i) normal type (Lamellar structure) and (ii) cylinder type consisting of two components with different refractive indexes, respectively. The latter type has a cylinder (tube)-shape component. When this cylinder part has a higher refractive index than the other component, the scintillation light can be guided through the cylinder like in an optical fiber cable (Fig. 1(ii-1)). Both types consist of LiF as a neutron-alpha ray converter and an emitter excited by the resulting alpha rays. Using Monte Carlo method including Bethe-Bloch formula, the stopping range of 2055-keV alpha ray (distance of passing through matter until their energy become to zero) is 5.5 μm in the LiF crystal, therefore the Lamellar pitch or phase width should be less than 5.5 μm so that the alpha ray can reach the emitter zone.

LiF and LaF_3 have refractive indexes of 1.41 and 1.64 at 300 nm[13,14] therefore LaF_3 is expected to guide or separate the scintillation light into position-sensitive photo detectors. In addition, Moses et al. reported that 1-mol% Ce doped LaF_3 has a scintillation light output of 440 photons/MeV excited by gamma rays,[15] thus, Ce:LaF_3 can be also used as emitter. Although the light output of Ce:LaF_3 is smaller when compared to well-known fluoride scintillators such as CaF_2, BaF_2, this material has a larger refractive index (n) than CaF_3 ($n = 1.46$ at 300 nm), BaF_2($n = 1.51$ at 300 nm). In this paper, first we report the structure of LiF/LaF_3 eutectic crystals grown by the μ-PD method, and then we show the optical properties of Ce-doped LiF/LaF_3 eutectic crystals in order to study its feasibility for the neutron scintillator.

2. Materials and methods

Pure and Ce-doped LiF/LaF_3 eutectic crystals were grown by the μ-PD method (details of the μ-PD method are described in Refs. 16,17). Here the eutectic point of LiF/LaF_3 system is LiF: LaF_3 = 83.8: 16.2 at approximately 770 °C,[18] so the initial composition was $(LiF)_{83.8}\{(La_{1-x}, Ce_x)F_3\}_{16.2}$ ($x = 0$ or 0.01). The powders of the crystal materials (LiF, LaF_3 and CeF_3) were of 99.99% purity, and the atmosphere was a gas mixture of Ar:CF_4 in a pressure ratio of 97:3 at 1 atm (sealed). A Pt wire was used as the seed, and the pulling rates were 0.1, 0.5, 1.0, 2.0 mm/min for pure LiF/LaF_3 eutectic crystals. In order to confirm that a phase width (Lamellar pitch) is of less than approximately 5.5 μm, field emission scanning Electron Microscope/Electron Probe Micro Analyzer (FE-SEM/EPMA, JEOL, JXA-8530F,10 kV, 20 nA) was used. After checking, 1-mol% Ce-doped LiF/LaF_3 eutectic crystals were grown by the μ-PD method with an optimized pulling rate (we decided the rate of 0.5 mm/min). In order to check the phase of the obtained crystals, powder X-ray diffraction analysis was performed from 20° to 80° using a diffractometer (RIGAKU, RINT2000). The X-ray source was using CuKα line with an accelerating voltage of 40 kV, and tube current of 40 mA.

We investigated some optical properties of the samples after cutting and polishing; (i) Transmittance was measured with a spectrophotometer (JASCO, V-530), (ii) photo-luminescence spectra (emission and excitation) were measured with a spectrofluorometer (Edinburgh Instruments, FLS920), (iii) photo-luminescence decay curves were obtained with the spectrofluorometer and a Flash lamp (Edinburgh Instruments, nF900) (iv) the radio-luminescence spectra at room temperature were measured using the same spectrofluorometer and 5.5-MeV alpha rays (^{241}Am) as the excitation source.