

Article

Fabrication of $\text{CeCl}_3/\text{LiCl}/\text{CaCl}_2$ Ternary Eutectic Scintillator for Thermal Neutron Detection

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Abstract: To date, ^3He gas has been commonly used to detect thermal neutrons because of their high chemical stability and low sensitivity to γ -rays, owing to their low density and large neutron capture cross-section. However, the depletion of ^3He gas prompts the development of a new solid scintillator for thermal neutron detection to replace ^3He gas detectors. Solid scintillators containing ^6Li are commonly used to detect thermal neutrons. However, they are currently used in single crystals only, and their ^6Li concentration is defined by their chemical composition. In this study, ^6Li -containing eutectic scintillators were developed. CeCl_3 was selected as the scintillator phase because of its low density (3.9 g/cm^3); high light yield (30,000 photons/MeV); and fast decay time with four components of 4.4 ns (6.6%), 23.2 ns (69.6%), 70 ns (7.5%) and $>10 \mu\text{s}$ (16.3%), owing to the Ce^{3+} 5d-4f emission peak at approximately 360 nm. Crystals of the CeCl_3 , LiCl and CaCl_2 ternary eutectic were fabricated by the vertical Bridgman technique. The grown eutectic crystals exhibited Ce^{3+} 5d-4f emission with a peak at 360 nm. The light yield was 18,000 photons/neutron, and the decay time was 10.5 ns (27.7%) and 40.1 ns (72.3%). Therefore, this work demonstrates optimization by combining a scintillator phase and Li-rich matrix phase for high Li content, fast timing, high light yield and low density.

Keywords: scintillator; melt growth; vertical Bridgman technique; thermal neutron detection; eutectic system



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1. Introduction

Technologies using neutron sources and their detectors are used in many application fields, such as materials analysis, crystal chemistry, imaging of interior structures, well logging, monitoring of nuclear facilities and basic research in condensed matter physics. Recently, Li-ion batteries have garnered attention with the increasing focus on environmental protection and demand for thermal neutron detectors suitable for the internal non-destructive testing of Li-ion batteries [1,2].

To date, thermal neutron detectors using ^3He gas have been used for many years due to their low density, low sensitivity for gamma-rays and large neutron capture cross-section. However, the recent increase in the demand for neutron detectors for national security purposes and to address the export restrictions on ^3He gas of the USA, which is the largest producer of ^3He gas, resulted in the significantly higher demand for ^3He gas than its supply [3]. Hence, the development of solid state scintillator materials for thermal neutron detection to replace ^3He gas counters has attracted considerable interest, and research is being conducted on alternative neutron detector technologies [4–6].

Prospective materials for solid state neutron scintillators contain ${}^6\text{Li}$ because of its high thermal neutron capture cross-section, producing ${}^6\text{Li}(n, \alpha){}^3\text{H}$ nuclear reaction with a high Q value of 4.8 MeV (Equation (1)) [7]. In addition, this high Q value can achieve ${}^6\text{Li}$ -containing scintillators with a high light yield for thermal neutrons. Therefore, in previous studies, ${}^6\text{Li}$ -based single crystalline scintillators, such as LiCaAlF_6 scintillators doped with Ce^{3+} and Eu^{2+} , have been developed and used in practice, demonstrating good scintillation properties under neutron excitation from a ${}^{252}\text{Cf}$ source [8–12] with the reaction:



However, as these solid-state scintillators are single crystals, the ${}^6\text{Li}$ content is fixed by their chemical compositions. Therefore, we developed eutectic crystals of fluorides, chlorides, bromides, iodides and other eutectic scintillators, such as ${}^6\text{LiF}/\text{CaF}_2$ [13], ${}^6\text{LiF}/{}^6\text{LiYF}_4$ [14], ${}^6\text{LiF}/{}^6\text{LiGdF}_4$ [15], ${}^6\text{LiF}/\text{SrF}_2$ [16], ${}^6\text{LiF}/\text{LaF}_3$ [17], ${}^6\text{LiCl}/\text{CeCl}_3$ [18], ${}^6\text{LiCl}/\text{Li}_2\text{SrCl}_4$ [19], ${}^6\text{Li}_2\text{SrCl}_4/{}^6\text{LiSr}_2\text{Cl}_5$ [20], ${}^6\text{LiCl}/\text{BaCl}_2$ [21], ${}^6\text{LiBr}/\text{CeBr}_3$ [22], ${}^6\text{LiBr}/\text{LaBr}_3$ [23] and ${}^6\text{LiSrI}_3/\text{LiI}$ [24]. The schematic for the use of eutectic materials in neutron scintillator applications is shown in Figure 1. In the Li-containing phase, only the incident neutrons were converted to secondary ionized particles by Equation (1). A certain number of α -rays passed through the small-sized, Li-containing phase to reach other phases. Subsequently, the α -ray is converted to scintillation light in the scintillator phase. Thus, the scintillator phase can be considered as a luminescent material excited by α -ray. Further, the most prominent advantage of eutectic scintillators is the ability to increase their ${}^6\text{Li}$ content, unlike single crystals, thereby enhancing their ability to capture thermal neutrons.

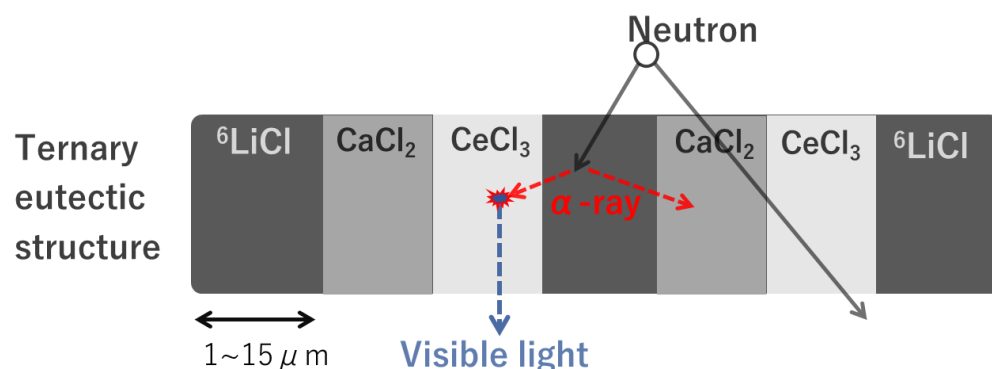


Figure 1. Schematic of the ternary eutectic scintillator in this study.

In this study, the ternary eutectic of $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ was developed using the vertical Bridgman (VB) technique as candidates for new thermal neutron detector materials. CeCl_3 single-crystal scintillators have low density of 3.9 g/cm^3 , fast decay time of approximately 25 ns owing to the Ce^{3+} 5d-4f emission peak at approximately 360 nm and good light yields of 30,000 photons/MeV under the irradiation of gamma rays [25]. Thus, CeCl_3 was chosen as the scintillator crystal phase in this study. In a previous study, the growth rate of the eutectic crystal in ${}^6\text{LiCl}/\text{CeCl}_3$, which is a eutectic scintillator using CeCl_3 as the scintillator phase, was investigated to adjust the grain size of each phase, which is an important factor in eutectic scintillators for thermal neutron detection. In particular, an excessively large grain shape resulted in the loss of the energy caused by the nuclear reaction ${}^6\text{Li}(n, \alpha){}^3\text{H}$ before reaching the scintillator phase. Therefore, in this study, in addition to the scintillator phase selection, we focused on ternary eutectic crystals and attempted to reduce the grain size of each phase by creating scintillator crystals in ternary eutectic systems to efficiently transfer energy to the scintillator phase. The luminescence and radiation responses under the irradiation of thermal neutrons were also evaluated.

2. Materials and Methods

2.1. Crystal Growth

CeCl₃, CaCl₂ and ⁶Li-enriched (95%) LiCl powders (4N purity) were prepared as the initial raw material. CeCl₃/⁶LiCl/CaCl₂ eutectic was fabricated at the chemical composition ratio of 17.4 mol% CeCl₃: 62.7 mol% ⁶LiCl: 19.9 mol% CaCl₂ [26]. The starting powders were weighed, mixed and placed in a 4-mm inner diameter quartz ampoule in a glove box filled with argon gas. The ampoule was then removed from the glove box and baked at 180 °C under $\sim 10^{-1}$ Pa vacuum to eliminate water and air from the ampoule. At the end of the baking process, the ampoule was sealed. The ampoule was heated until the raw material melted and then the eutectic was fabricated by the VB technique at a pulling rate of 0.2 mm/min. Details of the fabrication method were described in the previous report [18–23].

2.2. Structural Analysis of the Eutectic

Eutectic samples were cut and polished into 1- and 3-mm pieces with a wire saw. Moreover, 3-mm crystals were cut horizontally along the growth direction. Samples of 1-mm thickness in circumferential and perpendicular directions were taken polished from the fabricated eutectic and mirror. The eutectic structures on the transverse and vertical cross-sections were observed by backscattered electron imaging (BEI) using a Hitachi S3400N scanning electron microscope. The X-ray diffraction (XRD) in the 2 θ range of 10–90° was performed to identify crystal phases in the eutectic using a Bruker D8 Discover diffractometer using a CuK α X-ray source with a tube current of 40 mA and accelerating voltage of 40 kV.

2.3. Evaluation of the Luminescence and Radiation Responses

The radioluminescence (RL) spectra were obtained under X-ray excitation using an Andor Technology SR-163 spectrometer equipped with an Andor Technology iDus420-OE charge-coupled device (CCD) detector. In addition, the cathodoluminescence (CL) spectra were measured for each phase of the eutectic crystal using a Horiba JSM-7001F, MP-32M equipped with photomultiplier tube (PMT) as the CL detector (R943-0, Hamamatsu, Shizuoka, Japan) and CCD as the CL map (Synapse Plus BIUV, Horiba, Kyoto, Japan). The light yield was estimated from the pulse height spectra of the eutectic sample and a GS20 (Li-glass) standard with the light yield of 7000 photons/neutron under thermal neutron (²⁵²Cf) excitation at room temperature using a Hamamatsu R7600U-200 photomultiplier tube (PMT) at an operating voltage of 600 V. The output signal was fed via a ORTEC 572A shaping amplifier and a two-channel USB Wave Catcher module into a personal computer. The decay curve was obtained using the same PMT. The output signal was recorded using a Tektronix TD5032B digital oscilloscope.

3. Results and Discussion

3.1. Growth Crystal and Phase Identification

A CeCl₃/⁶LiCl/CaCl₂ ternary eutectic with a diameter of 4.0 mm and length of 5.8 mm was fabricated by the VB technique. The fabricated eutectic in the quartz ampoule and 1-mm thick polished circumferential wafer are shown in Figure 2. The polished wafer sample exhibited a visible transparency along the pulling direction, as indicated by the visibility of the black cross on the back.

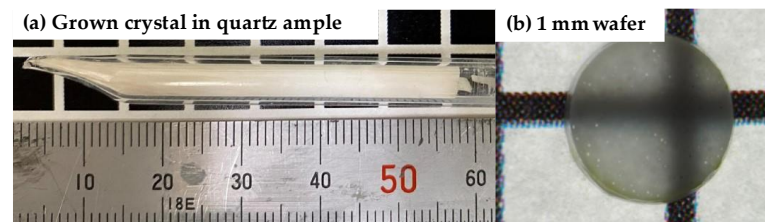


Figure 2. Images of the (a) as-grown crystal and (b) 1-mm polished wafer of the grown eutectic.

The powder XRD pattern of the fabricated eutectic is shown in Figure 3. Only CeCl_3 (hexagonal, $P6_3/m$, 176), LiCl (cubic, $Fm-3m$, 225) and CaCl_2 (bipyramidal, $Pnnm$, 58) phases were confirmed. The BEI of the wafer samples in the vertical and transverse cross-section are shown in Figure 4. The BEI results show the fiber-type eutectic structure of $\text{CeCl}_3/\text{LiCl}/\text{CaCl}_2$, which has the tendency to extend linearly along the crystal growth direction. The eutectic structure extended several tens to hundred micrometers in the growth direction with large variations in the grain sizes. Although the eutectic structures were obscured by the polishing scratches, their length was recorded to be several hundred micrometers. It is known as the Hunt–Jackson law that grain size in eutectics is inversely proportional to the square root of the solidification speed [27]. The difference in grain size in the eutectic suggests that there were areas where latent heat was efficiently eliminated, and the solidification rate was fast, and other areas where it was not. This is presumably due to the fact that unidirectional solidification did not follow the pulling direction and the solid–liquid interface was not flat. Uniformity of grain size is thought to be improved by optimizing eutectic growth conditions, such as the temperature gradient and pulling rate. From the powder XRD pattern and BEI results, the LiCl (black), CaCl_2 (gray) and CeCl_3 (white) phases were determined. In addition, the refractive indices of CeCl_3 , LiCl and CaCl_2 at 380 nm were determined to be 2.20, 1.67 and 1.52, respectively, denoting a large difference [28–30]. The transparency of the sample wafers can be ascribed the elongation of the eutectic structure, as similarly noted in previous reports [22,24].

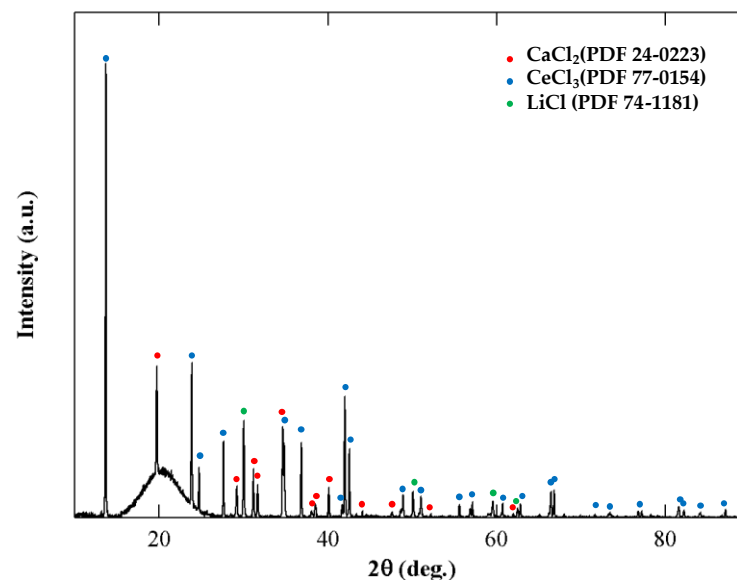


Figure 3. Powder XRD patterns of the $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ eutectic.

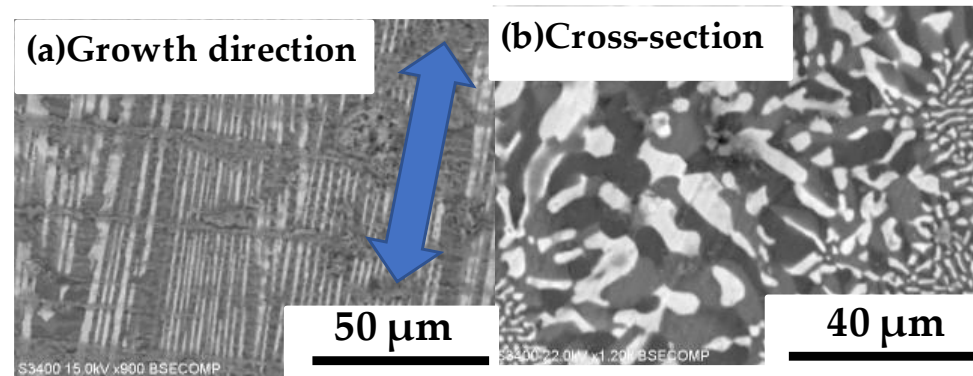


Figure 4. The BEI of the wafers of (a) vertical and (b) transverse cross-section. The blue arrow indicates the pulling direction.

Eutectics tend to take on a fiber-type structure when the volume ratio of the eutectic fiber phase is around 30% [22,24]. In this study, the theoretical volume ratio of $\text{CeCl}_3\text{:}^6\text{LiCl}\text{:CaCl}_2$ was 31.9:37.8:30.3, which is considered optimal for the ternary eutectic system. Thus, a fiber-type eutectic was easily obtained.

3.2. Luminescence and Radiation Responses

The RL spectra of the $\text{CeCl}_3\text{:}^6\text{LiCl}\text{:CaCl}_2$ eutectic sample under X-ray irradiation are shown in Figure 5. The fabricated eutectic sample showed the expected Ce^{3+} 5d-4f emission with a peak at approximately 360 nm, similar to that of CeCl_3 single crystal. This result is consistent with that of a previous report [25].

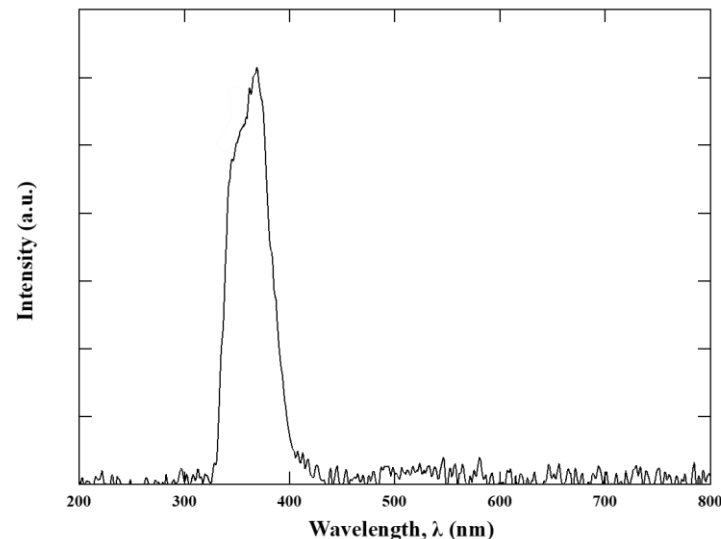


Figure 5. RL spectra of the eutectic sample under X-ray irradiation.

The CL spectra on the corresponding position of the CeCl_3 , LiCl and CaCl_2 phases in the wafer sample are shown in Figure 6. The electron beam irradiated all crystal phases, and the CL spectra in the wavelength range of 200–600 nm was obtained. The CL intensity on the CeCl_3 phase was much higher than that of the other crystal phases. Thus, CeCl_3 may be substituted for CaCl_2 . However, no Ce^{3+} 5d-4f emission was observed on the CaCl_2 phase. This indicates that only the CeCl_3 phase works as the scintillator phase, whereas only the $^6\text{LiBr}$ phase works as the neutron reaction phase. The CL measurements showed additional emission peaks at around 455, 680 and 750 nm. This may be due to the influence of hydrates deposited on the sample surface. In the CL measurement, the sample was polished and put into the apparatus in air, and the hydrates were gradually deposited on

the sample surface. The emission is obtained from the sample surface layer, so the influence of hydrates on the surface is inevitable in the CL.

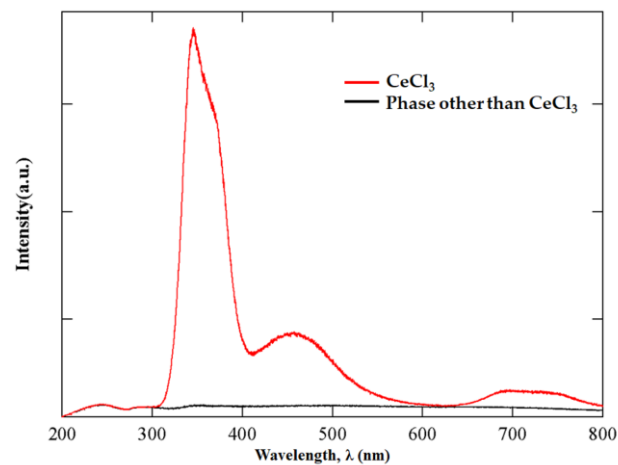


Figure 6. CL spectra on each crystal phase in the eutectic sample.

The pulse-height spectra of the fabricated eutectic and GS20 standard under thermal neutrons (^{252}Cf) irradiation are shown in Figure 7. The light yield of the eutectic was approximately 250% of GS20. Considering that the quantum efficiency (QE) at the emission peak of 395 nm for GS20 is 40% and the QE at the emission peak of 360 nm for the eutectic is 41%, the light yield of the eutectic was calculated to be 18,000 photons/neutron.

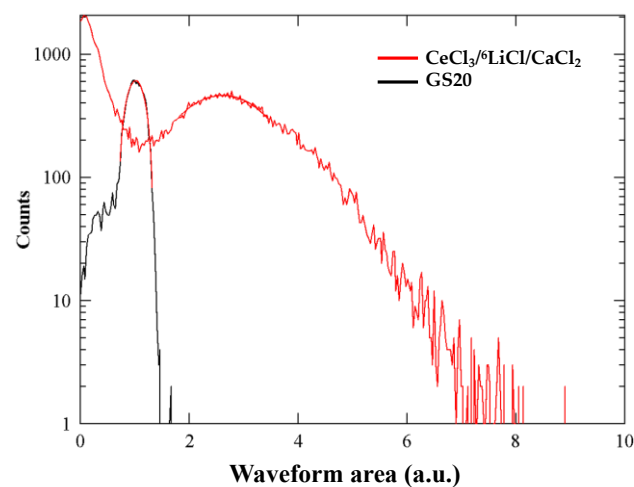


Figure 7. Pulse-height spectra of the $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ eutectic and GS20 standard under ^{252}Cf thermal neutron irradiation.

The scintillation decay curve of the eutectic excited by thermal neutrons (^{252}Cf) is shown in Figure 8. The decay time was approximated using Equation (2), which is the sum of two exponents:

$$I(t) = y_0 + \sum_{i=1}^2 A_i \exp\left(-\frac{t}{\tau_i}\right), \quad (2)$$

The ratio was calculated as

$$A_1 \tau_1 / (A_1 \tau_1 + A_2 \tau_2) \times 100\%, \quad (3)$$

where y_0 is the baseline; τ is the decay time; t is the time; A_i is a coefficient; and I is an intensity. The decay times were converged to two components of 10.5 ns (27.7%) and 40.1 ns (72.3%). The decay time and component ratio differed from the previous report of CeCl_3

single crystal [25]. This is assumed to be the effect of partial substitution of the Li^+ and Ca^{2+} into the Ce^{3+} site.

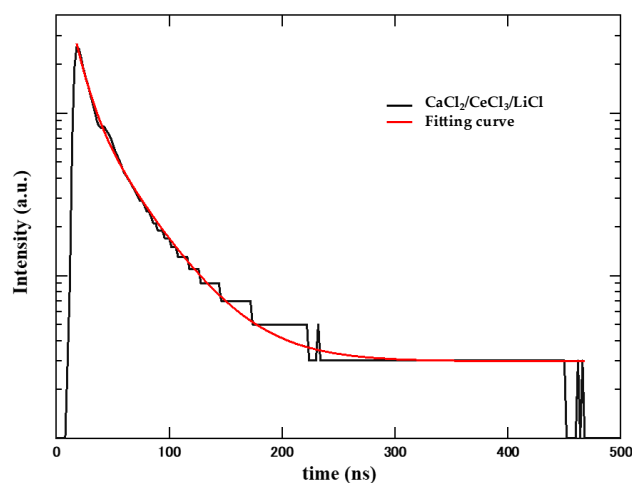


Figure 8. Scintillation decay curve of the grown eutectic sample under ^{252}Cf neutron excitation.

The density of the eutectic crystals was calculated to be as low as 2.98 g/cm^3 . The Li concentrations of the eutectic crystal, Li glass and Ce:LiCAF were 0.031, 0.028 and 0.016 mol/cm^3 , respectively [31]. The highest Li concentration was obtained in $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$. Therefore, $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ eutectic growth was successfully applied in developing a promising scintillator for thermal neutron detection.

4. Conclusions

Ternary eutectic $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ with the diameter of 4 mm was prepared by the VB technique in the quartz ampoule. Li-enriched (95%) LiCl powder was used for the eutectic fabrication to evaluate the thermal neutron responses. The results of the BEI and powder XRD analyses confirmed the existence of LiCl, CaCl_2 and CeCl_3 phases. In the $\text{CeCl}_3/{}^6\text{LiCl}/\text{CaCl}_2$ eutectic, Li has a high content of up to 0.031 mol/cm^3 and small density of 2.98 g/cm^3 . The eutectic has an Ce^{3+} 5d-4f emission peaking at 360 nm in the CeCl_3 phase, and it is consistent with previous studies on CeCl_3 single crystals. The light yield was approximately 18,000 photons/neutron with the two decay components of 10.5 ns (27.7%) and 40.1 ns (72.3%). These results indicate that the scintillator phase with excellent scintillation performance provides high light yield and fast decay time, while the Li-rich phase provides the high Li content thermal neutron scintillator. Although $\text{CeCl}_3/\text{LiCl}/\text{CaCl}_2$ has slight hygroscopic properties, it remains to be a promising material in terms of ${}^6\text{Li}$ concentration, density, light yield and decay time. As a fiber-type structure can be obtained in this eutectic, the transparency in the pulling direction and light yield can be enhanced by optimization of the eutectic fabrication conditions and diameter of fiber phases in future studies.

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