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Intrinsically activated $TlCaCl_3$: A new halide scintillator for radiation detection

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HIGHLIGHTS

- New intrinsically activated $TlCaCl_3$ has been grown by Bridgman technique.
- X-ray induced emission spectra show emission peak at 425 nm.
- Light yield of 30,600 ± 3060 ph/MeV is found under γ-rays excitation.
- Energy resolution of 5% (FWHM) is obtained under 662 keV γ-rays excitation.
- Two decay constants are obtained under 662 keV γ-rays excitation.

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ABSTRACT

A new intrinsically activated $TlCaCl_3$ scintillator has been developed by conventional Bridgman technique. The emission spectra under X-ray excitation show broad band spanning in the range of 350 −550 nm having maximum emission at 425 nm. Light output of 30,600 ± 3060 ph/MeV and energy resolution of 5% (FWHM) is recorded under 662 keV γ-rays excitation from a ^{137}Cs source. The scintillation decay time is bi-exponential with components of 317 ns and 727 ns having total light yield contribution of 44% and 56%, respectively. Due to high effective Z-number ($Z_{eff} = 65.5$) than the widely used commercial scintillators such as NaI: Tl ($Z_{eff} = 50.8$) and CsI: Tl ($Z_{eff} = 54$), the grown crystal will provide higher detection efficiency for X- and γ-rays. The measured scintillation properties showed that $TlCaCl_3$ could be the best candidate for the detection of X- and γ-rays in different applications.

1. Introduction

The discovery of $Tl_2LiGdCl_6$: Ce^{3+} (Kim et al., 2015) crystal has gained attention due to its promising scintillation properties and high effective atomic number ($Z_{eff} = 70$). Also, the presence of Tl^+ ion in the host lattice makes such compound very attractive due to their intrinsic luminescence, high Z_{eff}, and density (ρ). Therefore, a new research direction has been initiated to explore new Tl-based scintillators which can meet the requirements of different applications such as high Z_{eff} and density, excellent energy resolution, high photon yield, fast timing response and non-hygroscopic nature. Besides these, the host matrix offers high neutron detection efficiency. Recently, Tl_2LiYCl_6 (Kim et al., 2016a,b; Rooh et al., 2017a,b; Hawrami et al., 2017) crystal has been developed for neutron and γ-ray detection. Compared with the conventional scintillators CsI: Tl ($Z_{eff} = 54$, ρ = 4.53 g/cm^3) (Totsuka et al., 2012) and NaI: Tl ($Z_{eff} = 50.8$, ρ = 3.67 g/cm^3) (Hofstadter, 1948), the newly developed scintillators such as $Tl_2LiGdBr_6$: Ce^{3+} ($Z_{eff} = 66$, ρ = 5.30 g/cm^3) (Kim et al., 2016a,b), $Tl_2LiLuCl_6$: Ce^{3+} ($Z_{eff} = 71$, ρ = 5.06 g/cm^3) (Rooh et al., 2017a,b), Tl_2LaCl_5: Ce^{3+} ($Z_{eff} = 70$, ρ = 5.20 g/cm^3) (Kim et al., 2017a,b) and Tl_2LaBr_5: Ce^{3+} ($Z_{eff} = 67$, ρ = 5.90 g/cm^3) (Kim et al., 2017a,b) have high Z_{eff} and density. Therefore, these scintillators will provide higher detection efficiency for X- and γ-rays in different applications. In addition, high light yield, good energy resolution and fast decay time are also the advantages of these scintillators.

Since intrinsic scintillators need no dopant, therefore, the homogeneous growth of a crystal with low cost is an advantage. In the known commercial intrinsic scintillators, $Bi_4Ge_3O_{12}$ (Nestor and Huang, 1975) is widely used due to its high density and Z_{eff},

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however, it has low light yield, which limited its use in application where high light yield is mandatory. The recently reported Tl-based intrinsically activated $TlMgCl_3$ (Fujimoto et al., 2016) and $TlCdCl_3$ (Fujimoto et al., 2017) have shown good scintillation properties.

In this study, we focused on the growth of intrinsically activated $TlCaCl_3$, a new ternary halide scintillator. This crystal is grown from the melt of the desired material by vertical Bridgman method. The luminescence and scintillation properties of the grown single crystal are studied under X- and γ-rays irradiation at room temperature.

2. Experimental

The $TlCaCl_3$ crystal was grown from TlCl (5N) and $CaCl_3$ (3N) powders provided by Sigma Aldrich, by two zones vertical Bridgman technique. Due to the hygroscopic nature of the starting materials, all the preparations were performed inside an ultra-low humidity glove box in the continuous flow of Argon gas. The compounds were weighed according to the stoichiometric ratio and charged in the quartz ampoules. The powders were dried at 150 ^{O}C for several hours and sealed under vacuum conditions of ~10^{-7} Torr. A thermal gradient of 10 ^{O}C/cm was achieved by setting the upper and lower temperature zones at 700 ^{O}C and 400 ^{O}C, respectively. The crystal was grown at lowering speed of 12 mm/day. During crystal growth process no extra phase was observed and whole material melted at 680 ^{O}C as reported in ref (Korreng, 1914). Grown crystal was cut using diamond coated stainless steel wire. During cutting, crystal surface was covered with mineral oil to avoid direct exposure to air which could degrade the scintillation performance due to the hygroscopic nature of this crystal. The Z_{eff} and density of this material were measured to be 65.5 and 3.77 g/cm^3, respectively. The grown crystal is shown in Fig. 1.

The emission spectra of $TlCaCl_3$ crystal were measured by irradiating the crystal with X-rays. The X-rays was produced by operating the X-ray generator (DRGEM. Co.) with 80 kV and 1 mA power setting. A fiber optic spectrometer (QE65000) was used to analyze the emission spectrum. The scintillation response of the grown $TlCaCl_3$ crystal was measured under γ-rays excitation using ^{22}Na, ^{57}Co, ^{109}Cd,^{133}Ba and ^{137}Cs sources. Polished crystal of $TlCaCl_3$ was wrapped with Teflon tape (0.1 mm thickness) except one surface facing the entrance window of Hamamatsu R6233-100 PMT. The bias voltage of PMT during this measurement was set to be −500 V. Shaping time of 6 μs of Tennelec TC 245 spectroscopy amplifier was used to shape the PMT signals. The output signal from the amplifier was digitized by 25-MHz FADC and recorded to a personal computer by a USB2 connection. The recorded data were analyzed by root data analysis software (So et al., 2008).

The optically coupled $TlCaCl_3$ crystal to the Hamamatsu R6233-100 PMT was irradiated by 662 keV γ-rays for the measurement of decay time response. For this measurement, the PMT was biased with the high voltage of −1000 V. A 400-MHz FADC was used to convert the output analog signal of the PMT to digital signal (Kim et al., 2010). The recorded pulse height information was analyzed for the decay time measurement.

3. Results and discussion

3.1. X-ray induced luminesce

The radio luminescence spectrum of $TlCaCl_3$ measured at room temperature under X-ray excitation is shown in Fig. 2. The emission band is spanning in the range of 350–550 nm having the maximum emission peak at 425 nm. Similar broad band emission is also observed in the emission spectra of $TlMgCl_3$ (Fujimoto et al., 2016) and $TlCdCl_3$ (Fujimoto et al., 2017) scintillators. Therefore, the observed emission could be assigned to Tl^+ ion or self-trapped exciton (STE) emission. The luminescence obtained from this crystal is compatible with the quantum efficiency response of light sensors, therefore this crystal will be a good candidate for γ-rays detection.

3.2. Pulse height spectrum and light yield

The pulse height spectrum of the grown $TlCaCl_3$ crystal is shown in Fig. 3. The full width at half maximum (FWHM) energy resolution is calculated from the photopeak using the Gaussian function. An energy resolution of ~5% (FWHM) is obtained from the photopeak, which is measured at 662 keV γ-rays, as presented in Fig. 3. In Fig. 3, a peak observed below the 662 keV γ-rays full absorption peak is caused by K X-ray escape peak of Tl^+ ion (Kim et al., 2015; Koehler et al., 2012).

The light yield of the $TlCaCl_3$ crystal is calculated by comparing the pulse height spectrum recorded with 662 keV γ-rays irradiation from a ^{137}Cs source with that of LYSO: Ce^{3+} using PMT (Hamamatsu R6233-100) at room temperature. The light yield measurement is performed under similar experimental conditions. A windowless large area avalanche photodiode (LAAPD) (630-70-73-510,

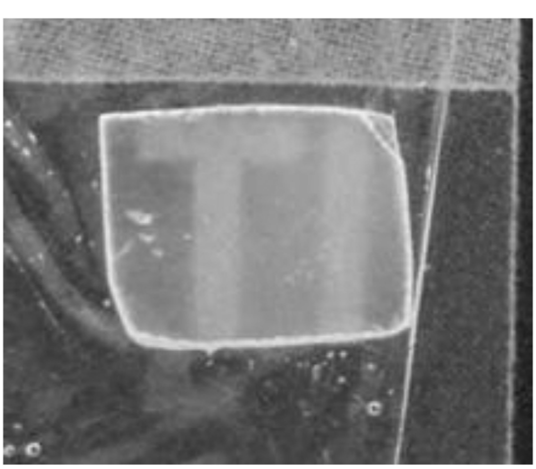

Fig. 1. A polished sample of the as grown $TlCaCl_3$ crystal.

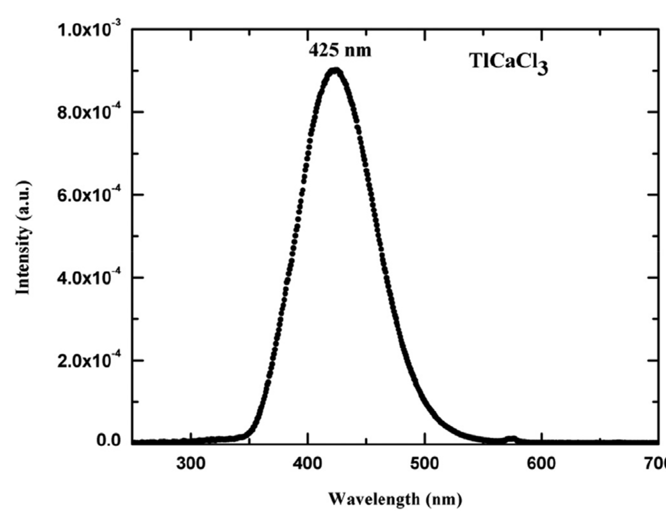

Fig. 2. Emission spectra of $TlCaCl_3$ crystal recorded under x-ray irradiation at room temperature.

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