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Scintillation characterization of thallium-doped lithium iodide crystals

Sajid Khan [a], H.J. Kim [a,*], Y.D. Kim [b]

[a] Department of Physics, Kyungpook National University, Daegu 702-701, South Korea
[b] Center for Underground Physics, Institute for Basic Science (IBS), Daejon 305-811, South Korea

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ABSTRACT

The paper discusses scintillation and luminescence properties of thallium-doped LiI crystals, grown by the Bridgman technique. X-ray induced emission spectrum is obtained between 380 nm and 600 nm, and is attributed to the Tl^+ ion. The photoluminescence measurement with the excitation wavelength of 305 nm revealed a similar emission spectrum. Light yield, energy resolution and scintillation decay time profiles were studied under 662 keV (^{137}Cs) γ-ray excitation. A maximum light yield of 14,000 ± 1400 ph/MeV and two exponential decay time components were obtained.

1. Introduction

Alkali halide scintillators are among the popular materials for scintillation applications and are widely used in the areas such as high energy physics, nuclear physics, medicine, geology, astrophysics and security [1,2]. Among the alkali halides, NaI(Tl) was the first inorganic single crystal scintillator developed by Hofstadter [3]. LiI crystals have attracted attention owing to the presence of ^{6}Li nuclei that have a large capture cross-section for thermal neutrons. LiI crystals have been grown with different activators and tested for scintillation properties. A LiI(Sn) scintillator was used for studying the detection of thermal neutrons [4]. The growth of LiI crystals with Tl and Ag doping were reported in [5,6]. The previous work has been focused on the response of the grown crystals to the thermal neutrons. Activation of LiI phosphor with Eu, Sm and Tl was reported in [7]. The investigations performed in [7] included measuring the β-luminescence and thermoluminescence at room and low temperatures. Among all of the activated LiI crystals, LiI(Eu) is the most intensively studied scintillator for neutron detection and γ-ray spectroscopy. The first Eu-doped LiI crystal and its luminescence properties were proposed by Schenck during early 1950s [8]. Later, Murray [9] reported LiI crystals activated with Eu for detecting neutrons with energies ranging from 1 to 14 MeV, at various temperatures. Recently, Syntfeld et al. [10] reported on the use of LiI(Eu) in neutron and γ-ray spectroscopy. Although LiI(Eu) has reasonable light yield (~15,000 ph/MeV) [10], its slow decay time (1.2 μs) [9] is a limiting factor for high count rate applications.

Despite of the interest in activation of LiI with various dopants, to the best of our knowledge, a detailed study on the crystal growth of LiI with optimal Tl concentration and investigation of its scintillation properties has not been performed. Therefore, for better understanding of the scintillation properties of LiI, more detailed studies on the dependence of these properties on Tl concentration are necessary.

In this paper, we report the crystal growth as well as the scintillation and luminescence properties of LiI(Tl) crystals. Optimal Tl concentration was determined by growing various crystals. Room temperature studies of scintillations properties include: X-ray excited emission spectrum, UV-luminescence, energy resolution, light yield and decay time analysis.

2. Experimental details

2.1. Crystal growth

Single crystals of LiI with various Tl concentrations (0.02%, 0.05%, 0.1%, 0.5% and 1% by mole) were grown by using two zone vertical Bridgman technique [11]. LiI and TlI powders from Alfa Aesar with 4 N and 5 N purity respectively were loaded into quartz ampoules located inside a glove box in Ar atmosphere. The ampoules were sealed under dynamic vacuum of $\sim 10^{-7}$ Torr. The crystals were grown with a speed of 0.5 mm/h. The obtained

* Corresponding author. Tel.: +82 539505323; fax: +82 539561739.
E-mail address: hongjoo@knu.ac.kr (H.J. Kim).

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crystals were crack-free and transparent. The concentrations of Tl in the LiI hosts were not determined after the growth of the crystals. Fig. 1 shows a photograph of the grown crystals in their corresponding quartz ampoules. The samples used in this work were cylinders (10 mm in diameter and 5 mm in height) and were cut from the middle parts of the crystal ingots and polished in Ar environment. Owing to the hygroscopic nature of lithium iodide, the grown crystals were stored in mineral oil.

2.2. Equipment

The photoluminescence excitation and emission spectra of the LiI(Tl) crystals were measured at room temperature by utilizing a Horiba Flurolog-3 (light source-450 W Xenon lamp) [12] in the spectral range of 250 to 800 nm. The X-ray luminescence was measured by using an X-ray tube from DRGEM. Co. having a W anode at room temperature. The power setting parameters of the tube were 80 kV and 1 mA. The emission spectra were acquired by using a fiber optic spectrometer (QE65000, Ocean Optics).

For measuring the pulse height spectra, the samples were wrapped with Teflon tape and directly coupled to the entrance window of a photomultiplier tube (PMT, Hamamatsu R6233) using index matching silicon oil. The measurements were performed at room temperature in the glove box with Ar atmosphere, and samples were excited by using a ^{137}Cs γ-source. The analog signals from the PMT were shaped by using a Tennelec TC-245 spectroscopy amplifier. The output signals were then fed into a 25-MHz flash analog-to-digital converter (FADC) [13]. The FADC output was recorded to a personal computer by using a USB2 connection and the recorded data were analyzed by using a C++ data analysis program [14].

The decay time measurements were performed under 662 keV γ-rays excitation (^{137}Cs-source). The signals from the PMT were fed into a 400-MHz FADC and the decay time spectra were acquired from the recorded pulse shape information. A detailed description of the experimental set-up for measuring the decay time can be found in Ref. [15].

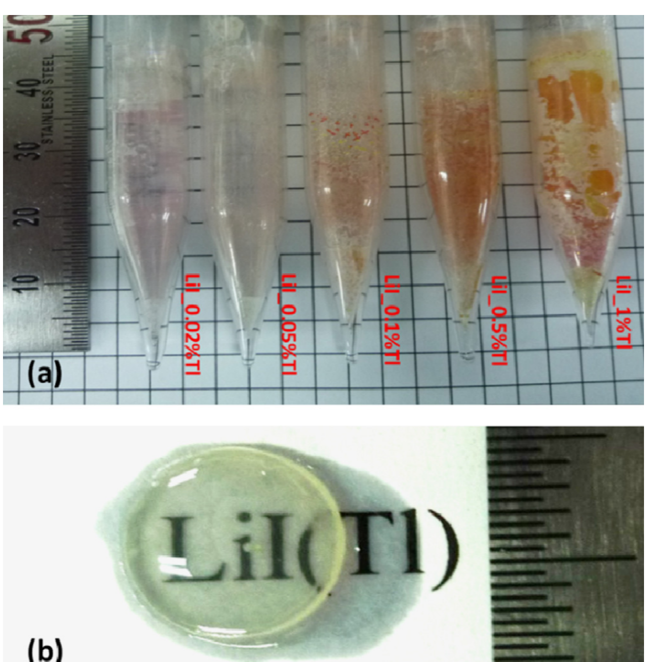

Fig. 1. (a) The LiI crystals with 0.02%, 0.05%, 0.1%, 0.5% and 1% Tl concentration (from left to right) are shown in their quartz ampoules. (b) The LiI(0.5% Tl) crystal, after cutting and polishing (10 mm diameter and 5 mm height). Since LiI is hygroscopic, the shown sample is immersed in mineral oil. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

3. Results and discussion

3.1. X-ray and UV luminescence spectra

X-ray induced emission spectra of the LiI crystals doped with 0.02%, 0.05%, 0.1%, 0.5% and 1% Tl are shown in Fig. 2. Broad emission bands were observed ranging from 380 nm to 600 nm, peaking at 470 nm. The observed emission bands are attributed to the transition between the ground state and excited state of the Tl^{+} ion [16,17]. A similar broad emission spectrum was reported for LiI(0.02% Tl) excited by β particles from ^{90}Sr [18]. A faint shoulder at ~360 nm and a low intensity peak at ~610 nm were also observed in the emission spectra of all crystals. The emission at ~360 nm corresponds to the intrinsic luminescence of LiI and it was earlier reported for pure LiI crystals in [19]. The small peak at ~610 nm was also observed previously in the emission spectra of LiI(0.1% Ag) and LiI(0.04% Yb) [18], but its origin remains unknown.

We found an increase in emission intensity while going to higher Tl concentrations. This behavior has also been observed in other alkali halides scintillators [20,21]. The maximum emission intensity was obtained for the sample with 0.1% Tl and no further increase was observed for the sample doped with 0.5% of thallium. At the Tl concentration of 1% the emission intensity showed its minimum value. The concentration quenching effect [22] might serves as explanation for this observation. Fig. 3 shows the emission intensity as a function of Tl concentration in the host lattice. A small shift in the emission maximum towards the longer wavelengths with increasing Tl concentration was observed. This could be attributed to the re-absorption effects between Tl ions. A similar effect with increasing dopant concentration has also been reported for other scintillating crystals [23,24].

The UV-luminescence spectrum of each LiI(Tl) sample is shown in Fig. 4. All LiI(Tl) crystals were excited at 305 nm and similar broad emission bands were observed between 380 nm and 600 nm. Emission spectra under X-ray and UV excitation showed similar spectral range with typical Tl^{+} emission. Compared with the X-ray induced emission spectra, no bands at ~360 nm and ~610 nm were found in the UV-excitation spectra of the different LiI(Tl) samples.

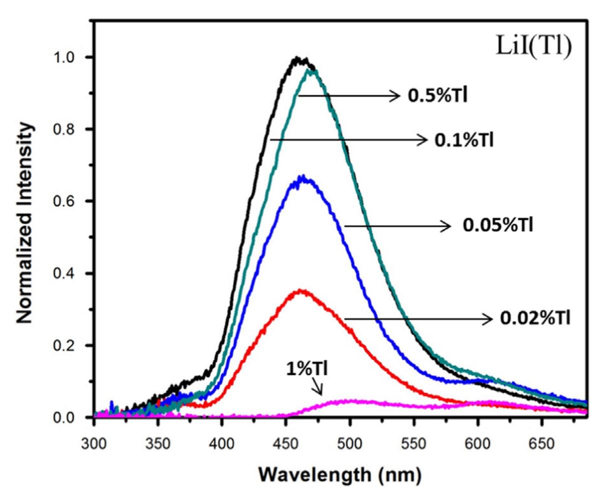

Fig. 2. X-ray induced luminescence spectra of LiI(Tl) crystals with various Tl concentrations, measured at room temperature. The intensity was normalized to the emission intensity of the 0.1% Tl sample. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)