# **Thermoluminescent Glasses for Neutron Detection**

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#### ABSTRACT

We have succeeded in neutron detection via thermoluminescent response of <sup>6</sup>Li- and <sup>10</sup>B- enriched glasses via subtraction of gamma-ray contribution using <sup>11</sup>B-enriched glasses containing Li of natural isotope abundance ratio.

#### 1 Introduction

Neutrons are widely used in various fields of basic science and electric power generation in nuclear plants. In addition, recent progress of boron neutron capture therapy (BNCT) further broadened the application field. Accordingly, demand for neutron detection technique is increasing. Neutrons have no electric charge and are commonly detected via nuclear reactions with several nuclei. Among the nuclei, <sup>3</sup>He has long been used in a proportional counter; however, use of other nuclei is necessary at present owing to serious shortage of <sup>3</sup>He. In solid state detectors, <sup>10</sup>B and <sup>6</sup>Li are widely used.

In this study, we aimed to develop storage phosphors for registering neutron fluence distribution in BNCT. Among the storage phosphors used for radiation detection, thermoluminescence (TL) phosphors were used in the present study because of the simple read out. In addition, glass was chosen as the host of the TL materials owing to the two advantages: one is that glass can contain B and Li as the main constituent, and the other is that many glass systems containing rare-earth ions as luminescent centers have excellent emission properties.

In the case of neutron detection, accompanying gamma rays also contribute to the detection signal. Hence, gamma-ray signal should be discriminated from neutron signal. To enable the discrimination, we used the TL responses of a pair of glasses, one of which contains <sup>6</sup>Li or <sup>10</sup>B as the main constituent and have sensitivity to neutrons and gamma rays, and the other of which contains <sup>7</sup>Li and <sup>11</sup>B as the main constituent and have sensitivity only to gamma rays. In this report, neutron detection capabilities of several TL glass systems based on Tb<sup>3+</sup>-doped Li<sub>2</sub>O<sub>3</sub>–B<sub>2</sub>O<sub>3</sub> [1] and CaO–Al<sub>2</sub>O<sub>3</sub>–B<sub>2</sub>O<sub>3</sub> [2] are introduced.

## 2 Experiments

 $27 Li_2 O - 20 Al_2 O_3 - 50 B_2 O_3 - 1.5 Tb_4 O_7$  and  $Tb^{3+} \text{-doped}$ 

CaO-Al<sub>2</sub>O<sub>3</sub>-B<sub>2</sub>O<sub>3</sub> glasses were synthesized via melt quenching method. Using enriched raw materials, we synthesized four glasses:  ${}^{6}Li{}^{-10}B$ ,  ${}^{6}Li{}^{-11}B$ ,  ${}^{n}Li{}^{-10}B$ , and  ${}^{n}Li{}^{-11}B$  enriched glasses, where  ${}^{n}Li$  denotes Li isotopes with the natural abundance ratio, for 27Li<sub>2</sub>O-20Al<sub>2</sub>O<sub>3</sub>-50B<sub>2</sub>O<sub>3</sub>-1.5Tb<sub>4</sub>O<sub>7</sub> glass. For Tb<sup>3+</sup>-doped CaO-Al<sub>2</sub>O<sub>3</sub>-B<sub>2</sub>O<sub>3</sub> glass,  ${}^{10}B$ - and  ${}^{11}B$ -enriched glasses were synthesized.

The glass samples were irradiated with neutrons using a research reactor UTR-KINKI at Kindai University, Japan, or NASBEE, QST, Japan. After the irradiations, the TL glow curves were obtained using our original apparatus for TL measurements. The samples were placed on a ceramic heater, and the temperature was controlled using a controller. TL photons from the samples were guided to a photon counting unit via an optical fiber.

## 3 Results and Discussion

The photograph of the enriched  $27Li_2O-20AI_2O_3-50B_2O_3-1.5Tb_4O_7$  glass samples is presented in Figure 1. The transparent glass samples were synthesized with strong green emission with UV excitation. Figure 2 presents the TL glow curves of the enriched glass samples after irradiation of neutrons at  $1.0 \times 10^8$  neutrons cm<sup>-2</sup>. A clear glow peak was observed at around 390 K for all samples, among which <sup>6</sup>Li-<sup>11</sup>B enriched sample showed the highest intensity. This result indicates that use of the nuclear reaction of neutrons with <sup>6</sup>Li is advantageous for neutron detection.



Fig. 1. Glass samples of  $27Li_2O-20Al_2O_3-50B_2O_3-1.5Tb_4O_7$  under room light and UV light of 365 nm. Copyright (2021) The Japan Society of Applied Physics [1].



Fig. 2. TL glow curves of enriched glass samples after irradiation of neutrons at  $1.0 \times 10^8$  neutrons cm<sup>-2</sup>. Copyright (2021) The Japan Society of Applied Physics [1].

The difference in the intensities of <sup>n</sup>Li-<sup>11</sup>B, whose TL response is mostly attributed to gamma rays, and the other samples corresponds to the neutron detection signal. The difference in the TL integral intensities at 300–500 K as functions of neutron fluence is described in Figure 2. The difference linearly increased with the neutron fluence. The <sup>6</sup>Li-<sup>11</sup>B enriched sample had the highest integral intensity. Based on the linear fit combined with the noise level of the measurement system, the detectable minimum neutron fluence is estimated to be 4.0×10<sup>6</sup> neutrons cm<sup>-2</sup>.



Fig. 3. Difference in TL integral intensities at 300–500 K with <sup>n</sup>Li-<sup>11</sup>B enriched sample as functions of neutron fluence. Copyright (2021) The Japan Society of Applied Physics [1].

The TL glow curves of 1 mol% Tb3+-doped CaO– Al<sub>2</sub>O<sub>3</sub>–B<sub>2</sub>O<sub>3</sub> glass 2 days after neutron irradiation at 10<sup>11</sup> neutrons cm<sup>-2</sup> are presented in Figure 4. As we expected, the TL intensity of the <sup>10</sup>B glass was much higher than that of <sup>11</sup>B glass, which indicates that both gamma-rays and neutrons contribute to the TL of <sup>10</sup>B glass, while only gamma-rays contribute to the TL of <sup>11</sup>B glass. The difference in TL intensities at 420 and 530 K between <sup>10</sup>B and <sup>11</sup>B glasses as functions of neutron fluence is shown in Figure 6. The difference increases with the neutron fluence, which indicates that the pair of <sup>10</sup>B and <sup>11</sup>B glasses have sensitivity to neutrons within the investigated

neutron fluence range.



Fig. 4. TL glow curves of 1 mol% Tb3+-doped CaO– Al<sub>2</sub>O<sub>3</sub>–B<sub>2</sub>O<sub>3</sub> glass 2 days after neutron irradiation at 10<sup>11</sup> neutrons cm<sup>-2</sup>. Copyright (2019) The Ceramic Society of Japan [2].



Fig. 5 Difference in TL intensities at 420 and 530K between <sup>10</sup>B and <sup>11</sup>B glasses as functions of neutron fluence. Copyright (2019) The Ceramic Society of Japan [2].

#### 4 Conclusions

We have succeeded in neutron detection using TL response of <sup>6</sup>Li- or <sup>10</sup>B- enriched glasses via subtraction of gamma-ray contribution using <sup>11</sup>B-enriched glasses or those containing Li of natural isotope abundance ratio.

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