# **Thermoluminescent Borate Ceramics for Neutron Detection**

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### ABSTRACT

We have succeeded in neutron detection via thermoluminescence response of <sup>10</sup>B- enriched borate ceramics via subtraction of gamma-ray contribution using <sup>11</sup>B-enriched ceramics.

### 1 Introduction

The importance of neutron detection technique is increasing owing to the widespread applications of neutrons. Among them, boron neutron capture therapy (BNCT) is an emerging cancer therapy that uses neutrons. To assure the quality of the therapy, neutron fluence distribution during the therapy should be registered. To enable the registration of the neutron fluence in a simple manner, we developed the measurement system of the neutron fluence on the basis of thermoluminescence (TL), which is luminescence of materials irradiated with ionizing radiation upon subsequent heating.

Neutrons are commonly detected via nuclear reactions with several nuclei: in solid state detectors,  ${}^{6}Li$  or  ${}^{10}B$  are generally used. In particular, based on the high natural abundance (20%) and high cross section of nuclear reaction (3840 barns), we chose  ${}^{10}B$  as the nuclei to be reacted with neutrons. Among compounds containing B as the main constituent, we used borate compounds in this study. Herein, we report the neutron detection capabilities of Tb- [1] and Dy- [2] doped Ca<sub>2</sub>B<sub>2</sub>O<sub>5</sub> ceramics.

#### 2 Experiments

Ceramic samples of Ca<sub>2</sub>B<sub>2</sub>O<sub>5</sub>:Tb and Ca<sub>2</sub>B<sub>2</sub>O<sub>5</sub>:Dy were fabricated in a solid state reaction from the stoichiometric ratio of CaCO<sub>3</sub> (99.99%; Rare Metallic Co., Ltd), H<sub>3</sub>BO<sub>3</sub>, (<sup>10</sup>B-enriched one: <sup>10</sup>B > 96%, Stella-Chemifa Co., <sup>11</sup>Benriched one: <sup>11</sup>B > 99%, Yamanaka Ceradyne, Inc.), Tb<sub>4</sub>O<sub>7</sub> (99.9%, Rare Metallic Co., Ltd.), and Dy<sub>2</sub>O<sub>3</sub> (99.9%; Rare Metallic Co., Ltd.). The concentrations of Tb were 0, 0.5, 1, and 2 mol% relative to Ca. The concentrations of Dy were 0, 0.25, 0.5, 1, 2, and 4 mol% relative to Ca. Using <sup>10</sup>B- and <sup>11</sup>B-enriched H<sub>3</sub>BO<sub>3</sub>, we synthesized <sup>10</sup>B- and <sup>11</sup>B-enriched samples to discriminate the contribution of neutrons and accompanying gamma rays: <sup>11</sup>B-enriched sample is sensitive only to gamma rays, whereas <sup>10</sup>Benriched sample is sensitive to neutrons and gamma rays. The TL glow curves of the samples after irradiations of X-rays or neutrons were measured. The samples were irradiated with X-rays using an X-ray generator (SA-HFM3, Rigaku) equipped with an X-ray tube having Cu target operated at 40 kV and 40 mA. The dose rate was 1 Gy/min. The samples were irradiated with neutrons in the UTR-Kinki nuclear reactor facility at Kindai University. The samples were irradiated at the radiography port at a flux of  $1.0 \times 10^4$  neutrons cm<sup>-2</sup> s<sup>-1</sup> up to a fluence of  $1.0 \times 10^8$  neutrons cm<sup>-2</sup> or at the central stringer irradiation port at a flux of  $1.2 \times 10^7$  neutrons cm<sup>-2</sup> s<sup>-1</sup> at fluences higher than  $1.0 \times 10^8$  neutrons cm<sup>-2</sup>.

#### 3 Results and Discussion

Figure 1 presents the TL intensity after X-ray irradiation as a function of Tb concentration. The highest TL intensity was obtained in the sample with 0.5 mol% Tb. The difference in the TL intensities between <sup>10</sup>B and <sup>11</sup>B ceramics was small. The TL intensity after X-ray irradiation as a function of Dy concentration is presented in Figure 2. The highest TL intensity was obtained at Dy concentration of 4 mol%. At higher Dy concentration, an alien phase was observed in the XRD patterns.



Fig 1. TL intensity as a function of Tb concentration.



Fig. 2. TL intensity as a function of Dy concentration.

The TL glow curves of the sample with 0.5 mol% Tb and 4% Dy are shown in Figures 3 and 4, respectively. TL glow peaks were observed at 340 and 440 K with a shoulder at 500 K for the Tb-doped sample. The glow curve shape of the Dy-doped sample is strongly different from that of the Tb-doped sample. Two glow peaks were observed 350 and 550 K with shoulders at 410 and 625 K. The difference strongly suggests that the trap sites are different in the Tb-and Dy-doped samples.



Fig. 3. TL glow curves of 0.5 mol% Tb-doped sample after X-ray irradiations.



Fig. 4. TL glow curves of 4 mol% Dy-doped sample after X-ray irradiations.

Figure 5 shows the TL glow curves of 0.5 mol% Tb<sup>3+</sup>doped sample after neutron irradiation at 5.0 × 10<sup>7</sup> neutrons cm<sup>-2</sup>. The TL glow curves of 4 mol% Dy<sup>3+</sup>-doped ceramics after neutron irradiation at 1.0 × 10<sup>8</sup> neutrons cm<sup>-2</sup> are presented in Figure 6. For both cases, the <sup>10</sup>Benriched sample had significantly higher TL intensity than that of <sup>11</sup>B-enriched sample, which indicates that we have succeeded in the neutron detection via the TL measurements of a pair of <sup>10</sup>B- and <sup>11</sup>B-enriched samples. The difference in TL intensities of the <sup>10</sup>B- and <sup>11</sup>B-enriche 4 mol% Dy-doped samples as a function of neutron fluence is presented in Figure 7. Excellent linearity in wide dynamic range has been achieved.

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Fig. 5. TL glow curves of 0.5 mol% Tb-doped sample after neutron irradiation at  $5.0 \times 10^7$  neutrons cm<sup>-2</sup>.



Fig. 6. TL glow curves of 4 mol% Dy-doped sample after neutron irradiation at  $1.0 \times 10^8$  neutrons cm<sup>-2</sup>.



Fig. 7. Difference in TL intensities of <sup>10</sup>B- and <sup>11</sup>Benriched 4 mol% Dy-doped samples as a function of neutron fluence.

#### References

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