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# Thermal Neutron Measurements Using Thermoluminescence Phosphor Cr-doped $Al_2O_3$ and Cd Neutron Converter

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In the neutron irradiation field in boron neutron capture therapy, neutron rays and γ-rays are mixed, and it is not easy to selectively measure only neutron rays. The currently used gold activation method is expensive and not suitable for measuring the dose distribution with high spatial resolution. Therefore, there is increasing demand for a simple neutron measurement method for periodic inspections and treatment planning in boron neutron capture therapy (BNCT). Cd has a large nuclear reaction cross section solely for thermal neutrons, and converts thermal neutrons to γ-rays by the (n, γ) reaction. Therefore, in this study, we investigated a method for calculating the thermal neutron fluence by installing Cd as a neutron converter on Cr-doped $Al_2O_3$ ($Al_2O_3$:Cr), which is a thermoluminescence dosimeter, and measuring the converted γ-ray dose with the $Al_2O_3$:Cr. After a 30 min irradiation experiment using the Kyoto University research reactor, the amount of thermoluminescence (TL) increased 101-fold after installing a Cd converter compared with the case of no Cd converter. In addition, the dependence of the TL response on the irradiation time showed excellent proportionality, suggesting the possibility of selectively measuring only the thermal neutron fluence.

## 1. Introduction

In boron neutron capture therapy (BNCT), γ-rays are generated at the same time as neutrons and have different biological effects. Therefore, it is necessary to accurately measure the doses of both when performing quality control of the neutron source and treatment planning for BNCT. Currently, BeO, which is a thermoluminescence dosimeter (TLD) powder,(1) is mainly used for γ-ray measurement in a mixed field of neutron rays and γ-rays, but it is highly toxic to the human body and its production has been discontinued. In addition, although the gold activation method has been used for neutron fluence measurement, gold foil and gold wires are expensive and provide only one-dimensional measurement. The subtraction method, using the difference

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between the cross sections of the nuclear reactions of $^6$Li and $^7$Li for neutrons, is used as a neutron fluence measurement method,[2] but it is not applicable to the dose range of BNCT, so a method that can measure neutron fluence more easily is required. Therefore, we are studying a thermal neutron fluence measurement method using $Al_2O_3$:Cr, which is an inexpensive TLD and capable of two-dimensional imaging.[3]

Previous studies[4] have shown that $Al_2O_3$:Cr has excellent dose responsiveness to photon rays and enables imaging with high spatial resolution but has a small nuclear reaction cross section for neutrons.[5] Therefore, it cannot measure neutron fluence by itself. Therefore, we considered the use of Cd, which is typically employed as a shielding material for thermal neutrons. Cd has a large nuclear reaction cross section solely for neutrons with energy of 0.18 eV or less, absorbs thermal neutrons, and emits γ-rays. Utilizing these properties of Cd, we propose a method to indirectly measure thermal neutron fluence by measuring the dose of γ-rays converted from thermal neutrons by sandwiching $Al_2O_3$:Cr with Cd and irradiating it with neutrons. In this report, the Kyoto University research reactor (KUR) was used to perform irradiation experiments to test this measurement method, and the results are shown.

## 2. Materials and Methods

### 2.1 $Al_2O_3$:Cr and Cd converter

$Al_2O_3$:Cr is $Al_2O_3$ doped with 0.05 wt% $Cr_2O_3$ and has thermoluminescence (TL) characteristics. The effective atomic number of $Al_2O_3$:Cr is 11.13, the density is 3.7 g/cm$^3$, the glow peak temperature is about 300 °C, and the TL wavelength is 693 nm (visible light). In addition, it can be reused after annealing. We measured the TL glow curve with an independently developed measuring instrument.[4] The glow curves were recorded from room temperature up to 400 °C at a heating rate of 0.1 °C $s^{-1}$ in air. The sampling time was 10 s.

In this experiment, we used three 10 × 10 × 0.7 mm$^3$ $Al_2O_3$:Cr TLDs side by side for each irradiation condition (Fig. 1). Since the amount of TL of $Al_2O_3$:Cr is decreased by heat and light, $Al_2O_3$:Cr was covered with black drawing paper to shield it from light and stored at a low temperature (5 °C or less).

Cd (Fig. 2) is a metallic element with atomic number 48 and is used as a thermal neutron absorber in nuclear reactors. Among the isotopes of Cd, $^{113}$Cd, which has a natural abundance ratio of 12.22%,[6] has a large nuclear reaction cross section (20170 barn) for neutrons with energies up to about 0.18 eV (Fig. 3).[7] When $^{113}$Cd is irradiated with thermal neutrons, it induces an (n, γ) reaction and converts thermal neutrons into γ-rays.

In this study, we investigated a method to measure thermal neutron fluence by installing a Cd converter on either side of $Al_2O_3$:Cr and measuring the γ-rays converted from thermal neutrons with the $Al_2O_3$:Cr. In this experiment, we prepared a Cd plate with a purity of 99.99% and processed it into 10 × 30 × 0.5 mm$^3$ pieces so that it was the same size as the three $Al_2O_3$:Cr TLDs. $Al_2O_3$:Cr sandwiched between Cd converters and $Al_2O_3$:Cr without converters were separately irradiated with neutrons using KUR, and the amount of TL was investigated.

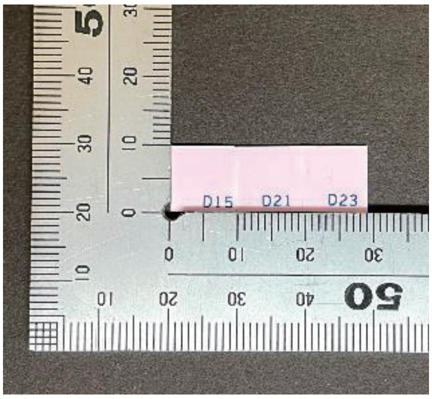


Fig. 1. (Color) $Al_2O_3$:Cr.

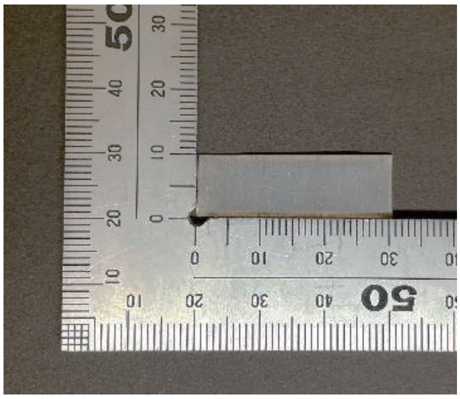

Fig. 2. (Color) Cd converter.

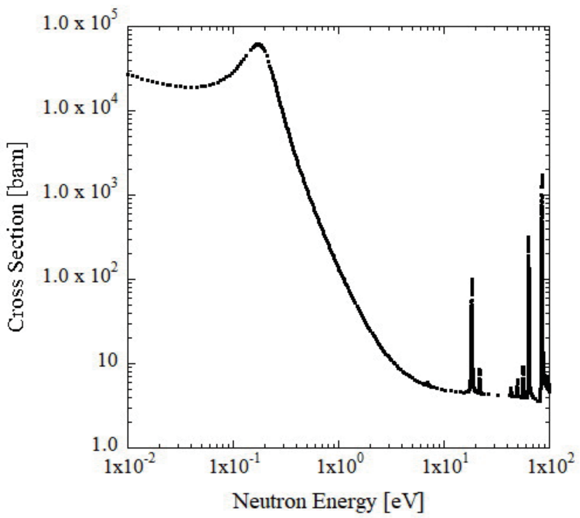


Fig. 3. Nuclear reaction cross section of Cd to neutrons.(7)

## 2.2 Neutron irradiation

KUR uses a plate fuel of about 20% enriched uranium. It has an average thermal neutron flux inside the reactor core of about $3 \times 10^{13}$ [$cm^{-2}s^{-1}$] and a maximum thermal neutron flux at the irradiation position of about $5 \times 10^9$ [$cm^{-2}s^{-1}$].(8) We conducted experiments using the Mix mode (OO-0000), which contains neutron beams of various energies taken from the heavy-water neutron irradiation facility at KUR (KUR-HWNIF). The neutron energy spectrum of KUR-HWNIF in the Mix mode is shown in Fig. 4.(9) $Al_2O_3$:Cr sandwiched between Cd converters was attached to a flat plate and uniformly irradiated with neutron rays. In addition, gold foil and BeO (UD-170LS, Panasonic) were attached to the same plate, and neutron fluence measurement with the gold foil activation method and γ-ray dosimetry using BeO TLDs in the irradiation field were performed. The TL of BeO was measured using a TLD reader (UD-512, Panasonic). The irradiation time was set to 5, 15, 30, 60, and 80 min, and the relationship between the TL response and the irradiation time was also investigated. The Cd converter was removed

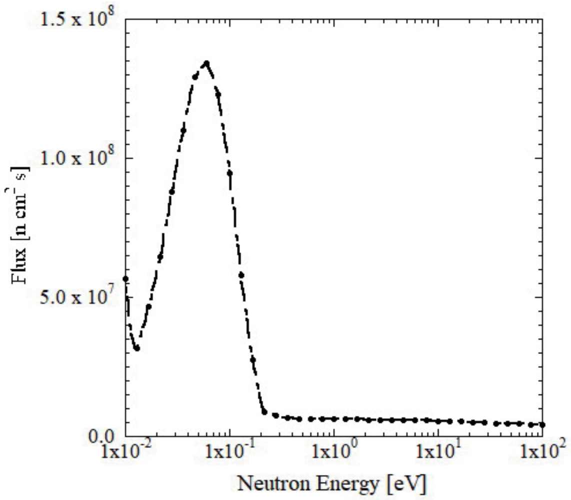


Fig. 4. Neutron energy spectrum focused on thermal and epithermal energy region in the Mix mode of KUR-HWNIF.(9)

immediately after irradiation to eliminate the contribution of radiation emitted by the activation of Cd.

## 3. Results and Discussion

### 3.1 Thermal and epithermal neutron fluence and γ-ray dose in KUR-HWNIF

The results of neutron fluence measurement with the gold foil activation method and γ-ray dosimetry using BeO TLDs for KUR-HWNIF in the Mix mode are shown in Table 1. Thermal and epithermal neutron fluences were calculated using the values measured by the gold radiation method, the Cd ratio data, and the nuclear reaction cross section of Cd. The results showed that the neutron fluence and γ-ray dose increased with the irradiation time, and there was an excellent proportional relationship between them.

### 3.2 TL measurement of $Al_2O_3$:Cr with Cd converter

The TL glow curves(10) of $Al_2O_3$:Cr with the Cd converter (i) and $Al_2O_3$:Cr without the Cd converter (ii) after irradiation for 30 min in the Mix mode of KUR-HWNIF are shown in Fig. 5.

Table 1
Measurement results of neutron fluence and γ-ray dose in Mix mode of KUR-HWNIF.

| Irradiation time (min) | Neutron fluence (cm²) Thermal neutrons | Neutron fluence (cm²) Epithermal neutrons | γ-ray (Gy) (BeO) |
|---|---|---|---|
| 5 | 4.39E+11 | 7.80E+10 | 4.68.E−02 |
| 15 | 1.26E+12 | 2.24E+11 | 1.31.E−01 |
| 30 | 2.48E+12 | 4.40E+11 | 2.57E−01 |
| 60 | 5.55E+12 | 9.88E+11 | 5.44E−01 |
| 80 | 7.30E+12 | 1.30E+12 | 7.21E−01 |

In both cases, the glow peak temperature was about 300 °C. The amount of TL near the glow peak increased about 101-fold after installing the Cd converter. The main reason for this increase in the amount of TL is considered to be that $Al_2O_3$:Cr absorbs γ-rays generated by the (n, γ) reaction of $^{113}$Cd with the internal conversion electrons that are emitted from the excited $^{114}$Cd produced by $^{113}$Cd neutron absorption. This result suggests that the amount of TL derived from thermal neutrons is much larger than that derived from γ-rays, making it possible to selectively measure only thermal neutrons.

In addition, the measurement results for the amount of TL for $Al_2O_3$:Cr with and without the Cd converter and $Al_2O_3$:Cr as a function of the irradiation time are shown in Table 2. In each case, the amount of TL increased with the irradiation time.

The TL response as a function of the irradiation time of $Al_2O_3$:Cr is shown in Fig. 6, and the TL response obtained by subtracting the amount of TL of $Al_2O_3$:Cr without the Cd converter from that of $Al_2O_3$:Cr with the Cd converter is shown in Fig. 7. The figures show that when the Cd converter was installed, the amount of TL of $Al_2O_3$:Cr during irradiation in the mixed mode increased quadratically with the irradiation time. This is due to the superlinearity of $Al_2O_3$:Cr for photon rays of 10 Gy or more.(4) This is because the TL intensity of neutron-derived γ-rays converted by Cd corresponds to that of photon rays of 10 Gy or more. In addition, the TL

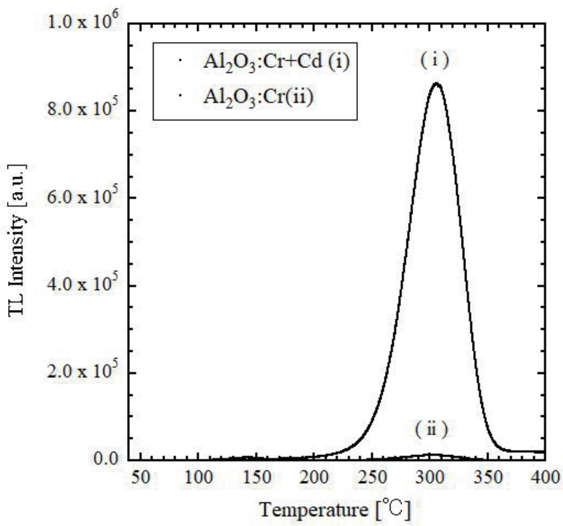


Fig. 5. TL glow curves of $Al_2O_3$:Cr with the Cd converter (i) and $Al_2O_3$:Cr without the Cd converter (ii) after irradiation for 30 min in the KUR-HWNIF Mix mode.

Table 2
Amount of TL of $Al_2O_3$:Cr as a function of irradiation time in the KUR-HWNIF Mix mode.

| Irradiation time (min) | $Al_2O_3$:Cr +Cd (a.u.) | $Al_2O_3$:Cr (a.u.) |
|---|---|---|
| 5 | 3.39E+07 | 4.49E+05 |
| 15 | 1.15E+08 | 1.40E+06 |
| 30 | 3.11E+08 | 3.09E+06 |
| 60 | 8.37E+08 | 6.71E+06 |
| 80 | 1.27E+09 | 8.56E+06 |

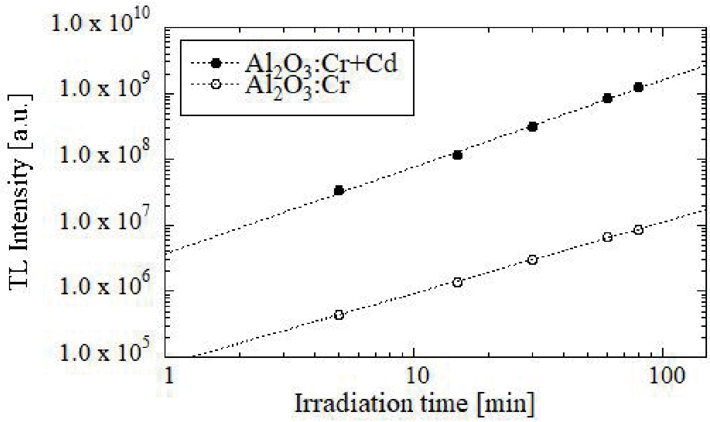


Fig. 6. TL response of $Al_2O_3$:Cr as a function of irradiation time in the Mix mode of KUR-HWNIF (5, 15, 30, 60, and 80 min).

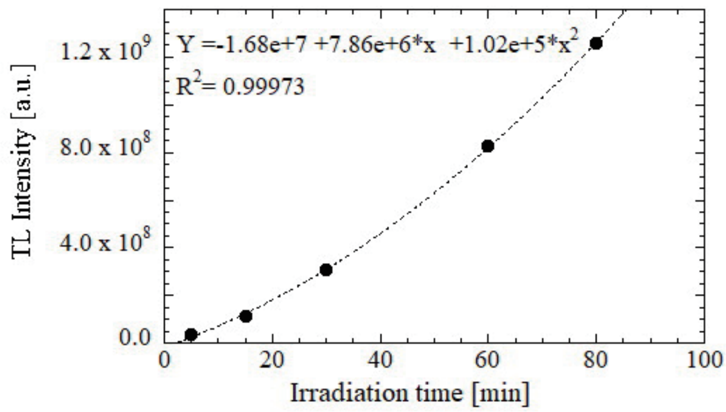


Fig. 7. TL response as a function of irradiation time obtained by subtracting the amount of TL of $Al_2O_3$:Cr without the Cd converter from that of $Al_2O_3$:Cr with the Cd converter.

response curve can be approximated with high accuracy by the quadratic equation in the figure. This suggests that the neutron fluence in the Mix mode of KUR-HWNIF can be obtained from the TL of $Al_2O_3$:Cr using this equation.

## 4. Conclusions

In this study, we irradiated $Al_2O_3$:Cr sandwiched between Cd converters with neutrons using KUR-HWNIF and investigated the amount of TL. From the experimental results obtained using the Mix mode of KUR-HWNIF, it was found that the amount of TL of $Al_2O_3$:Cr was increased after installing the Cd converter. We attributed this increase to the (n, γ) reaction of $^{113}$Cd. After irradiation for 30 min, the TL of $Al_2O_3$:Cr with the Cd converter installed was about 101 times that of $Al_2O_3$:Cr without the Cd converter. This result suggests that the thermal neutron fluence can be calculated easily without the need to subtract the influence of γ-rays. In addition, the TL response as a function of the irradiation time was excellent, suggesting the possibility of performing thermal neutron fluence measurements. Therefore, this measurement system holds promise as a technique capable of selectively measuring only thermal neutrons. In the future, we will proceed with research on the application of the method to two-dimensional neutron fluence measurement using this measurement system.

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