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Investigation of the Thermally Transferred Optically Stimulated Luminescence Characteristics in BeO Ceramics: Dual Thermal Process and Dependence on Irradiation History

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In this study, we investigated the thermally transferred optically stimulated luminescence (TT-OSL) characteristics of BeO ceramic plates with and without annealing treatment, focusing on the effects of preheating temperature and irradiation history. OSL and TT-OSL measurements were performed after X-ray irradiation at doses of 1, 5, and 10 Gy. The OSL intensity reached a maximum at a preheating temperature of approximately 250 ℃, showing a significant enhancement particularly in the low-dose region. This enhancement was attributed to the thermal transfer of electrons from deep traps to shallow optically active traps. Even after annealing at 1000 ℃, which completely erased the irradiation history, TT-OSL signals were observed, indicating that deep traps were reoccupied upon reirradiation and thermally activated during preheating. The results revealed that the TT-OSL characteristics of BeO ceramics are governed by a dual process: the thermal transition of electrons from deep traps to shallow optically active traps and the thermal depletion of initially active traps. Because this dual process depends strongly on the irradiation history, it significantly affects measurement accuracy and sensitivity. Therefore, proper annealing and thermal management are essential for achieving stable and precise dose measurements using BeO-based OSL dosimeters.

1. Introduction

Beryllium oxide (BeO) has long been used as an inorganic material capable of storing radiation energy,(1–4) and BeO emits luminescence when stimulated optically or thermally after exposure to ionizing radiation. These phenomena are referred to as optically stimulated luminescence (OSL) and thermally stimulated luminescence (TSL), respectively. In BeO, electrons and holes generated by ionizing radiation are trapped at defect sites or impurity levels in the crystal, and luminescence occurs when they recombine upon stimulation. Because the luminescence intensity is proportional to the absorbed dose, BeO can be utilized as a dosimetric material.

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The accurate measurement of radiation dose is essential in various fields, such as radiation therapy and environmental monitoring. In recent years, OSL dosimetry using BeO, which exhibits high tissue equivalence, has attracted increasing attention. However, OSL signals strongly depend on the trap structure and defect states of the material, leading to problems such as sensitivity variation, fading, and nonlinear dose response. To address these issues, many studies have focused on the fundamental OSL characteristics of BeO and on improving measurement techniques aimed at enhancing and stabilizing OSL sensitivity.(1–9)

Among OSL dosimeters, α-Al_2O_3:C has excellent thermal and chemical stabilities and has been the most widely used material.(7,10–12) However, its high effective atomic number results in poor tissue equivalence, and saturation and fading occur at high doses. In contrast, BeO, with an effective atomic number of 7.1, close to that of soft tissue (water), and excellent thermal and chemical stabilities, is regarded as a promising material that combines tissue equivalence and high sensitivity. While the TSL properties of BeO have been investigated for decades,(1–3) recent studies have focused on its OSL and thermally transferred OSL (TT-OSL) characteristics,(8,9,13–15) suggesting the potential for further enhancement of luminescence sensitivity.

Yukihara reported strong TT-OSL signals in BeO ceramics and demonstrated that charge transfer from deep traps to shallow optically active traps during heating contributes to luminescence.(8,9) This phenomenon represents a novel sensitivity regeneration mechanism in which electrons that are not released by optical stimulation are thermally redistributed and subsequently contribute to OSL emission. However, the quantitative analysis of these processes requires the precise evaluation of trap structures and transition probabilities, as well as the suppression of signal fluctuations caused by factors such as heating rate, stimulation intensity, and pretreatment temperature.

Okada *et al.* developed a TSL–OSL–RPL Automatic and Integrated Measurement System (TORAIMS) to evaluate these luminescence characteristics under identical experimental conditions.(5) This system eliminates the need for sensitivity correction between different stimulation modes and the positional adjustment of samples, enabling the acquisition of highly reproducible luminescence data.

Previous studies(5–15) have shown that preheating at approximately 250 ℃ is most effective for enhancing TT-OSL sensitivity,(8,9) but these measurements were conducted either without annealing or under annealing conditions below 400 ℃. Therefore, the effect of residual electrons in deep traps may not have been completely eliminated.

In this study, the TT-OSL characteristics of BeO ceramic plates annealed at 1000 ℃ after OSL measurement were evaluated, with the aim of clarifying the effects of preheating on OSL sensitivity and dose–response behavior.

2. Materials and Methods

2.1 BeO ceramic plates

The BeO ceramic plates (Materion Corporation, Thermalox 995) used in this study (Fig. 1) are OSL detector media containing more than 99.5 wt% BeO. Each plate has dimensions of

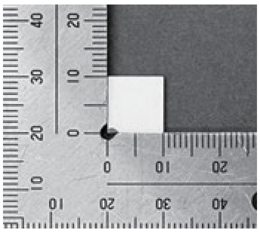


Fig. 1. BeO ceramic plate.

10 × 10 × 0.7 mm^3, a density of 2.85 g/cm^3, and an effective atomic number of 7.13. The chemical composition of the BeO ceramic plates is summarized in Table 1.

2.2 X-ray irradiation and OSL measurement

OSL measurements were performed using the TORAIMS developed by Okada *et al.*[5] This system enables a fully automated operation of all measurement processes, including irradiation, preheating, cooling, and luminescence measurement, under programmable control. A schematic diagram of the TORAIMS configuration and its internal structure is shown in Fig. 2.

X-ray irradiation was carried out using an X-ray generator (Model XRB80N, Spellman High Voltage Electronics, USA). BeO ceramic plates were irradiated with 40 kV X-rays inside a shielded cabinet. All irradiations were performed at a constant rate of 8.33 mGy s^{-1}, corresponding to 120 s Gy^{-1}, for all dose levels. For OSL stimulation, a blue LED module (center wavelength: 450 nm, Thorlabs Inc., USA) was used as the excitation light source. The emitted OSL signals were detected using a photomultiplier tube (Model H11890-01, Hamamatsu Photonics, Japan). To eliminate scattered excitation light, a band-pass filter (center wavelength 340 ± 25 nm, Asahi Spectra Co., Ltd., Japan) was placed in front of the detector.

2.3 Experimental procedures

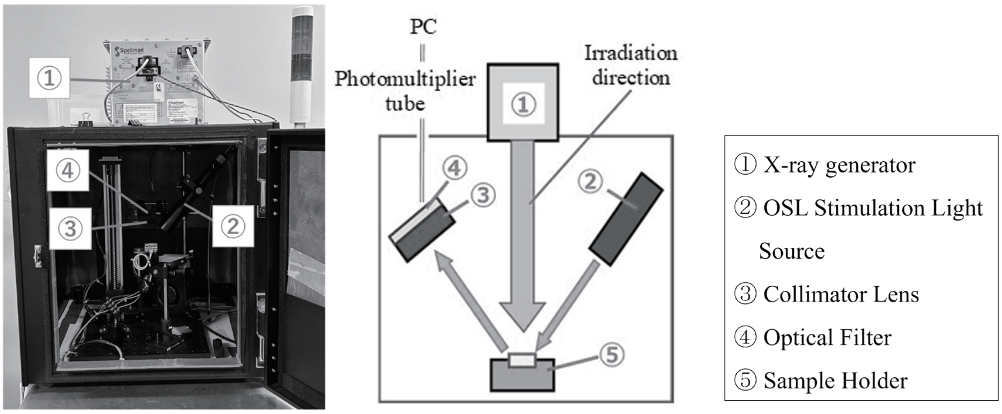
In this study, two types of experiment were performed to investigate the effect of annealing treatment on the TT-OSL characteristics of BeO ceramic plates.

2.3.1 Experiment 1: TT-OSL characteristics of non-annealed samples

The experimental flow chart is shown in Fig. 3, and the measurement conditions are summarized in Table 2. To verify the enhancement of OSL intensity due to preheating reported in previous studies,[5,9] X-ray doses of 1, 5, and 10 Gy were used. For each dose, the following sequence was repeated: X-ray irradiation → preheating → cooling → OSL measurement. The preheating temperature was increased from 100 to 400 °C in 50 °C increments. Each preheating step was maintained for 100 s after the target temperature was reached, followed by cooling to 40 °C before OSL measurement. The first measurement at each dose was performed without

Table 1
Composition of BeO ceramic plates.

Composition	BeO	Si	Mg	Mn
wt%	99.5	0.186	0.0922	0.0002



(a) (b)

Fig. 2. TORAIMS configuration and setup. (a) TORAIMS system and (b) its internal structure.

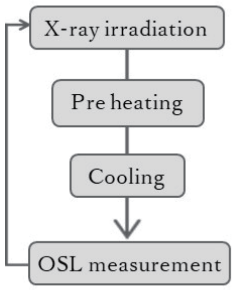


Fig. 3. Measurement flow chart for Experiment 1.

Table 2
Conditions for Experiment 1.

Dose	Preheating temperature	Cooling	OSL measurement time
1, 5, 10	–	No	1000
	100	Yes	
	150		
	200		
	250		
	300		
	350		
	400		

preheating and used as a reference. The stimulation time for OSL measurement was fixed at 1000 s for all experiments.

2.3.2 Experiment 2: TT-OSL characteristics after annealing at 1000 ℃

The measurement conditions are listed in Table 3. BeO ceramic plates were annealed at 1000 ℃ for 30 min to completely erase the irradiation history, and the TT-OSL characteristics were evaluated under X-ray doses of 1, 5, and 10 Gy.

The following four measurement procedures were compared: (1) OSL measurement, (2) consecutive OSL measurements (two successive OSL readings), (3) TT-OSL measurement (with preheating at 250 ℃), and (4) OSL measurement followed by TT-OSL measurement.

On the basis of the results of Experiment 1, the preheating temperature was fixed at 250 ℃, where the TT-OSL intensity was found to be the highest, and the preheating duration was set to 100 s.

Measurements (3) and (4) involving preheating were conducted following the same flow chart shown in Fig. 3. All operations, except during heating, were performed at 40 ℃. For each measurement, the OSL stimulation and acquisition time was fixed at 1000 s. Except between procedures (1) and (2), each experiment was followed by annealing at 1000 ℃ for 30 min to remove any residual irradiation history before proceeding to the next measurement. In procedure (4), OSL and TT-OSL measurements were performed consecutively without intermediate annealing to obtain the cumulative luminescence intensity.

2.4 Annealing

Annealing treatments after each measurement in Experiment 2 were carried out using an electric furnace (Model ETSS-430, Yamada Denki, Japan). The annealing condition was set at 1000 ℃ for 30 min. This temperature is sufficiently lower than the sintering temperature of BeO ceramics (approximately 1200 ℃), allowing heating without damaging the crystal structure. In addition, the temperature and duration were consistent with those reported by Yukihara(8,9) for completely releasing trapped electrons in deep traps and eliminating the irradiation history in BeO. Therefore, this annealing condition was suitable for restoring the BeO samples to their initial state before subsequent measurements.

Table 3
Conditions for Experiment 2.

Dose (Gy)	Measurement procedures	OSL measurement time (s)
1, 5, 10	1. **Annealing (1000 ℃ 30 min)**	–
	2. X-ray irradiation (8.33 mGy/s)	–
	3. OSL measurement 1st	1000
	4. OSL measurement 2nd	1000
	5. **Annealing (1000 ℃ 30 min)**	–
	6. X-ray irradiation (8.33 mGy/s)	–
	7. TT-OSL preheating at 250 ℃	1000
	8. **Annealing (1000 ℃ 30 min)**	–
	9. X-ray irradiation (8.33 mGy/s)	–
	10. OSL measurement + TT-OSL preheating at 250 ℃	1000

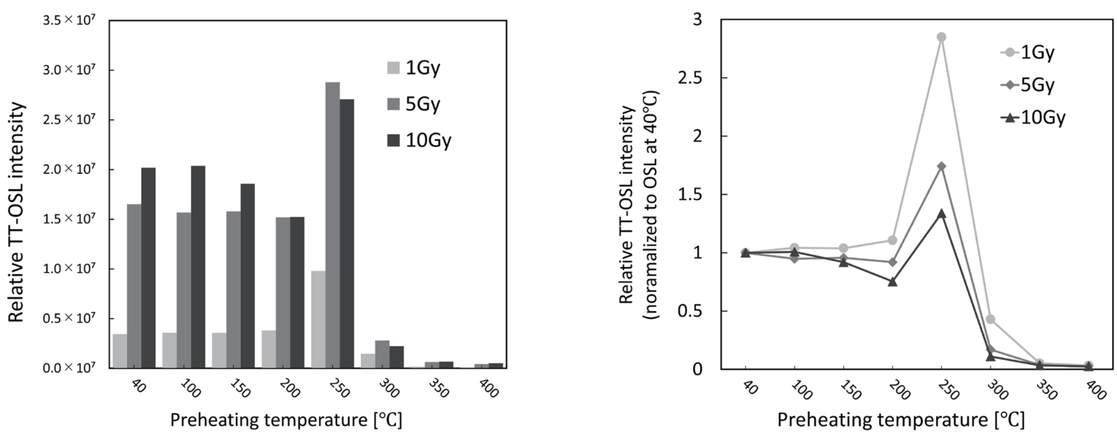
3. Results

3.1 Experiment 1: TT-OSL characteristics of non-annealed samples

The results of Experiment 1 are shown in Fig. 4. For all X-ray doses, the relative TT-OSL intensity—defined as the ratio of the TT-OSL signal (with preheating) to the normal OSL signal (without preheating)—reached its maximum at a preheating temperature of 250 ℃. This result is consistent with those reported by Okada *et al.*[5] and Yukihara.[9] When the preheating temperature was varied between 100 and 400 ℃, the relative TT-OSL intensity decreased above 250 ℃. This suggests that the activation of trap levels contributing to OSL emission occurs most efficiently around 250 ℃. The enhancement ratio of TT-OSL intensity compared with normal OSL intensively was determined to be approximately 2.8 at 1 Gy, 1.7 at 5 Gy, and 1.3 at 10 Gy, indicating that the preheating-induced sensitivity enhancement is more pronounced at lower dose regions.

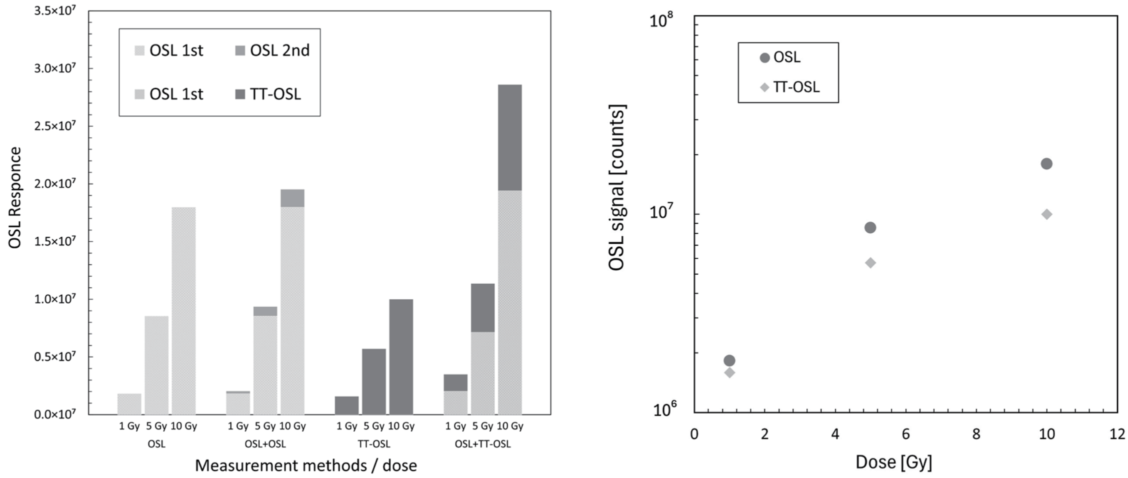
3.2 Experiment 2: TT-OSL characteristics of BeO ceramics after annealing at 1000 ℃

The results of Experiment 2 are shown in Fig. 5. After annealing at 1000 ℃, the OSL intensity obtained from TT-OSL measurements with preheating at 250 ℃ exhibited a decreasing trend compared with that of the non-annealed samples in Experiment 1. At all doses (1, 5, and 10 Gy), the OSL intensity after preheating was lower than that observed during normal OSL measurement. This indicates that the annealing treatment released almost all electrons trapped in deep levels, thereby reducing the possibility of thermal redistribution during subsequent preheating. In other words, annealing at 1000 ℃ likely suppressed the sensitivity regeneration mechanism caused by preheating. Such a decrease in TT-OSL intensity and suppression of charge redistribution after high-temperature annealing have not been clarified in the previous reports by Okada *et al.*[5] and Yukihara.[9]



(a) (b)

Fig. 4. Effects of preheating temperature on TT-OSL intensity and dose (X-ray doses of 1, 5, and 10 Gy) response of BeO ceramic plates. (a) TT-OSL intensity as a function of preheating temperature. (b) Relative TT-OSL intensity, expressed as the ratio of TT-OSL to normal OSL (40 ℃ preheating).



(a) (b)

Fig. 5. TT-OSL characteristics of BeO ceramic plates after annealing at 1000 °C. (a) Comparison of OSL and TT-OSL intensities obtained with different measurement methods after annealing. (b) Dose-response curves of OSL and TT-OSL.

In contrast, a comparison between procedure (2) (two consecutive OSL measurements) and procedure (4) (OSL measurement followed by TT-OSL measurement) showed that the second OSL intensity increased in the latter case. Under the present conditions, a single 1000-s OSL read did not completely deplete the optically active traps. The residual OSL observed in the subsequent read was about 10% of the initial signal, suggesting the partial thermal transfer of electrons from optically inactive traps into optically active levels during the interval between the two consecutive measurements. This increase became more pronounced with increasing dose, with the largest enhancement observed at 10 Gy. These results suggest that, even after the irradiation history was erased by annealing at 1000 °C, X-ray irradiation caused electrons to be trapped again in both shallow and deep levels.

During normal OSL measurement, electrons trapped in shallow levels are released by optical stimulation and contribute to luminescence, whereas those trapped in deeper levels are not easily released. When preheating is applied, electrons in deep traps are thermally transferred to shallow, optically active levels, resulting in additional OSL emission (TT-OSL). Therefore, the second OSL intensity increased when preheating was applied, and this increase was particularly evident at higher doses.

As shown in Fig. 5(b), the comparison of the dose-response characteristics of OSL and TT-OSL revealed that the TT-OSL signal tends to lose linearity at doses higher than those in the case of the OSL signal. This result agrees with the report by Yukihara[9] and suggests that the nonlinear response of the TT-OSL component reflects the contribution of deep-trap levels in BeO.

4. Discussion

On the basis of the above results, Fig. 6 schematically illustrates the luminescence mechanism of TT-OSL in BeO ceramics. The decrease in TT-OSL signal observed after annealing suggests

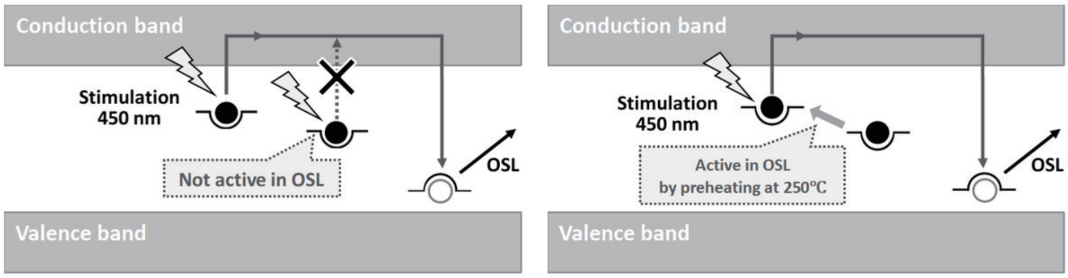


Fig. 6. Schematic energy-band model illustrating the TT-OSL mechanism in BeO ceramics.

that thermal transitions from deep traps to shallow optically active traps were suppressed. The details of this mechanism are discussed below.

In this study, the OSL intensity reached its maximum at a preheating temperature of approximately 250 ℃, and a pronounced enhancement effect was observed, particularly in the low-dose region. These results are consistent with the findings of Yukihara,(8,9) indicating that TT-OSL emission is induced by the thermal transfer of electrons from deep traps. When BeO is irradiated with ionizing radiation, electrons are captured in both shallow and deep traps. Electrons in shallow traps recombine under optical stimulation, producing normal OSL, whereas those in deep traps are not easily released by light alone. During preheating, electrons in deep traps thermally migrate to shallower, optically active levels and can again contribute to luminescence upon optical stimulation. This process constitutes the fundamental mechanism of TT-OSL.

The observation of TT-OSL signals even after annealing at 1000 ℃ indicates that, although the irradiation history was erased, deep traps were reoccupied upon reirradiation and subsequently activated during preheating. In particular, in the combined OSL + TT-OSL measurements, the second luminescence intensity increased, and this increase became more pronounced at higher doses. These results suggest that the occupancy of deep traps depends on the irradiation dose, and that the thermal redistribution process strongly affects the degree of sensitivity enhancement.

Furthermore, in this study, we revealed that two competing phenomena occur simultaneously during preheating: (1) the thermally induced depletion of shallow traps that are originally optically active and (2) the thermal activation of deeper, optically inactive traps that transfer electrons to shallow active levels.

The relative contribution of these two processes depends strongly on the irradiation history and the distribution of trapped electrons. Annealing at 1000 ℃ resets the trap population by emptying optically inactive traps. Consequently, during preheating, the observed TT-OSL signal primarily reflects the thermally induced transfer of electrons—generated by the new single irradiation—from optically inactive (deep) traps into optically active (shallow) levels, allowing the evaluation of this transfer process with a minimal effect from the prior irradiation history. Reproducibility tests confirmed that repeated OSL measurements after each annealing at 1000 ℃ (30 min) showed a variation of less than 1%, and no notable change in TL glow curves was observed. These results indicate that annealing at 1000 ℃ does not alter the luminescence properties of BeO ceramics while effectively removing the effect of the prior irradiation history. Therefore, the enhancement of TT-OSL sensitivity is likely to depend on the irradiation history, making it a key factor affecting the reproducibility and stability of dose measurements.

Even when BeO is used as a conventional OSL dosimeter, the effect of TT-OSL cannot be neglected. An incomplete annealing or residual irradiation history may cause partial thermal contributions from deep traps to be mixed into the measured OSL signal, leading to systematic errors in the estimated dose. Thus, the uniform control of annealing temperature is critically important for ensuring the accuracy of BeO-based OSL dosimetry.

In this study, we experimentally demonstrated that the TT-OSL characteristics of BeO ceramics are governed by a dual process: the thermal transition of electrons from deep traps to shallow optically active traps and the thermal depletion of active shallow traps. These findings provide fundamental insights not only for utilizing TT-OSL phenomena to achieve high-sensitivity dosimetry, but also for stabilizing the sensitivity and calibration of conventional OSL dosimeters.

Future work focusing on the quantitative evaluation of trap energy structures and transition probabilities is expected to further optimize TT-OSL signals and contribute to the development of highly accurate BeO-based dosimeters.

During preheating, electrons trapped in deep inactive levels are thermally transferred to shallower optically active traps, while some electrons in initially active traps are thermally released. These competing processes lead to both the enhancement and depletion of OSL intensity, depending on the irradiation history and annealing condition.

5. Conclusions

In this study, the thermally transferred optically stimulated luminescence (TT-OSL) characteristics of BeO ceramic plates were systematically investigated with respect to annealing and preheating conditions. The following conclusions were obtained:

(1) Sensitivity enhancement by preheating: The OSL intensity became maximum at a preheating temperature of approximately 250 ℃, with a pronounced enhancement observed particularly at low doses. This behavior is attributed to the TT-OSL mechanism, in which electrons trapped in deep levels are thermally transferred to shallow, optically active levels and contribute again to OSL emission.

(2) Effect of annealing treatment: Although annealing at 1000 ℃ effectively erased the irradiation history, the enhancement of TT-OSL sensitivity was still observed after reirradiation. This indicates that deep traps were reoccupied during irradiation and subsequently reactivated by thermal stimulation during preheating.

(3) Dual process during preheating: Preheating was found to involve two competing thermal processes: the thermal depletion of shallow traps that are originally optically active and the thermal activation of deep, optically inactive traps that transfer electrons to active shallow levels. The relative contributions of these processes depend on the irradiation history and affect the reproducibility and accuracy of TT-OSL measurements.

(4) Implications for dose measurement and calibration: Both TT-OSL enhancement and OSL sensitivity variation strongly depend on the irradiation history, representing a major source of uncertainty in BeO-based dosimetry. Therefore, when BeO is used as an OSL dosimeter, it is essential to consider the effect of TT-OSL and to carefully control the annealing temperature to ensure accurate and stable dose measurements.

From these results, it was clarified that the TT-OSL characteristics of BeO ceramics are governed by a dual process: the thermal transition of electrons from deep traps to shallow optically active levels and the thermal depletion of active shallow levels.

The findings of this study provide fundamental insights for stabilizing and improving the sensitivity of BeO-based OSL dosimeters and are expected to contribute to the quantitative control of TT-OSL signals and the development of high-sensitivity dosimeters in the future.

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