




## Article

# Radiation Response Properties of Tb-Doped $\text{MgGa}_2\text{O}_4$ Single Crystals

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**Abstract:** Tb-doped  $\text{MgGa}_2\text{O}_4$  single crystals (0.3, 1, 3, and 5%) were synthesized by the floating zone method. The synthesized crystals had a single phase of  $\text{MgGa}_2\text{O}_4$  confirmed by X-ray diffraction and high transparency in the visible wavelength.  $\text{Tb}^{3+}$  acted as a luminescence center in both photoluminescence (PL) and thermally stimulated luminescence (TSL) processes. In the TSL dose response functions, the minimum detectable dose of the crystals was 0.01 mGy, which was comparable with some commercial dosimetric materials. In the X-ray imaging test taken based on TSL, the spatial resolution of the 1% Tb-doped crystal was estimated to be 8.90 LP/mm (56.2  $\mu\text{m}$ ).

**Keywords:** radiation detection; dosimetry; single crystal; antisite defect



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## 1. Introduction

Dosimetric materials are a kind of phosphors for ionizing radiation detection, which have the function of storing absorbed radiation energy via the carrier trapping phenomenon. When ionizing radiation is absorbed by dosimetric material, numerous electrons and holes (carriers) are generated. The carriers are temporally stored at trapping centers and have a metastable state. Then, they are released by receiving external stimulation, for example, heat or light. After the re-excitation of carriers from trapped states, they recombine at the luminescence center with emitting photons; the emission due to heat or light stimulation is called thermally or optically stimulated luminescence (TSL or OSL), respectively [1]. Because the luminescence intensity is proportional to the absorbed dose, TSL and OSL can derive the total amount and distribution of the absorbed dose and are applied to a wide range of applications, including personal and environmental dosimetry [2], radiation therapy [3,4], and medical imaging [5]. The main required properties for dosimetric materials are high luminescence intensity, low fading, high thermal and chemical stability, and wide dynamic range. Until now, many dosimetric materials such as C-doped  $\text{Al}_2\text{O}_3$  [6], Tb-doped  $\text{Mg}_2\text{SiO}_4$  [7], Cu-doped  $\text{Li}_2\text{B}_4\text{O}_7$  [2], Tm-doped  $\text{CaF}_2$  [8], and Eu-doped  $\text{BaFBr}$  [9] have been investigated and commercialized. However, no dosimetric material meets all of the above requirements; therefore, there is still a need to develop novel and effective dosimetric materials.

Although many dosimetric materials have been developed as opaque ceramics or powder form [5,10–14], a single crystal is advantageous in terms of luminescence intensity. A single crystal has higher transparency than ceramics or powder, and emitted photons can be obtained from not only the surface but also from inside the material. Therefore, more luminescence can reach a readout photodetector. On the other hand, fewer defects can be a disadvantage since it means a lower concentration of trapping centers. To use the advantage and solve the disadvantage, spinel-type crystalline materials are attractive. Spinel-type materials, which have a chemical formula of  $\text{AB}_2\text{O}_4$ , are basically composed of tetrahedral  $\text{A}^{2+}$  ions and octahedral  $\text{B}^{3+}$  ions. However, a part of  $\text{A}^{2+}$  and  $\text{B}^{3+}$  ions are

swapped due to cation inversion, and intrinsic antisite defects are generated [15–18]. The antisite defects are useful for trapping centers, and  $AB_2O_4$  single crystals are expected to have both high transparency and high concentration of trapping centers. Up to now, we have reported that some  $MgAl_2O_4$  single crystals doped with rare earths, including Tb, showed good dosimetric properties [19–22]. Analogically,  $MgGa_2O_4$ , which also has a spinel-type structure, may also exhibit good dosimetric properties. Additionally, in radiation-induced luminescence, luminescence efficiency is inversely proportional to the band gap energy ( $E_g$ ) of the host material [23]. Therefore,  $MgGa_2O_4$  ( $E_g = 4.9$  eV), which has smaller  $E_g$  than  $MgAl_2O_4$  ( $E_g = 7.8$  eV), is expected to have higher luminescence efficiency than  $MgAl_2O_4$  in terms of  $E_g$  [24,25]. However, as far as we know, the dosimetric properties of  $MgGa_2O_4$  have been investigated only in ceramic form, and research into their dosimetric properties has been limited to a few studies, such as TSL glow curves at certain doses [26,27]. In this study, we synthesized  $MgGa_2O_4$  single crystals doped with various concentrations of Tb and investigated the photoluminescence and dosimetric properties.

## 2. Experimental Methods

Tb-doped  $MgGa_2O_4$  single crystals were synthesized by the floating zone (FZ) method. The raw materials were MgO (4N, High Purity Chemicals, Saitama, Japan),  $Ga_2O_3$  (4N, Furuuchi Chemical, Tokyo, Japan), and  $Tb_4O_7$  (4N, Furuuchi Chemical, Tokyo, Japan) powders. Tb concentrations were 0.3, 1, 3, and 5% in molar ratios. To compensate for the evaporation of  $Ga_2O_3$ , 30% excess  $Ga_2O_3$  was added. This experimental condition was tested by different excess amounts of  $Ga_2O_3$  from 5 to 40%. The crystals were grown in air atmosphere by using FZ furnace (Canon Machinery Inc., FZD0192, Shiga, Japan) under conditions of 5 mm/h and 10 rpm for pull-down and rotation rate, respectively. Other operations were performed according to a previous study [28].

A part of obtained crystalline rod was cut and polished for measurement. The portions with few cracks were selected by visual inspection and cut manually. A polishing machine (Buehler, MetaServ 250, Lake Bluff, USA) was used for mechanical polishing. Other remaining parts were crushed into powder for X-ray diffraction (XRD) measurement. An X-ray diffractometer (Rigaku, MiniFlex600, Tokyo, Japan) was used to analyze the XRD patterns. A spectrophotometer (Shimadzu, SolidSpec-3700, Kyoto, Japan) was utilized for diffuse transmittance spectrum measurements.

The PL excitation and emission spectra were investigated by a spectrofluorometer (JASCO, FP-8600, Tokyo, Japan), and PL quantum yields were acquired using Quantaaurus-QY (Hamamatsu Photonics, C11347, Shizuoka, Japan). The shortest excitation wavelength of Quantaaurus-QY was 250 nm. The PL decay curves, as well as decay time constants, were evaluated by Quantaaurus- $\tau$  (Hamamatsu Photonics, C11367, Shizuoka, Japan).

In this study, TSL properties were measured as dosimetric properties. As a radiation source, an X-ray generator (Spellman, XRB80P&N200X4550, Hauppauge, USA) was utilized. The measurements were taken immediately after irradiation. The TSL glow curves were studied by a TSL reader (NanoGray Inc., TL-2000, Osaka, Japan) at 1 °C/s in the temperature range of 50–490 °C [29]. The TSL reader was equipped with an optical shortcut filter that cuts photons longer than 520 nm to eliminate signals due to thermal radiation. TSL spectra were measured by using a CCD-based spectrometer (Ocean Optics, QE Pro, Orland, USA) under heating by an electric heater (Sakaguchi E.H Voc, SCR-SHQ-A, Tokyo, Japan) [30]. The TSL dose response functions were obtained from the peak height at 90 °C of the TSL glow curves at each irradiation dose. The irradiation dose range was from 0.01 mGy to 1 Gy, which was calibrated by ionization chamber.

The X-ray imaging test was carried out by our original setup [31]. Pb-based square wave chart (DIAGNOMATIC, Pro-Res RF BarType7, Oksow, Poland) was used as an imaging object. The tested line patterns were 4.50, 6.30, and 8.90 LP/mm (spatial resolution of 111.1, 79.3, and 56.2  $\mu\text{m}$ , respectively). The crystals were irradiated by 10 Gy X-ray through the imaging object and then heated on hot plate (As One, CHO-170AF, Osaka, Japan) at approximately 300 °C. The emission was read by a CCD camera (Bitran Corp., BK-54DUV, Saitama, Japan) located 40 cm away from the heater and the sample.

### 3. Results and Discussion

Figure 1 shows the photograph of the crystals under room light (bottom) and 254 nm UV light (top). All the crystals were visibly transparent and colorless, although some small cracks were observed. Under 254 nm UV light, all the crystals exhibited green luminescence. Figure 2 depicts the XRD patterns of the crystals. The diffraction patterns of all the crystals corresponded to the reference pattern of  $\text{MgGa}_2\text{O}_4$  (COD 9006817) and indicated that obtained crystals had a single phase of  $\text{MgGa}_2\text{O}_4$ . Since no peak shift was observed within the accuracy of our XRD apparatus, Tb would be doped into interstitial [32].

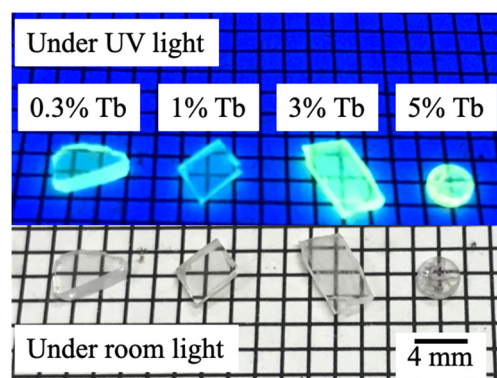


Figure 1. Photograph of the crystals under room light (bottom) and UV light (top).

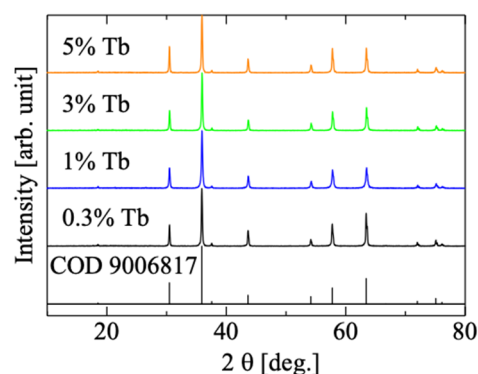


Figure 2. XRD patterns of synthesized Tb-doped  $\text{MgGa}_2\text{O}_4$  and reference (COD 9006817).

Figure 3 displays the diffuse transmittance spectra of the crystals. All the crystals showed absorption at a wavelength shorter than 250 nm and a high transmittance of approximately 80% in the range of 300 to 850 nm. The variation in the transmittance in each crystal may be due to cracks inside the crystals. Absorption at wavelengths shorter than 250 nm was considered due to the overlap of the band gap of the host material and 4f-5d transitions of  $\text{Tb}^{3+}$  [24,33–37].

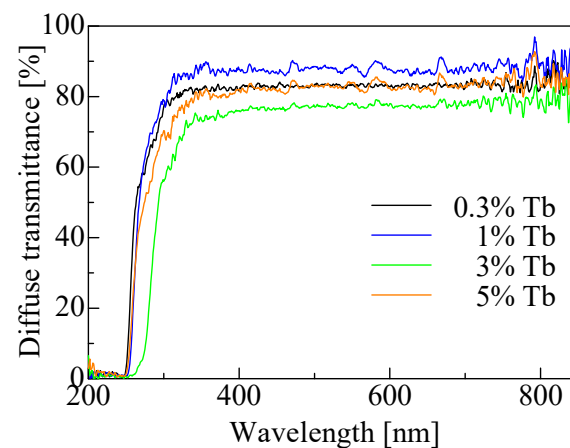


Figure 3. Diffuse transmittance spectra of Tb-doped  $\text{MgGa}_2\text{O}_4$ .

Figure 4 presents the PL excitation (dashed line) and emission (solid line) spectra of the crystals. When the monitoring wavelength was set to 550 nm, an excitation peak was observed at around 230 nm. Since the wavelength of the excitation peak was shorter than the absorption edge observed in Figure 3, the excitation peak might be independent of the absorption edge. Referring to previous studies, the origin of the excitation peak was ascribed to the 4f-5d transitions of  $\text{Tb}^{3+}$  [35–37]. Under excitation at 230 nm, all the crystals exhibited sharp luminescence lines at 380, 420, 440, 460, 470, 490, 550, 590, and 620 nm. According to previous reports, these peaks were attributed to the 4f-4f transitions of  $\text{Tb}^{3+}$  [19,38–41]. The 0.3 and 1% Tb-doped crystals showed a broad emission band around 440 nm as well. The broad emission band was considered due to the oxygen and cation vacancies of the host material [42,43]. The PL QYs of the crystals upon 250 nm excitation are summarized in Table 1. The highest QY was obtained from the 3% Tb-doped crystal among the present crystals, and concentration quenching occurred in the 5% Tb-doped crystal. Our device for QY can allow only excitation wavelengths longer than 250 nm. Therefore, if the crystals are excited by 230 nm, which is a suitable excitation wavelength, a higher QY will be expected.

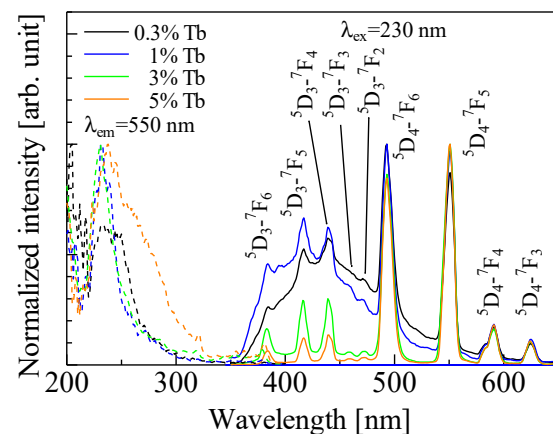


Figure 4. PL excitation (dashed line) and emission (solid line) spectra of Tb-doped  $\text{MgGa}_2\text{O}_4$ .

Table 1. PL QY and decay time constants of the crystals.

Sample	QY at 250 nm (%)	$\tau$ ( $\mu\text{s}$ )
0.3% Tb	5.0	856
1% Tb	6.5	627
3% Tb	10.7	703
5% Tb	4.4	877

Figure 5 shows the PL decay curves of the crystals. Obtained decay time constants are summarized in Table 1 as well as QY. Here, the excitation wavelength was set to 228 nm by using an optical filter, and the monitoring wavelength was 550 nm. All the decay curves were approximated with a single exponential decay function, and the obtained decay time constants were 627–877  $\mu$ s. These values are typical for the 4f-4f transitions of  $\text{Tb}^{3+}$  [44–46].

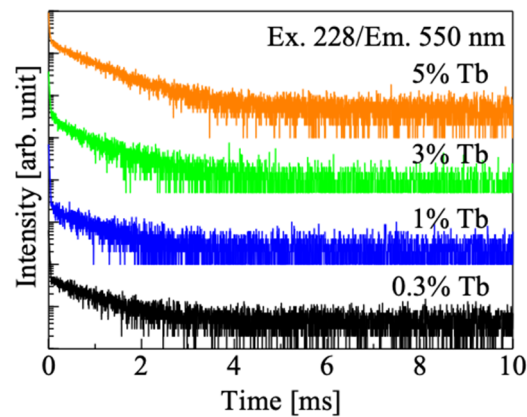


Figure 5. PL decay curves of Tb-doped  $\text{MgGa}_2\text{O}_4$ .

Figure 6 shows the TSL glow curves of the crystals after X-ray irradiation of 100 mGy. All the crystals exhibited a peak around 90  $^{\circ}\text{C}$ . The peak was also observed in powder and ceramic forms of  $\text{MgGa}_2\text{O}_4$ , and the origin was related to intrinsic antisite defects due to cation inversion [27,47]. Thus, antisite defects were useful as the trapping center, even in a single crystal form. Luminescence intensity followed PL QY, and the highest luminescence intensity was confirmed from the 3% Tb-doped crystal. The crystals were stable when heated at 490  $^{\circ}\text{C}$ . Therefore, the crystals can be used stably as a TSL dosimetric material.

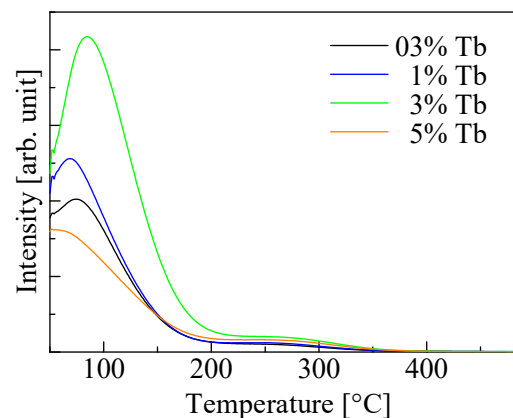
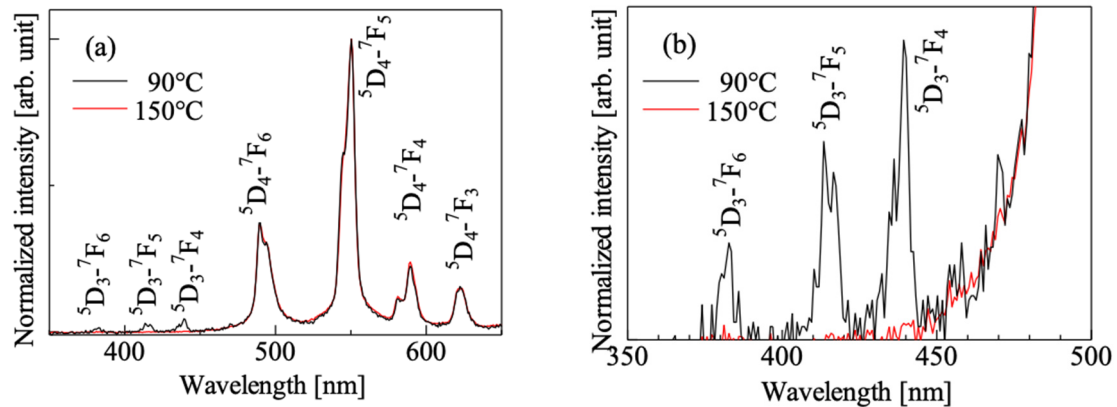


Figure 6. TSL glow curves of Tb-doped  $\text{MgGa}_2\text{O}_4$  after X-ray irradiation of 100 mGy.

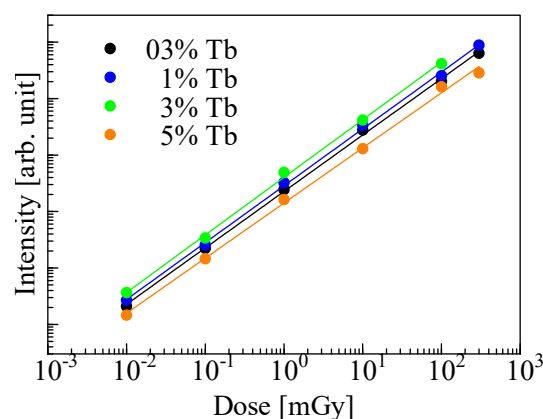
To clarify the luminescence origin of the crystals in TSL, TSL spectra were measured. Here, the result of the 3% Tb-doped crystal, which exhibited the highest TSL intensity, is shown as the representative since all the crystals showed the same trend. Figure 7a indicates the TSL spectra of the 3% Tb-doped crystal stimulated by 90 and 150  $^{\circ}\text{C}$  after X-ray irradiation with 10 Gy. Figure 7b shows an enlarged view in the range of 350 to 500 nm. The emission peaks due to 4f-4f transitions of  $\text{Tb}^{3+}$  were observed from 380 to 620 nm, as well as PL. On the other hand, there was no broad emission band due to oxygen and cation vacancies, unlike in PL. Therefore,  $\text{Tb}^{3+}$  played a major role in the TSL luminescence center, and oxygen and cation vacancies probably acted as trapping centers in TSL together with antisite defects. In addition, as shown in Figure 7b, the emission peaks related to the  $^5\text{D}_3$  level vanished over 150  $^{\circ}\text{C}$  because of thermal quenching [48]. In addition, TSL intensity in

Figure 6 might have been underestimated, especially above 150 °C, because the apparatus for TSL glow curves had an optical filter that cut photons with a wavelength longer than 520 nm.



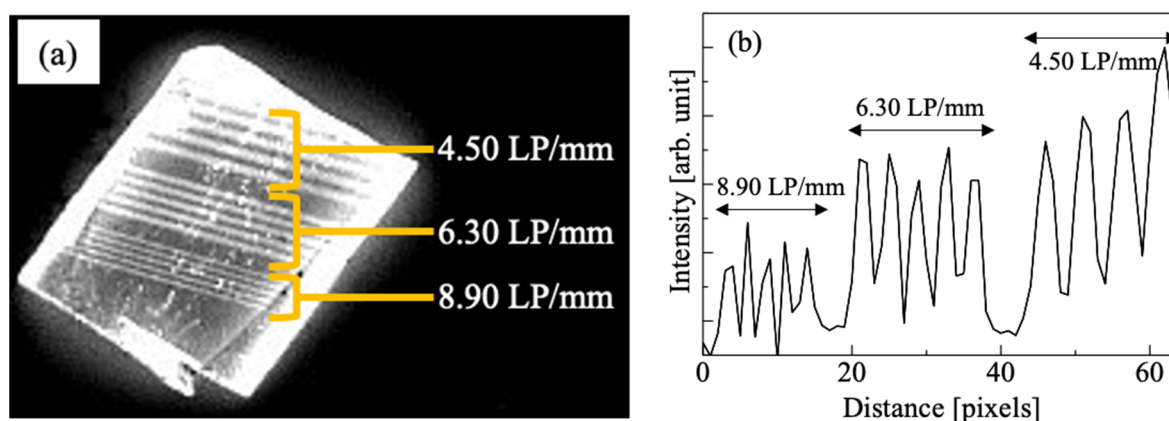
**Figure 7.** (a) TSL spectra of 3% Tb-doped  $\text{MgGa}_2\text{O}_4$  stimulated by 90 and 150 °C after X-ray irradiation with 10 Gy; (b) shows enlarged view between 350 and 500 nm.

Figure 8 presents the TSL dose response functions of the crystals. The intensity was defined as the peak height at 90 °C. The minimum detectable dose of all the crystals was 0.01 mGy, which was comparable with those of some commercial dosimeters, although fading of the samples may be strong [49]. In addition, 0.01 mGy was the lowest dose that could be tested in our setup, and the signal intensity saturated when the irradiated dose exceeded 100 mGy for the 3% Tb-doped crystal and 300 mGy for the other crystals. Therefore, the crystals may be used in a wider dose range.



**Figure 8.** TSL dose response functions of Tb-doped  $\text{MgGa}_2\text{O}_4$ .

Figure 9 shows the results of the X-ray imaging test of the 1% Tb-doped crystal taken by using TSL. Here, the clearest image was obtained from the 1% Tb-doped crystal among the present crystals because of the fewest cracks. Figure 9a,b indicates the obtained image and intensity profiles of each line pattern, respectively. As shown in Figure 9a, luminescence was observed only in the area irradiated by X-rays, while luminescence was not observed in the rest since X-rays were blocked by the Pb substrate. From Figure 9b, the line patterns were well resolved, and the spatial resolution of the 1% Tb-doped crystal was 8.90 LP/mm (56.2  $\mu\text{m}$ ). This result indicates that a Tb-doped  $\text{MgGa}_2\text{O}_4$  single crystal can be applied to 2D dosimetry (TL imaging plate) as well. In addition, the 3% Tb-doped crystal, which showed the highest luminescence intensity, can demonstrate higher spatial resolution than a 1% Tb-doped crystal if a crystal with no cracks is grown.



**Figure 9.** Results of X-ray imaging test of 1% Tb-doped  $\text{MgGa}_2\text{O}_4$  taken by TSL: (a) obtained image; (b) intensity profiles of each line pattern.

#### 4. Conclusion

$\text{MgGa}_2\text{O}_4$  single crystals doped with various concentrations of Tb were synthesized by the FZ method. The XRD patterns showed a single phase of  $\text{MgGa}_2\text{O}_4$ . In PL,  $\text{Tb}^{3+}$  and oxygen and cation vacancies worked as luminescence centers. The highest QY value was obtained in the 3% Tb-doped crystal. In TSL,  $\text{Tb}^{3+}$  acted as a luminescence center, as well as PL; on the other hand, oxygen and cation vacancies played the role of trapping centers, unlike PL. The 3% Tb-doped crystal exhibited the highest TSL intensity, as well as QY. The minimum detectable dose of the crystals was 0.01 mGy, which was comparable with some commercial dosimeters. In the X-ray imaging test using TSL, the spatial resolution of the 1% Tb-doped crystal was estimated to be 8.90 LP/mm (56.2  $\mu\text{m}$ ). Although the TL glow peak at 90  $^\circ\text{C}$  was unstable owing to fading, these results indicate that Tb-doped  $\text{MgGa}_2\text{O}_4$  single crystals are applicable to personal and environmental dosimetry and X-ray imaging.

**Author Contributions:** Conceptualization, Y.T.; methodology, Y.T.; validation, K.N., T.K., D.N. and N.K.; formal analysis, Y.T., S.H., T.K. and T.Y.; investigation, Y.T. and S.H.; resources, T.Y.; data curation, Y.T.; writing-original draft preparation, Y.T.; writing-review and editing, Y.T. and T.Y.; visualization, Y.T.; supervision, K.N., T.K., D.N., N.K. and T.Y.; funding acquisition, T.K., D.N., N.K. and T.Y. All authors have read and agreed to the published version of the manuscript.

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