Dosimeter properties of AlN

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Optical, scintillation, and dosimeter properties of AlN ceramic were reported. AlN sample was SHAPAL, a commercial product of Tokuyama Corp. for mainly a heat sink application. From ultra violet to near infrared wavelengths, it showed 0.02–1% transmittance with visibly translucent appearance. In photoluminescence (PL), it exhibited mainly two emission bands around 340 and 550 nm with primary decay times of 0.92 and 0.24 ns, respectively. X-ray induced radioluminescence spectrum was investigated and 340 nm peak showed high emission intensity. As a dosimeter property, thermally stimulated luminescence (TSL) was studied and glow peaks appeared at 80 and 320°C. The linearity between the irradiated dose and TSL intensity was studied and AlN ceramic exhibited a good linear response from 0.002 to 0.1 Gy.

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1. Introduction

Ionizing radiation detectors using solid state luminescent materials are widely applied to many industrial and scientific fields such as medical,¹⁾ security,²⁾ well-logging,³⁾ environmental monitoring,⁴⁾ personal monitoring,⁵⁾ and high energy physics.^{6),7)} Luminescent materials for radiation detectors are mainly classified as two types: scintillators which convert a single ionizing radiation to thousands of visible photons immediately⁸⁾ and dosimeters which storage the absorbed energy.⁹⁾ By stimulation of light (optically stimulated luminescence, OSL) or heat (thermally stimulated luminescence, TSL), the absorbed energy is released and read out from dosimeter materials. In personal dosimeter applications, ceramic Ti and Mg doped LiF¹⁰⁾ for TSL, crystalline powder C:Al₂O₃ for OSL,⁹⁾ and Ag-doped phosphate glass for radiophotoluminescence (RPL)¹¹⁾ are practically used.

The present work demonstrates basic optical, scintillation and TSL properties of ceramic AlN fabricated by Tokuyama Corp. for a heat sink application, namely SHAPAL. AlN is a wide band-gap material ($E_g = 6.2$ eV) with a wurtzite structure. Generally, ceramic AlN is used for the heat sink and crystal AlN is investigated for deep UV LED¹²⁾ due to a high thermal conductivity of $320 \text{ W m}^{-1} \text{ K}^{-1}$ and a high-electrical resistivity. Although basic TSL properties of AlN were studied few decades ago,^{13),14)} sample AlNs in previous works were not commercialized ones and radiation induced properties were not studied so deeply. Basic studies of commercially available AlN ceramic are important for practical applications so that we investigated SHAPAL which was a product of Tokuyama Corp. Since personal dosimeter applications required tissue-equivalent luminescent materials, AlN is suited due to its relatively low density and effective atomic number close to Al₂O₃ which is one of the famous OSL materials.

2. Experimental procedure

The sample AlN was a product (SHAPAL) of Tokuyama Corp. and the size was $15 \times 15 \times 1 \text{ mm}^3$. Backscattered electron image

was observed by using scanning electron microscope (SEM, Hitachi S-3400). Optical straight line transmittance was studied by using JASCO V670 spectrometer and photoluminescence (PL) emission map was investigated by Hamamatsu Quantaurus-QY. Then, PL decay time monitoring at 340, 505, and 550 nm were evaluated by using Hamamatsu Quantaurus- τ .

X-ray induced radioluminescence (RL) spectrum was measured by utilizing our original setup.^{15),16)} The excitation source was X-ray generator equipped with Cu target (OURSTEX Corporation) and supplied with 80 kV bias voltage and 2 mA tube current. The emission spectra were measured using Andor DU-420-BU2 CCD spectrometer. Its CCD-based detector (cooled down to 188 K by a Peltier module) was coupled with a monochromator SR163 (Andor, 1200 grooves/mm, 300 nm blaze wavelength). The scintillation light was fed into spectrometer through a 2 m optical fiber to avoid direct X-ray hit to CCD.

TSL glow curves at each X-ray irradiated dose were evaluated by using TL-2000 (Nanogray). In this instrument, photons from 300 to 500 nm were accumulated and a heating rate was 1°C/s to correctly determine a glow peak. The X-ray was irradiated by the generator same with the radioluminescence evaluation and X-ray dose was calibrated by the ionization chamber. In order to evaluate TSL spectrum, TL-2000 was improved to equip a port for optical fiber connection to feed TSL to the CCD based spectrometer (Andor DU-420-BU2).

3. Results and discussion

Figure 1 demonstrates an appearance and SEM image of AlN sample. AlN had a gray color and was visibly translucent. From SEM image, typical grain size was few μm . Transmittance spectrum of AlN ceramic is shown in **Fig. 2**. The optical straight line transmittance was 0.2% at wavelength longer than 400 nm and rapidly decreased to 0.001% around the bandgap. The dip around 800 nm was an instrumental artifact. The observed data was consistent with the bandgap ($6.2 \text{ eV} = 200 \text{ nm}$) of AlN. Compared with the recent research about the transmittance of AlN,¹⁷⁾ overall transmittance was not so good.

PL emission map of AlN is exemplified in **Fig. 3**. In PL, two emission bands around 505 and 550 nm appeared and the

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Fig. 1. A photograph (left) and SEM image (right) of AlN ceramic.

Fig. 2. Transmittance of AlN ceramic.

Fig. 3. PL emission map of AlN. The horizontal axis shows an emission and the vertical axis represents an excitation wavelengths.

Fig. 4. PL decay time profiles of AlN. (a) monitoring at 550 nm under 400 nm excitation, (b) monitoring at 340 nm under 280 nm excitation, (c) monitoring at 505 nm under 280 nm excitation, and (d) expanding image of the fast component of (c).

strongest emission band was 550 nm upon 380 nm excitation. In this evaluation, the absolute PL quantum yield of AlN was less than 1%. Up to now, there were many reports about PL of AlN, and the origin of 505 and 550 nm emissions would be related to defects or oxygen impurity.^{18),19)} PL decay time profiles of 340, 505, and 550 nm emission bands are displayed in Fig. 4. The

decay time of 550 nm emission that was the strongest luminescence peak in PL resulted quite fast, 0.24 ns. Then, secondary luminous peak at 505 nm showed two decay time components: one was fast 4.1 ns and the other was slow 6.65 μs. In addition, 340 nm peak only detected in scintillation (discussed later) was investigated and fast decay of 0.92 ns was observed.

Fig. 5. X-ray induced scintillation spectrum of AlN.

Fig. 7. TSL spectra at 80 and 320°C glow peaks of AlN.

Fig. 6. TSL glow curve of AlN after 50 mGy exposure.

X-ray induced scintillation spectrum is shown in **Fig. 5**. Unlike PL spectrum, intense emission band at 340 nm appeared and a sharp peak around 600 nm would be diffraction light of the main emission peak around 340 nm because no other work reported such a long wavelength emission. This intense UV emission was observed in the cathode luminescence spectrum¹⁷⁾ and the origin of this peak was still under discussion.^{20)–22)} When compared with other famous scintillating materials evaluated in the same manner, scintillation of AlN was not so high due to a low X-ray absorption probability and a low emissivity.

Figure 6 depicts TSL glow curve of AlN after 50 mGy exposure. Glow peaks appeared at 80 and 320°C. In the previous work, glow peak below 300°C was reported²³⁾ and the actual glow peak in the past report would be exist at lower temperature because the heating rate was quite high (10°C/s).²³⁾ In addition, glow peak below 100°C was not observed at all. Therefore the glow curve of the present sample AlN was largely different with previous results due to the difference of the fabrication process. By applying the initial rise method²⁴⁾ for 320°C peak, the trap depth of 0.18 eV was deduced. Then, **Fig. 7** compared TSL spectra around 80 and 320°C. The dominant component was the 340 nm luminescence and in higher temperature the peak shifted to longer wavelength. In the past study the dominant emission wavelength of TSL of AlN was around 400 nm²³⁾ and this discrepancy suggested that different TSL origins worked in the present the present AlN. Though the difference of the glow peak temperature was possibly explained by the difference of the heating rate, the difference in the wavelength could not be explained by experimental conditions and further studies would be required.

In **Fig. 8** the dose response curves of AlN in the dose range from 5 to 100 mGy of 80 and 320°C glow peaks are presented. The TSL intensity was obtained by an area of single Gaussian approximation to each peak. Since the 320°C peak was quite

Fig. 8. Dose response functions of AlN using 80 and 320°C glow peaks.

luminous, we could not evaluated at higher dose due to the upper limit of the instrument. A linear dose response function within this dose range confirmed that the mass production AlN (SHAPAL) was applicable for dosimeter application.

4. Conclusion

Optical, scintillation, and dosimeter properties of commercially available AlN ceramic were investigated. In spite of the translucent appearance, ceramic AlN showed intense PL and TSL. Dose response functions at 80 and 320°C glow peaks were linear from 5 to 100 mGy and it was proved that commercially available AlN ceramic can be applicable for dosimeter application.

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