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Thermoluminescence of Laser-Irradiated Mg_2SiO_4:Tb

Tadaki Miyoshi, Takanobu Kobayashi, Taketsugu Shinohara, Koji Okumura and Teruhisa Kaneda[1]

Department of Electrical and Electronic Engineering, Yamaguchi University, Tokiwadai, Ube, Yamaguchi 755-8611, Japan

[1]*Ube National College of Technology, Tokiwadai, Ube, Yamaguchi 755-8555, Japan*

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Thermoluminescence has been observed in laser-irradiated Mg_2SiO_4:Tb. Three glow peaks were observed at 130, 220 and 310°C. The glow peak observed at 220°C has not been previously reported for light-irradiated samples, and is similar to that observed in X-ray irradiated samples. The origin of these glow peaks was investigated.

KEYWORDS: phosphor, thermoluminescence, radiation dosimeter

Thermoluminescence is observed in rare earth-doped phosphors and used for radiation dosimetry.[1] Mg_2SiO_4:Tb is a phosphorus material, which can be applied as a thermoluminescence dosimeter of high sensitivity. This material shows thermoluminescence after light, X-ray and γ-ray irradiation,[2] as shown in Fig. 1. In ref. 2, the light source used was a fluorescent lamp. However, we cannot find any report on the thermoluminescence of this material when irradiated with laser light. Here, we report on the thermoluminescence of Mg_2SiO_4:Tb after laser irradiation. Comparing the glow curves of the thermoluminescence caused by various irradiation sources, we propose a possible mechanism for the thermoluminescence of this material.

The sample used was a commercial thermoluminescence dosimeter (TLD), Kyokko MSO-S (Mg_2SiO_4:Tb). The TLD was exposed to light from a frequency-tripled Nd:YAG laser (Quanta-Ray GCR-230T-10; wavelength = 355 nm, pulse duration = 5 ns, repetition rate = 11 Hz) and a frequency-doubled Ti:sapphire laser with a regenerative amplifier (Spectra Physics Tsunami and Spitfire; wavelength = 400 nm, pulse duration = 130 fs, repletion rate = 1 kHz) at 300 K. The thermoluminescence was measured using a TLD reader (Kyokko 2500). The glow curves were recorded using a pen recorder.

The thermoluminescence measurements were performed by heating the irradiated sample to 500°C. The heating rate was 4 K/s. Figure 2 shows the glow curves of the thermoluminescence of the MSO-S sample after Nd:YAG laser irradiation. Three glow peaks are observed at 130°C, 220°C and 310°C. The glow peaks at 130°C and 310°C are similar to those observed after light irradiation (dashed curve in Fig. 1), and the glow peak at 220°C is similar to that observed after X-ray irradiation (solid curve in Fig. 1). Although the relative intensity of the glow peak at 220°C is weaker than that of the other peaks for low irradiation intensity (dashed curve in Fig. 2), it becomes stronger than that of the other peaks for high irradiation intensity (solid curve in Fig. 2). Figure 3 shows the glow curves of the thermoluminescence of the MSO-S sample after Ti:sapphire laser irradiation. One glow peak is observed at 220°C. This glow peak is similar to that observed after X-ray irradiation (solid curve in Fig. 1).

From the glow curves, we evaluated the location of the traps using the initial rise method.[1] The energy difference between the traps and the valence-band or conductionband edge of glass is 1.1 eV for the glow peak at 220°C, 0.70 eV for the 130°C peak and 1.4 eV for the 310°C peak. The location of the traps corresponding to the glow peak at 310°C was evaluated by the following technique to eliminate the 130°C peak.[1)] The sample was first heated to 280°C at a heating rate of 4 K/s. The sample was then cooled to room temperature and reheated, at the same heating rate, to 500°C.

Two glow peaks at 130°C and 310°C are observed after Nd:YAG laser (355 nm) irradiation and fluorescent lamp irradiation. These peaks are not observed after Ti:sapphire laser (400 nm) irradiation. These peaks have been reported to have been observed after light irradiation, for light wavelengths shorter than 360 nm.[2)] Since the absorption edge of Mg_2SiO_4 is shorter than 285 nm,[3)] light irradiation is not capable of producing electron-hole pairs by direct excitation of electrons from the valence band. However, the ionization of excited impurities is possible. Electrons may be produced from impurities in Mg_2SiO_4, and these electrons could be trapped. Upon heating, these electrons would be released at temperatures reflecting their trap binding energy, and would produce two glow peaks at 130°C and 310°C. Although the impurities responsible for the glow peaks have not been established, an irradiation-induced ionization of impurities has been observed in Al_2O_3:Cr.[4)]

Irradiation-induced valence reduction and thermally induced reoxidation of rare-earth (RE) ions have been observed in rare-earth-doped phosphors.[5)] X-ray or γ-ray irradiation produces both electrons and holes. Electrons are trapped by RE^{3+} which produces RE^{2+}. Holes are trapped by hole traps. If heated, these holes would be released and would recombine with RE^{2+} to yield RE^{3+*}, the resulting excited state, which shows emission. Such an irradiation-induced valence reduction, and the thermally induced reoxidation of Tb^{3+} ions could be responsible for the peak at 220°C observed in Mg_2SiO_4:Tb.

The temperature of the glow peak at 220°C shifts to the lower temperature side, when the Nd:YAG laser irradiation intensity is weak (Fig. 2). This shift is not observed for X-ray irradiation or Ti:sapphire laser irradiation. Therefore, this shift may be due to trapped electrons, which are responsible for the glow peaks at 130°C and 310°C.

Since laser light is intense, both electrons and holes may be produced by two-photon absorption. Consequently, the glow peak at 220°C is observed in the laser-irradiated samples. Figure 4 shows the intensity of thermoluminescence at the glow peaks as a function of the irradiation intensity of Nd:YAG laser light. The thermoluminescence intensity of the glow peak at 220°C (solid circles) increases supralinearly with increasing irradiation intensity. The glow peak thermoluminescence intensity is proportional to $J^{1.3}$, where J is the irradiation intensity. The glow peak thermoluminescence intensity also increases supralinearly for Ti:sapphire laser irradiation. This supports the hypothesis that the glow peak at 220°C is attributable to two-photon absorption. Supralinearity is observed for X-ray irradiation, when the X-ray dose is larger than 1 Gy.[2)] However, the thermoluminescence intensity is 600000 (arb. units) for an X-ray dose of 1 Gy. The intensity of thermoluminescence induced by Nd:YAG laser irradiation is markedly smaller than that induced by X-ray irradiation. Therefore, the origin of the supralinearity shown in Fig. 4 may be different from that of thermoluminescence induced by X-ray irradiation.

The thermoluminescence intensity of the glow peak at 310°C (open triangles) increases sublinearly with increasing irradiation intensity. The glow peak thermoluminescence intensity is proportional to $J^{0.7}$. On the other hand, glow peak thermoluminescence intensity is proportional to J for light irradiation using a fluorescent lamp.[2)] While only electrons may be generated as a result of the fluorescent lamp irradiation, both electrons and holes may be generated as a result of the YAG:laser irradiation. Consequently, electrons recombine with holes during laser irradiation. This recombination may reduce the slope of the curve.

We measured thermoluminescence intensity as a function of irradiation time. The intensities of the glow peaks increased linearly with increasing irradiation time.

When MSO-S was exposed to light from a frequency-doubled Nd:YAG laser (Quanta-Ray GCR-230T-10; wavelength = 532 nm, pulse duration = 5 ns, repetition rate = 11 Hz), thermoluminescence was not observed. This suggests that a photon energy of 532 nm is too low to generate electron-hole pairs by two-photon absorption.

In summary, thermoluminescence was observed in Mg_2SiO_4:Tb after laser irradiation. Three glow peaks were observed. The glow peak at 220°C, which has not been observed in light-irradiated samples, was observed in the laser-irradiated samples. The origin of the glow peak at 220°C is considered to be explained by the electron-hole pairs produced by two-photon absorption, and the other glow peaks by the electrons produced from impurities in the samples.

This work was performed using the laser in the Venture Business Laboratory, Yamaguchi University. The authors are grateful to Associate Professor K. Kasatani for his cooperation in the laser experiments.

References

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Figure captions

Fig. 1. Glow curves of thermoluminescence of TLD, MSO-S (Mg_2SiO_4:Tb), after X-ray irradiation (solid curve) and white-light irradiation (dashed curve). X-ray dose is 1 Gy. Effective X-ray energy is approximately 48 keV. Light intensity is 1500 lx (power density of light is 0.5 mW/cm^2), and irradiation time is 2 h. Light source is a fluorescent lamp. Thermoluminescence intensities at glow peaks are normalized. Peak intensities are 600000 (arb. units) for X-ray irradiation and 110 (arb. units) for light irradiation.

Fig. 2. Glow curves of thermoluminescence of MSO-S after Nd:YAG laser irradiation. Peak power density of laser light is 1.2 MW/cm^2 (solid curve) and 38 kW/cm^2 (dashed curve). Irradiation time is 2 min. Thermoluminescence intensities at glow peaks are normalized. Peak intensities are 2400 (arb. units) for 1.2 MW/cm^2 and 50 (arb. units) for 38 kW/cm^2.

Fig. 3. Glow curves of thermoluminescence of MSO-S after Ti-sapphire laser irradiation. Peak power density of laser light is 400 MW/cm^2 (solid curve) and 100 MW/cm^2 (dashed curve). Irradiation time is 8 min.

Fig. 4. Intensity of thermoluminescence as a function of irradiation intensity of Nd:YAG laser light for MSO-S. Irradiation time is 2 min. Solid circles represent the intensities of glow peak at approximately 220°C, and open triangles those at 310°C. Lines are drawn through the data points as a visual guide.

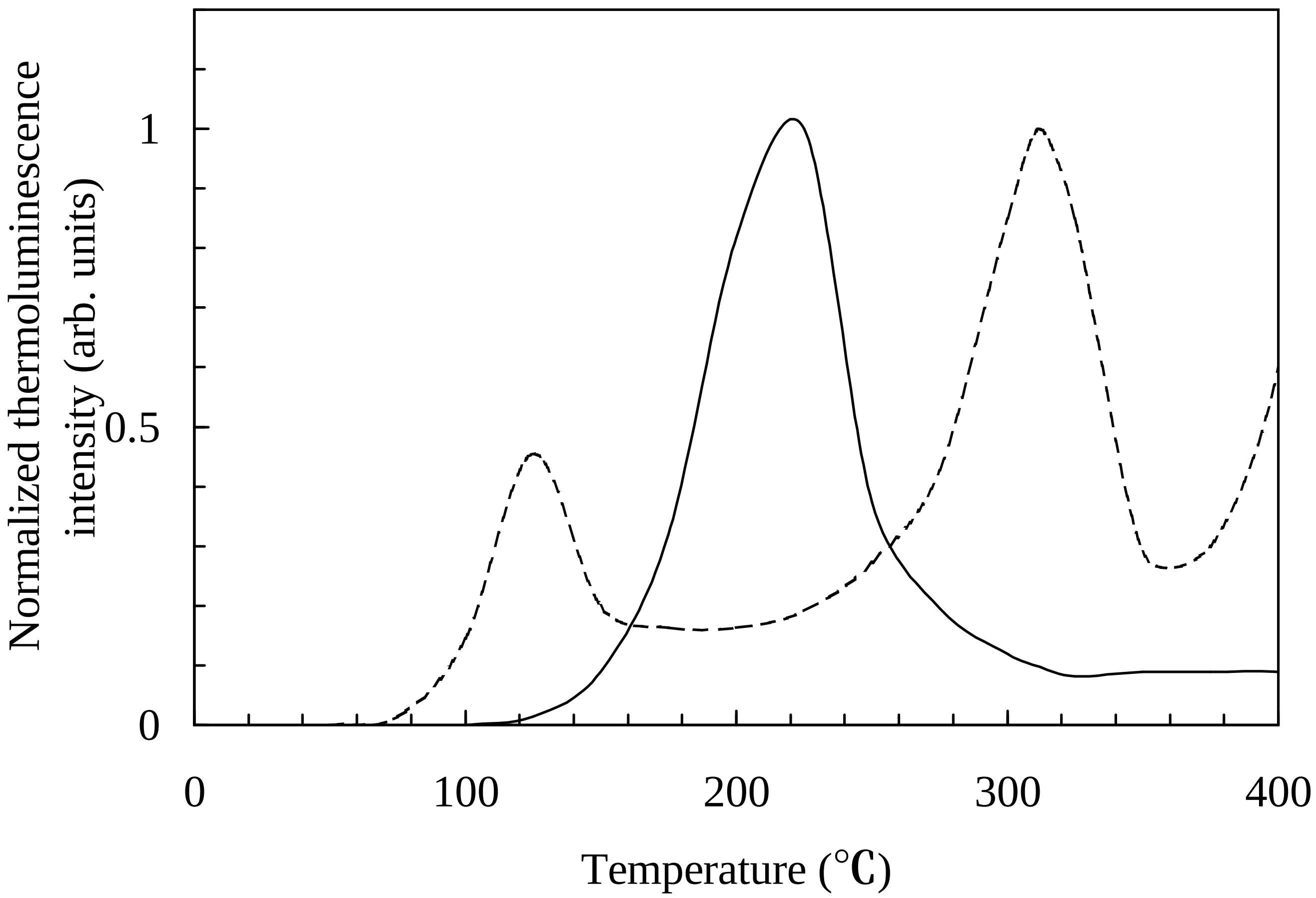
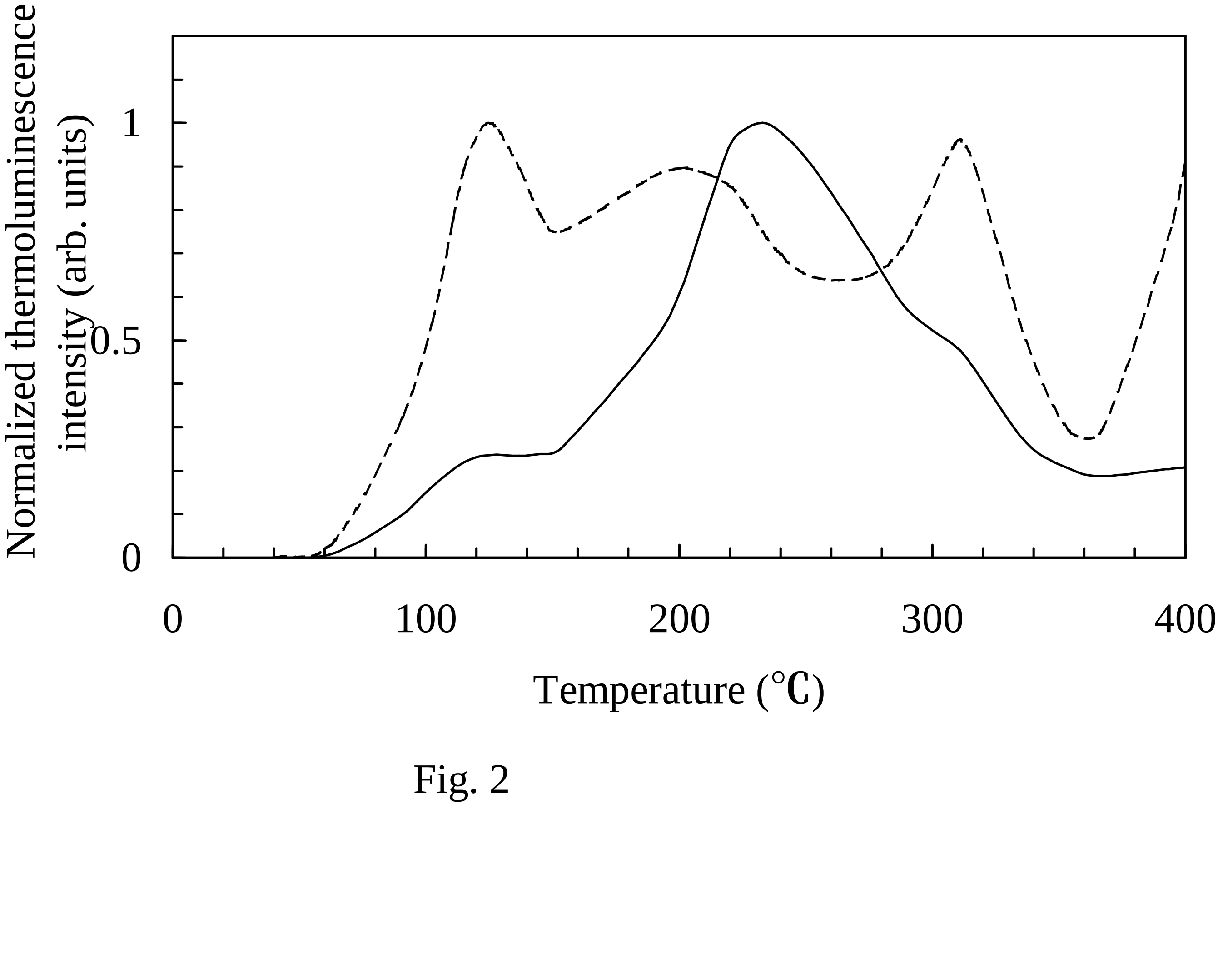

Normalized thermoluminescence intensity (arb. units)
1
0.5
0
0
100
200
300
400
Temperature (°C)

Fig. 1


Normalized thermoluminescence intensity (arb. units)
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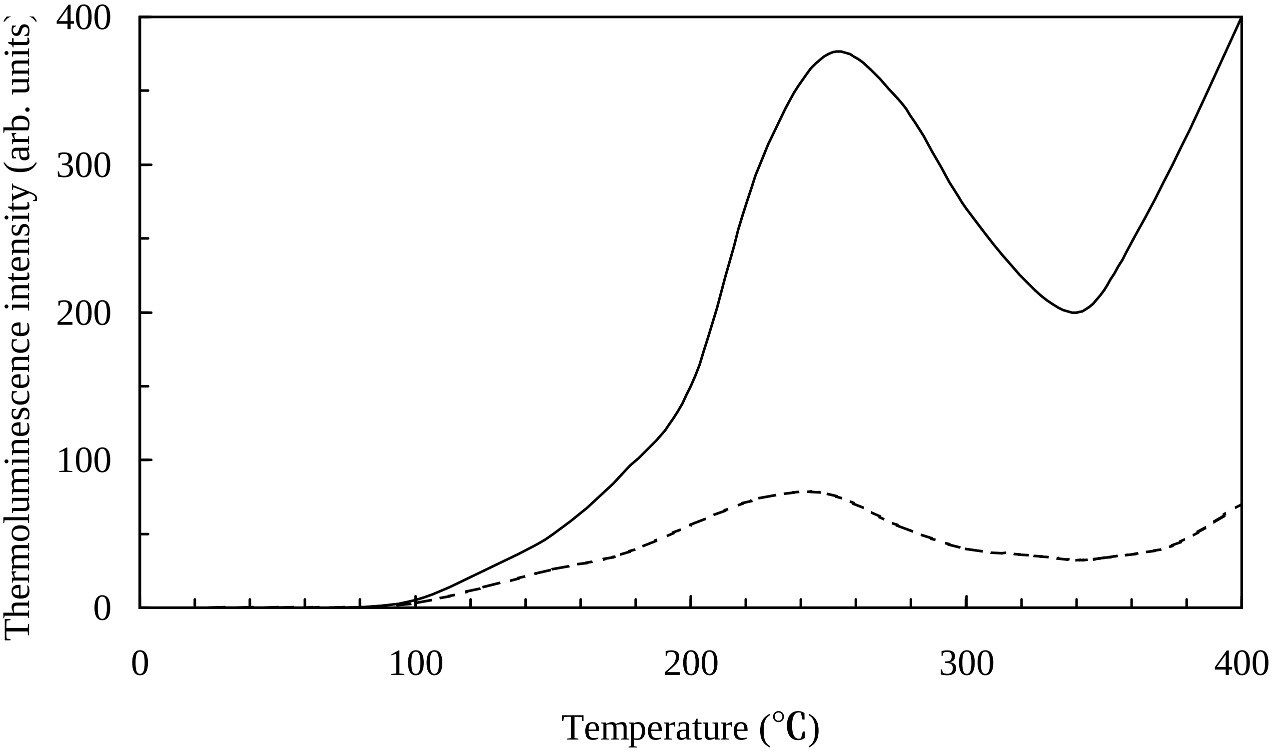
0.5
0
0
100
200
300
400
Temperature (°C)

Fig. 2



Thermoluminescence intensity (arb. units)
400
300
200
100
0
0
100
200
300
400
Temperature (℃)

Fig. 3

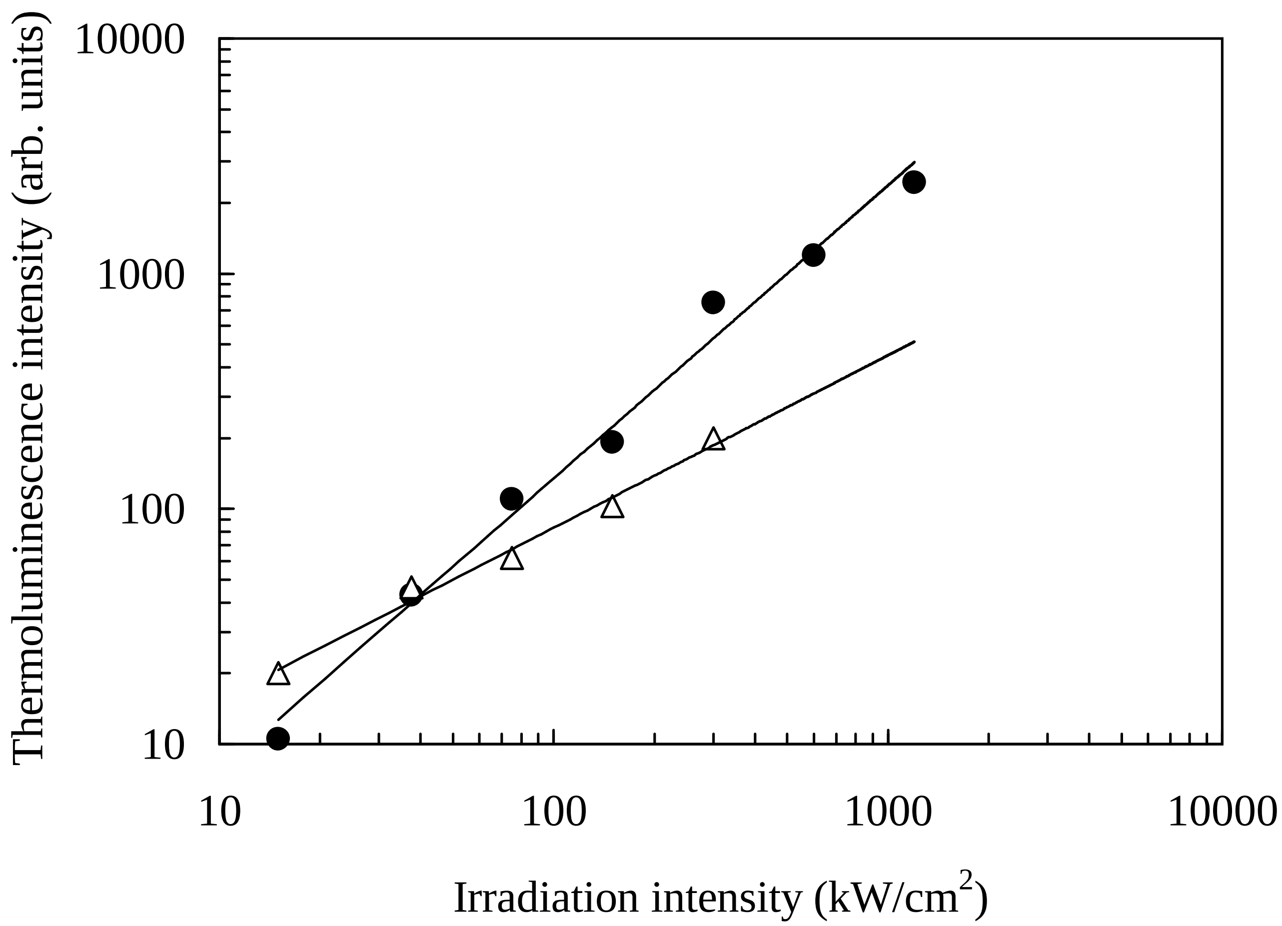

Thermoluminescence intensity (arb. units)
10000
1000
100
10
10
100
1000
10000
Irradiation intensity (kW/cm2)

Fig. 4