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Induced Absorption in X-ray-Irradiated CdS-Doped Glass

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Optical transmission spectra have been measured in X-ray-irradiated CdS-doped glass as functions of X-ray dose at 0.5-100 Gy and effective X-ray energy at 10-65 keV. Induced absorption is observed in the X-ray-irradiated glass with a dose higher than about 0.5Gy, and it increases linearly with a dose of up to 80 Gy. The induced absorption decreases with decreasing effective X-ray energy. Thermal stability was also studied. The origin of the induced absorption was discussed.

KEYWORDS: radiation dosimeter, optical absorption, X-ray irradiation, semiconductor, nanocrystal

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Since Jain and Lind$^{1)}$ first reported that semiconductor-doped glasses have a large optical nonlinearity and a fast response time, the optical properties of these materials have been investigated extensively. However, X-ray irradiation effects have been hardly investigated.$^{2)}$ We have reported that an electron spin resonance (ESR) signal$^{3)}$ and thermoluminescence$^{4)}$ are observed in X-ray-irradiated CdS-doped glasses. In previous papers, we reported the properties of thermoluminescence$^{5)}$ and the ESR signal,$^{6)}$ and we proposed that CdS-doped glass can be used as a material for radiation dosimetry with thermoluminescence and ESR. Here, we report the optical transmission spectra of X-ray-irradiated CdS-doped glass. Induced absorption was measured as functions of X-ray dose and effective X-ray energy. Although Grabovskis *et al.*$^{2)}$ reported on induced absorption in X-ray-irradiated CdS-doped glass, they did not report detailed results such as the dependence of the induced absorption on X-ray dose and effective X-ray energy. The thermal stability of the induced absorption was also examined. The results were compared with those of thermoluminescence and ESR.

The sample investigated was commercial CdS-doped filter glass, Asahi Y-44. Thermoluminescence and ESR signal have been observed in the sample,$^{5,6)}$ and the sample shows the most intense thermoluminescence in all the glasses investigated. The absorption edge of the glass was approximately 430 nm. The size of the sample was 10 mm in width and length and 2.5 mm in thickness. The concentration of CdS was approximately 0.4 wt%.$^{7)}$ The size of the CdS nanocrystals was approximately 3 nm.$^{8)}$ The glass composition was 70% SiO_2, 10% Na_2O, 10% ZnO, 6% K_2O, and 3% B_2O_3.$^{7)}$

The glass was exposed to X-rays from an X-ray source (Hitachi Medico MBR-1520R, W target, 150 kV, 20 mA) at 300 K. The effective X-ray energy was 10 keV. When low-energy X-rays were eliminated using a filter, which was composed of an Al plate and a Cu plate, effective X-ray energy increased. The effective X-ray energies were 33 keV using an Al plate with a thickness of 1.0 mm, 48 keV using both an Al plate with a thickness of 0.5 mm and a Cu plate with a thickness of 0.1 mm, and 65 keV using both an Al plate with a thickness of 0.5 mm and a Cu plate with a thickness of 0.3 mm. The filter mainly used was composed of an Al plate with a thickness of 0.5 mm and a Cu plate with a thickness of 0.1 mm, and consequently the effective X-ray energy was 48 keV.

Isochronal annealing was performed by heating irradiated samples at temperatures varying from 50 to 350°C. The annealing time was 15 min. Transmission spectra were measured using a spectrophotometer (JASCO V-550) at 300 K.

Figure 1 shows the transmission spectra of X-ray-irradiated and unirradiated CdS-doped glass, Y-44, at 300 K. The transmittance of the irradiated glass decreases around the absorption edge. The differential transmission spectrum is also shown in Fig. 1.

Figure 2 shows the dose response of the induced absorption of Y-44. The induced absorption increases linearly with a dose of up to 80 Gy, and it tends to saturate at higher dose levels. The induced absorption is observed in the glass with a dose higher than about 0.5 Gy. Therefore, the lower limit of X-ray detection is approximately 0.5 Gy. This value is almost the same as that for ESR.6)

The isochronal annealings of Y-44 for temperatures ranging from 50 to 350°C are shown in Fig. 3. Solid circles show data for the induced absorption and open triangles for ESR. The data points for ESR are from ref. 6. Signal intensity decreases with increasing temperature. The induced absorption decreases to about 80% of its original value at 100°C, and it almost vanishes at 350°C. On the other hand, the ESR signal vanishes at 250°C. Therefore, the thermal stability of the induced absorption is higher than that of ESR. Since the ESR signal is stable at room temperature,6) the induced absorption is stable at room temperature

The energy dependence of the induced absorption of Y-44 was examined and compared with that of the ESR signal. Figure 4 shows the results. The X-ray dose is 20 Gy for the induced absorption (solid circles) and 10 Gy for ESR (open triangles). The Data points for ESR are from ref. 6. The intensities are normalized. The induced absorption decreases with decreasing effective X-ray energy. The energy dependence of the induced absorption is different from that of ESR.

The effect of thermal annealing and the dependence of X-ray energy on the induced absorption are different from those of ESR, as shown in Figs. 3 and 4. Therefore, defects, which are responsible for the induced absorption, are considered to be different from those for ESR: the ESR signals may be ascribed to a trapped electron on a Cd^{2+} ion in the glass matrix and to the oxygen hole center.6)

Grabovskis *et al.*[2] reported that the induced absorption was attributable to the color centers in glass matrix. However, they did not report the origin of the color centers. Gomonnai *et al.*[9,10] reported the radiation-induced absorption increment in the alkali borosilicate glass matrix exposed to X-ray and electron irradiations. The additional absorption bands are attributed to the intrinsic hole radiation color centers ${H_3}^+$, ${H_2}^+$, and ${H_4}^+$.[10] The peak energy positions are 2.03 eV for ${H_3}^+$, 2.73 eV for ${H_2}^+$, and 4.0 eV for ${H_4}^+$. The bands corresponding to the ${H_3}^+$ and ${H_2}^+$ centers vanish with thermal annealing at 200°C. On the other hand, the band corresponding to the ${H_4}^+$ centers does not vanish with annealing at 200°C. Therefore, the induced absorption in Fig. 1 is considered to be attributable to a lower energy tail of the absorption band corresponding to the ${H_4}^+$ centers.

Thermoluminescence has been observed in Y-44. The effect of thermal annealing and the dependence of X-ray energy on thermoluminescence are the same as those of ESR. Therefore, defect, which is responsible for thermoluminescence, is considered to be the same as that for ESR. The intensity of thermoluminescence is proportional to an X-ray dose of up to 10 Gy, and shows supralinearity at higher dose levels.[5] The lower limit of X-ray detection is approximately 1 mGy. Although the lower limit of X-ray detection is approximately 1/500 that for the induced absorption, repeat measurement is impossible for thermoluminescence.

In summary, induced absorption has been observed in the transmission spectrum of X-ray-irradiated CdS-doped glass, Y-44. The induced absorption increases linearly with a dose of up to 80 Gy, and it decreases with decreasing effective X-ray energy. The induced absorption is stable at room temperature. The induced absorption is considered to be due to the intrinsic hole radiation color centers in a glass matrix.

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Figure captions

Fig. 1. Transmission spectra of X-ray-irradiated and unirradiated CdS-doped glass, Y-44, and differential transmission spectrum of Y-44 at 300 K. The X-ray dose is 20 Gy and the effective X-ray energy is 48 keV.

Fig. 2. Induced absorption of Y-44 as function of X-ray dose. The effective X-ray energy is 48 keV. A curve was drawn through the data points as a visual guide.

Fig. 3. Isochronal annealings of Y-44. The X-ray dose is 20 Gy for induced absorption and 10 Gy for ESR. The effective X-ray energy is 48 keV. The annealing time is 15 min. The data points for ESR are from ref. 6. Curves were drawn through the data points as visual guides.

Fig. 4. Intensities of induced absorption and ESR signal of Y-44 as function of effective X-ray energy. The intensities are normalized. The X-ray dose is 20 Gy for induced absorption and 10 Gy for ESR. The data points for ESR are from ref. 6. Curves were drawn through the data points as visual guides.

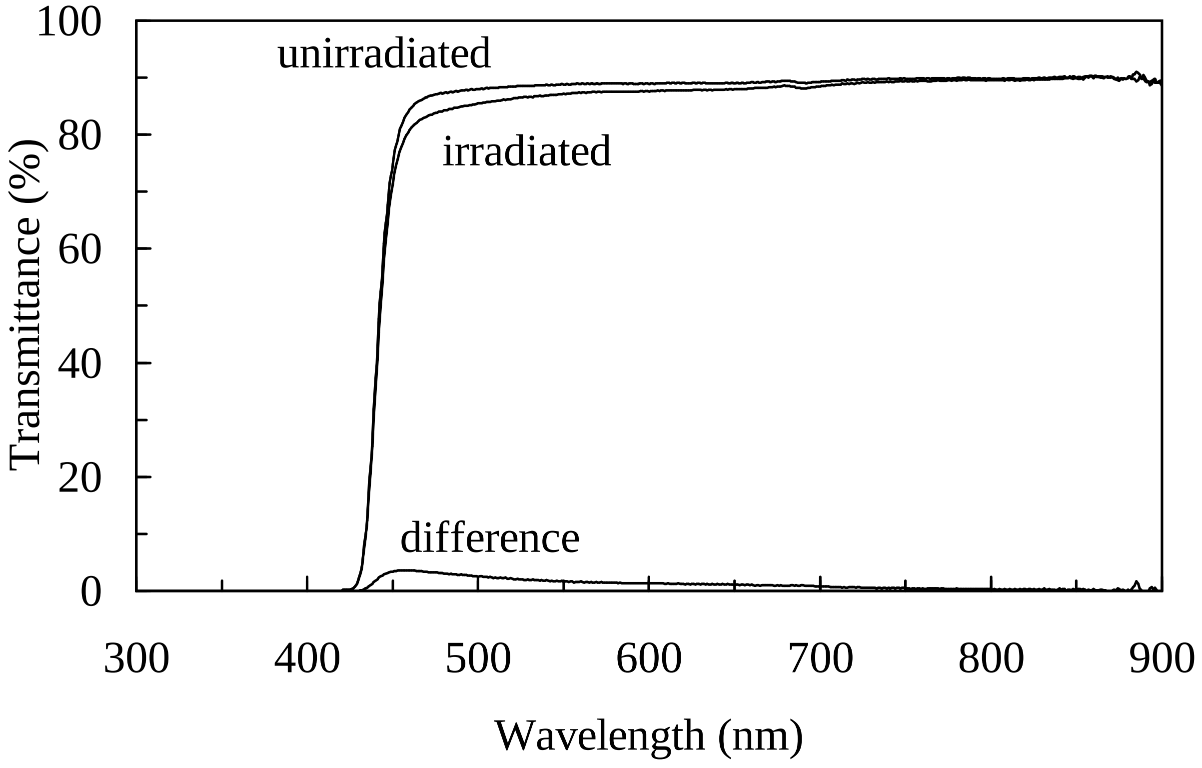

100
80
60
40
20
0
Transmittance (%)
unirradiated
irradiated
difference
300
400
500
600
700
800
900
Wavelength (nm)

Fig. 1

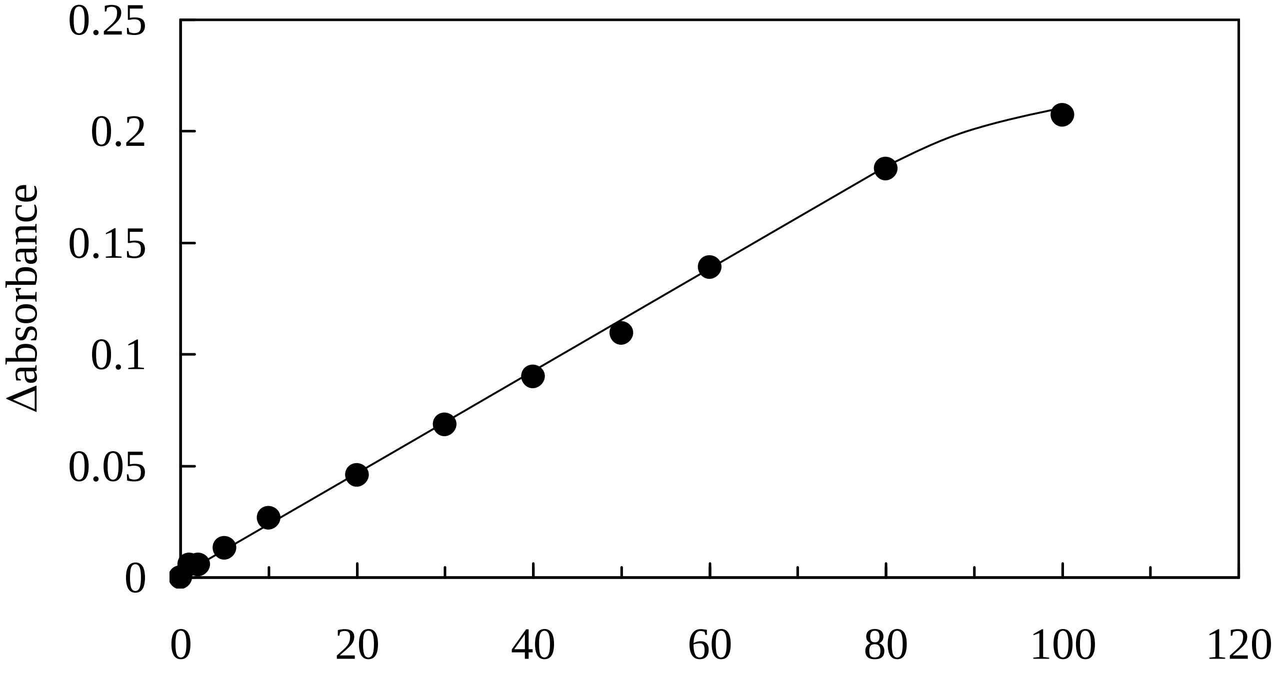

0.25
0.2
0.15
0.1
0.05
0
Δabsorbance
0
20
40
60
80
100
120
X-ray dose (Gy)

Fig. 2

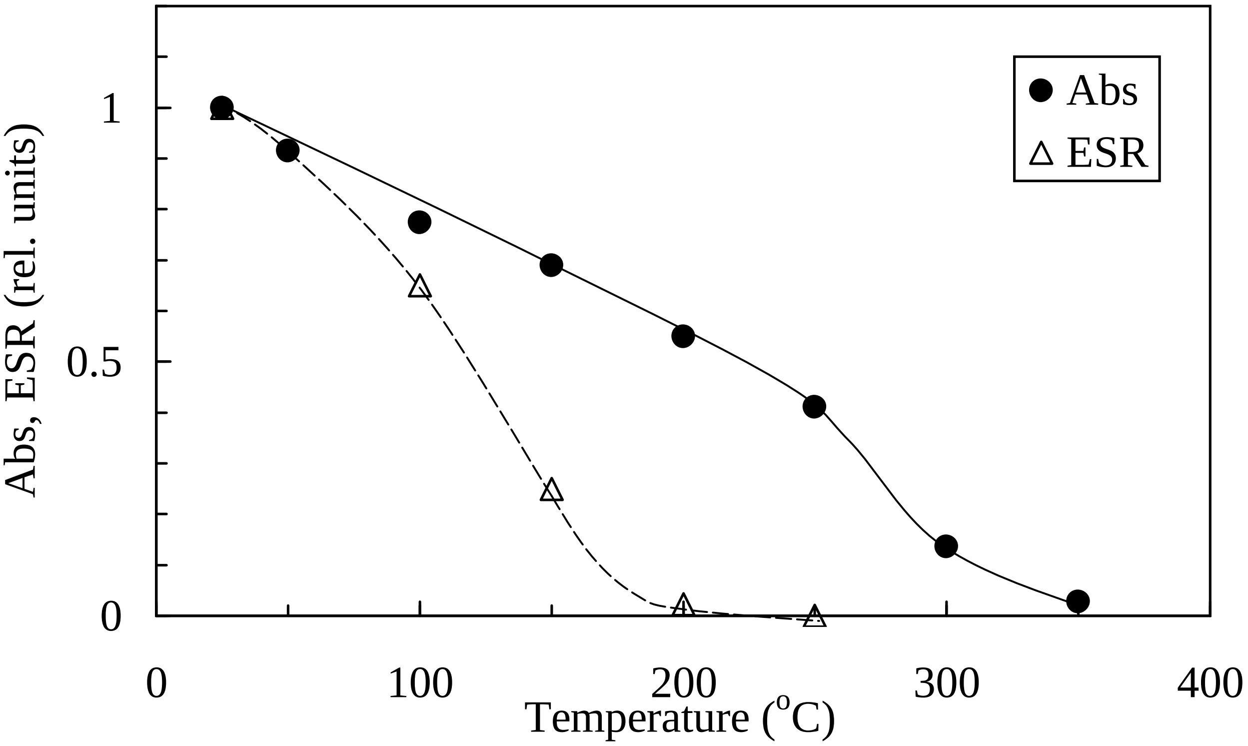
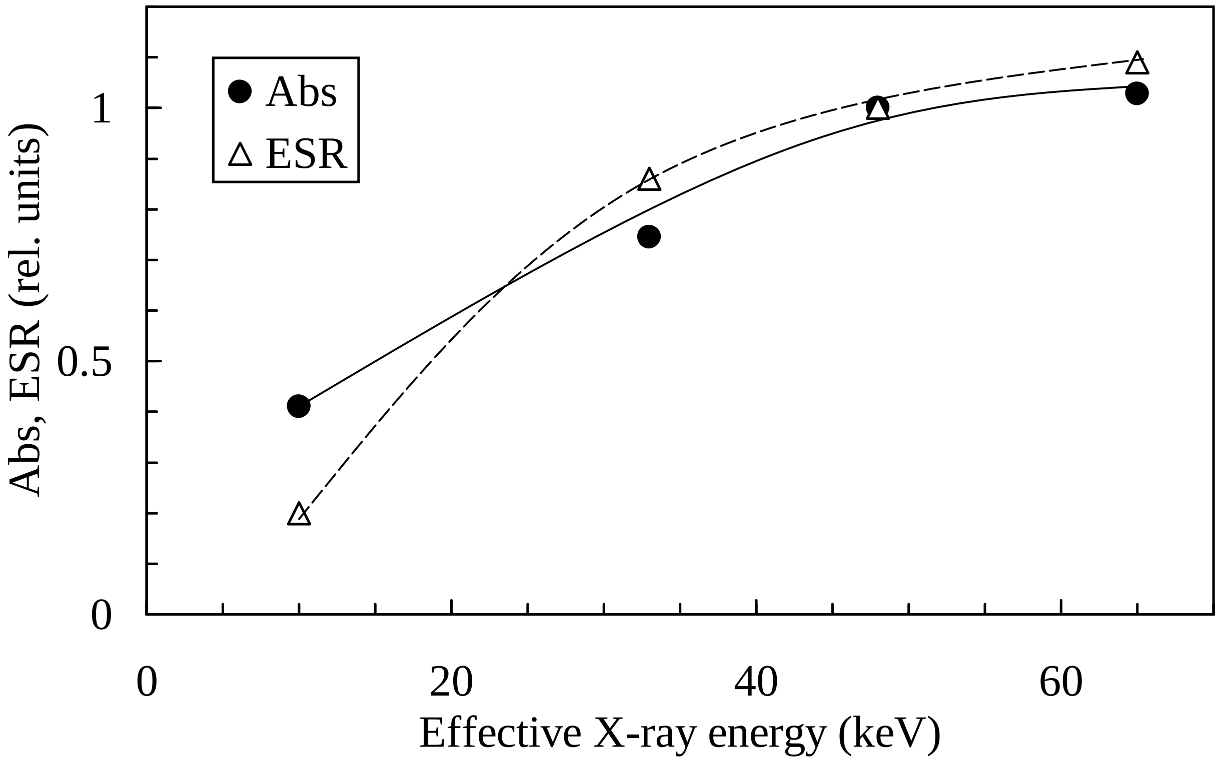

Abs, ESR (rel. units)
1
0.5
0
● Abs
△ ESR
0
100
200
300
400
Temperature (^oC)

Fig. 3



● Abs
△ ESR
Abs, ESR (rel. units)
0
0.5
1
0
20
40
60
Effective X-ray energy (keV)

Fig. 4