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CdS-Doped Glass as Dosimetric Material with Electron Spin Resonance

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Electron spin resonance (ESR) spectra have been measured in X-ray-irradiated CdS doped glass as functions of X-ray dose and effective X-ray energy. The intensity of the ESR signal is proportional to X-ray dose up to 40Gy. CdS-doped glass is more sensitive than a commercial alanine dosimeter. The intensity of the ESR signal decreases with decreasing effective X-ray energy for energies lower than 33 keV. Thermal stability was also studied. The ESR signal is relatively stable at room temperature.

KEYWORDS: radiation dosimeter, electron spin resonance, X-ray irradiation, semiconductor, nanocrystal

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Alanine is an amino acid used for radiation dosimetry using electron spin resonance (ESR). However, the sensitivity of alanine dosimeters is low: the lower limit is approximately 10 Gy. In recent years, Hassan *et al.*[1] have reported that bioglass (45% SiO_2, 24.5% CaO, 24.5% Na_2O, and 6% P_2O_5) was tenfold more sensitive than commercial alanine dosimeters. In a previous paper,[2] we reported that an ESR signal was observed in X-ray irradiated CdS-doped glass, which contains CdS nanocrystals in silicate glass. CdS nanocrystals act as the source of electrons and holes. Electrons are considered to be trapped in glass and holes at the glass-semiconductor interface. These traps are detected by ESR measurement. CdS-doped glass is anticipated to be more sensitive than bioglass and the alanine dosimeter. Here, we report the intensity of the ESR signal as functions of X-ray dose and effective X-ray energy. The thermal stability of the ESR signal was also examined. The sensitivity and X-ray energy dependence of CdS-doped glass were examined by ESR spectroscopy and compared with those of a commercial alanine dosimeter.

The sample investigated was commercial CdS-doped filter glass, Asahi Y-44. The size of the sample was 2 mm in width, 2.5 mm in thickness and 10 mm in length. The concentration of CdS was approximately 0.4 wt%.[3] The size of CdS nanocrystals was approximately 3 nm.[4] The glass composition was 70% SiO_2, 10% Na_2O, 10% ZnO, 6% K_2O, and 3% B_2O_3.[3] We also measured the ESR spectra of undoped glass, which does not contain CdS nanocrystals, and the alanine dosimeter (Hitachi Cable Aminogray) for comparison.

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, 39 keV using an Al plate with a thickness of 2.0 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, 58 keV using both an Al plate with a thickness of 0.5 mm and a Cu plate with a thickness of 0.2 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 the Al plate with a thickness of 0.5 mm and the Cu plate with a thickness of 0.1 mm, and consequently

the effective X-ray energy was 48 keV.

ESR spectra were measured using an X-band ESR spectrometer (Bruker ELEXSYS E-500) at 300 K using a cylindrical cavity operating at 9.8 GHz with a 100 kHz modulation frequency. The microwave power was 10 mW, and the modulation amplitude was 0.1 mT. The response time constant was 0.16 s with a field-sweeping rate of 20 mT/84 s.

Isochronal annealing was performed by heating irradiated samples at temperatures varying from 100°C to 250°C. The annealing time was 15 min.

Figure 1 shows ESR spectra of X-ray irradiated CdS-doped glass, Y-44, and undoped glass, Y-0, at 300 K. The ESR spectrum for the CdS-doped glass is characterized by two main signals: $g = 2.01$ and $g = 1.99$. The signal at $g = 1.99$ may be ascribed to a trapped electron on a Cd^{2+} ion in the glass matrix.[2)] The signal at $g = 2.01$ is similar to that in bioglass.[1)] Therefore, this signal may be ascribed to the oxygen hole center. These signals were not observed in undoped glass, Y-0, which does not contain CdS nanocrystals. The ESR signal was not observed in unirradiated CdS-doped glass, Y-44.

Figure 2 shows the dose response of ESR intensity of Y-44. The observed relationship between the absorbed dose and the peak-to-peak intensity of the ESR signal suggests a linear function in the dose range of 1–40 Gy. The intensity of the ESR signal is 4540 (arbitrary units) for 40 Gy. On the other hand, the intensity of the ESR signal is 2140 for the alanine dosimeter. The sensitivity of Y-44 seems to be two times that of the alanine dosimeter. However, the sample size of Y-44 is smaller than that of the alanine dosimeter: 2 mm in width, 2.5 mm in thickness and 10 mm in length for Y-44 compared with 3 mm in diameter and 30 mm in length for the alanine dosimeter. Therefore, Y-44 is considered to be approximately five times more sensitive than the alanine dosimeter.

Hassan *et al.*[1)] reported that bioglass was ten times more sensitive than a commercial alanine dosimeter. The sensitivity of bioglass seems to be higher than that of Y-44. However, the size of the alanine dosimeter (Bruker) used by them is considered to be 5 mm in diameter and 4 mm in length. The volume of their dosimeter is about 1/3 of that of the dosimeter (Hitachi Cable) used in the present experiment. Moreover, the volume of bioglass is approximately two times larger than that of Y-44, since the weight of the sample is 200 mg for bioglass and 100 mg for Y-44. These factors increase the sensitivity

of bioglass relative to that of the alanine dosimeter. This suggests that Y-44 is more sensitive than bioglass. In fact, Hassan *et al.*[1] observed a linear relationship between ESR intensity and dose for bioglass in the dose range of 5–1000 Gy, and we observed a linear relationship between ESR intensity and dose for Y-44 in the lower dose range of 1–40 Gy. Therefore, Y-44 is considered to be more sensitive than bioglass. In addition to this, their experimental conditions were different from those of the present experiment. The microwave power was 70 mW, and the modulation amplitude was 0.7 mT in their experiment. On the other hand, the microwave power was 10 mW, and the modulation amplitude was 0.1 mT in the present experiment. We measured the intensity of the ESR signal of Y-44, when the modulation amplitude was 0.5 mT. The intensity of the ESR signal was five times higher than that at 0.1 mT.

The isochronal annealings of Y-44 and bioglass (Bio-G)[1] for temperatures ranging from 100°C to 250°C are shown in Fig. 3. Relative intensity decreased with increasing temperature. Signal intensity decreased to 65% of its original value at 100°C. This indicates that trapped electrons and holes in Y-44 are relatively stable at room temperature and unstable at higher temperatures. The fading was investigated for Y-44 at 300 K. There was a small decrease (10%) in signal intensity after two months. The thermal stability and fading are similar to those in bioglass.[1]

Figure 4 shows the energy response of ESR intensity for Y-44 and the alanine dosimeter. Although the intensity of the ESR signal depends on the energy of X-ray, it is almost independent of effective X-ray energy for energies higher than 33 keV. The intensity of the ESR signal of the alanine dosimeter should be independent of X-ray energy, since alanine is a tissue equivalent material. However, the signal intensity for 10 keV is 34% of that for 48 keV. This is considered to be due to the fact that the sensitivity of the X-ray sensor in the X-ray source depends on X-ray energy. The effective atomic number of Y-44 is 15. This value is larger than that of tissue equivalent materials (7.5). However, the energy dependence for Y-44 is not very different from that for the alanine dosimeter in the X-ray energy region. The cause or mechanism of this energy dependence has not yet been clarified.

Thermoluminescence has been observed in Y-44.[5] The intensity of thermoluminescence

is proportional to X-ray dose up to 10 Gy.[6] The lower limit of X-ray detection is approximately 1 mGy (the value reported in ref. 6 is underestimated). Although the sensitivity of the thermoluminescence dosimeter using CdS-doped glass is higher than that of the ESR dosimeter using the same material, repeat measurement is impossible for the thermoluminescence dosimeter.

In summary, ESR signals have been observed in X-ray-irradiated CdS-doped glass, Y-44. The intensity of the ESR signal is proportional to X-ray dose up to 40 Gy. Y-44 is more sensitive than an alanine dosimeter. The intensity of the ESR signal is almost independent of effective X-ray energy for energies higher than 33 keV. The ESR signal is relatively stable at room temperature.

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Figure captions

Fig. 1. ESR spectra of X-ray-irradiated CdS-doped glass, Y-44, and undoped glass, Y-0, at 300 K. X-ray dose is 10 Gy, and the effective X-ray energy is 48 keV.

Fig. 2. Intensity of ESR signal of Y-44 as function of X-ray dose. The effective X-ray energy is 48 keV. A line was drawn through data points as a visual guide.

Fig. 3. Isochronal annealings of Y-44 and bioglass (Bio-G).[1)] X-ray dose is 10 Gy, and the effective X-ray energy is 48 keV for Y-44. The annealing time is 15 min. Curves were drawn through data points as visual guides.

Fig. 4. Intensities of ESR signal of Y-44 and the alanine dosimeter as functions of effective X-ray energy. The intensities are normalized. X-ray dose is 10 Gy for Y-44 and 40 Gy for the alanine dosimeter. Curves were drawn through data points as visual guides.

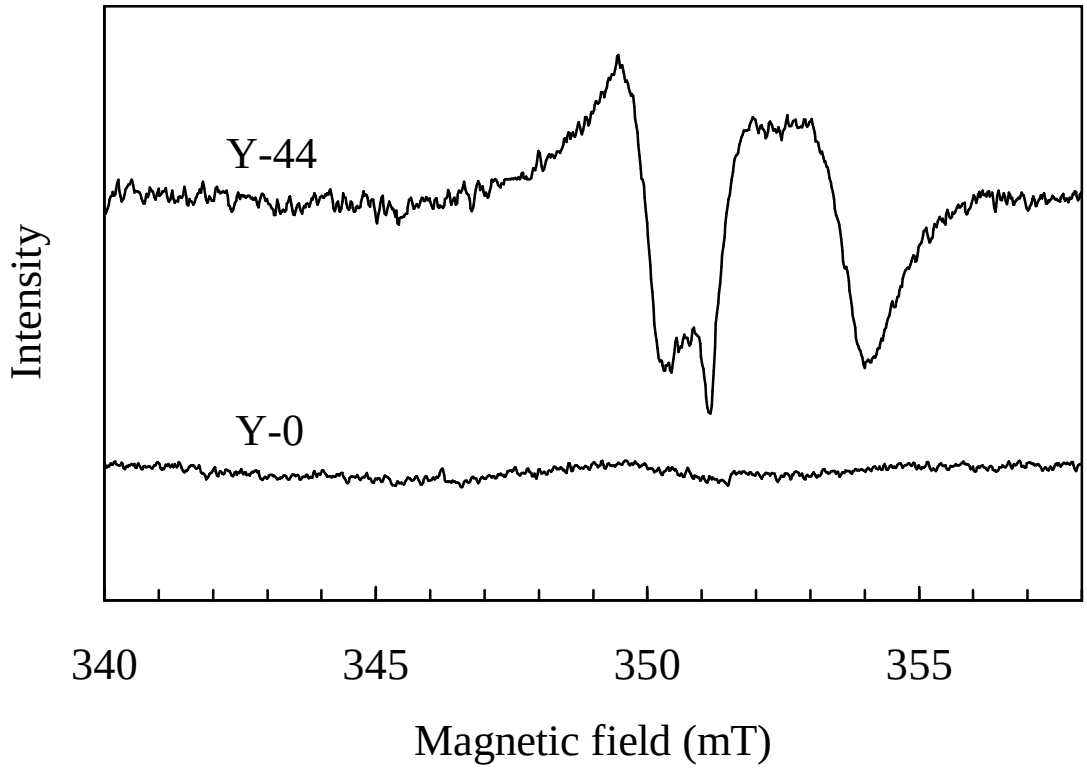

Fig. 1

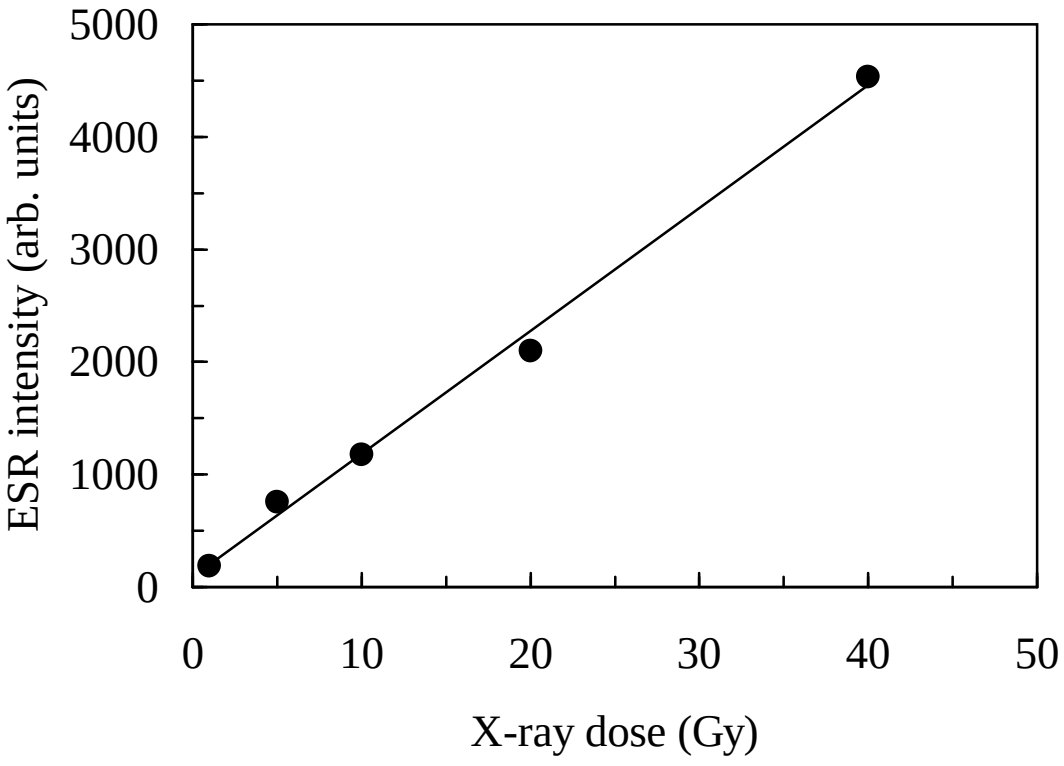
5000
4000
3000
2000
1000
0
ESR intensity (arb. units)
0
10
20
30
40
50
X-ray dose (Gy)

Fig. 2

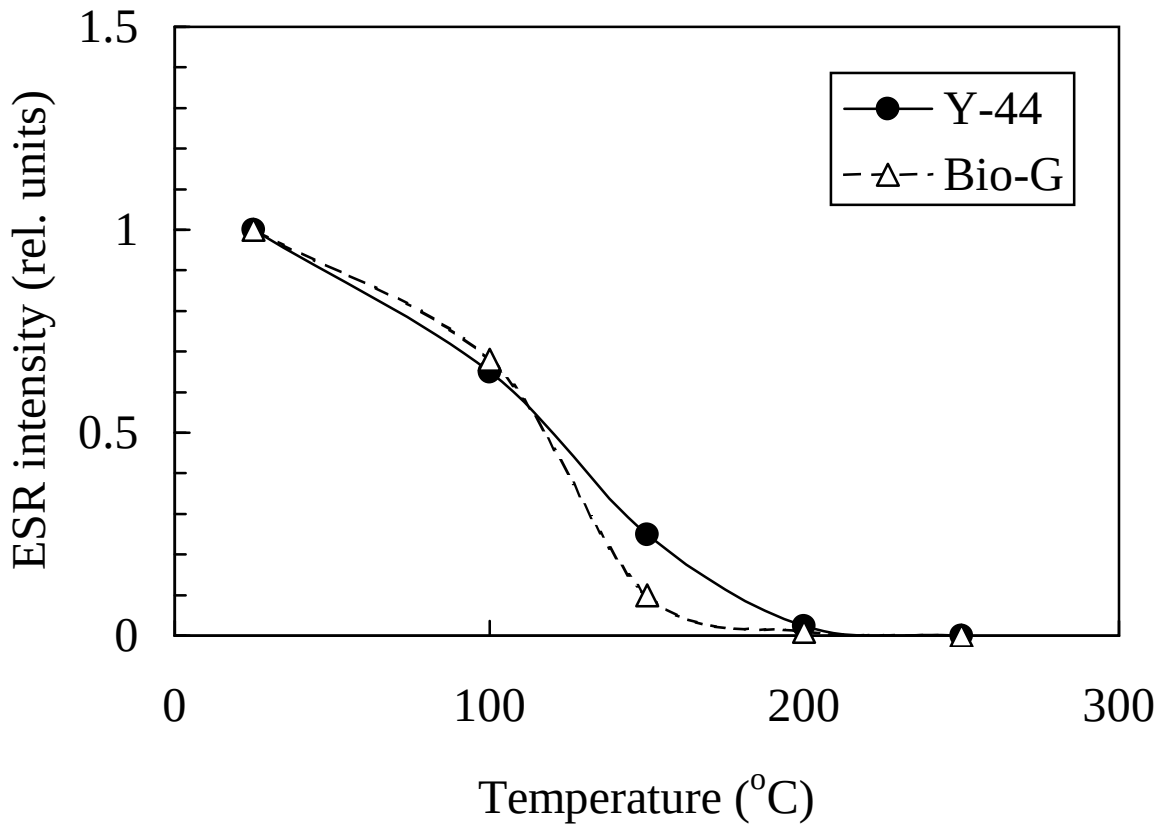
1.5
1
0.5
0
ESR intensity (rel. units)
Y-44
Bio-G
0
100
200
300
Temperature (°C)

Fig. 3

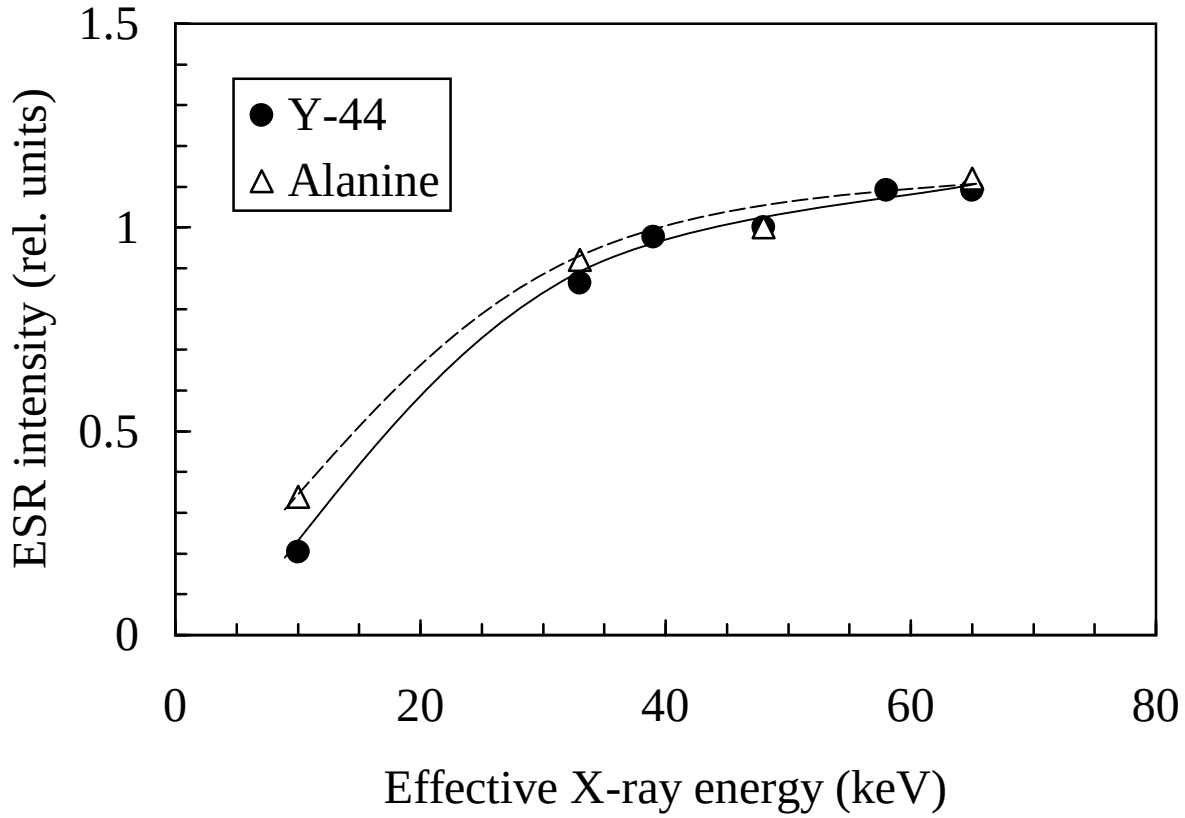
1.5
1
0.5
0
ESR intensity (rel. units)
Y-44
Alanine
0
20
40
60
80
Effective X-ray energy (keV)

Fig. 4