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Radiation damage of CsI(Tl) scintillators: blocking of energy transfer process of V_k centers to Tl^+ activators

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Abstract

This paper reports the emission spectra, light output, transmission and decay curves of CsI(Tl) crystals irradiated with gamma rays at different doses, ranging from 1×10^5 to 5×10^5 Gy. The crystals were coated with black or white tapes. Significant decreases in the emission spectra and light output values were observed for the crystals coated with white tape as the radiation dose increased. The decrease in the degree of the rising part of the decay curve in the irradiated crystals is attributed to the blocking of the energy transfer processes of V_k lattice disorders, which were produced in irradiated crystals. The scintillation mechanism is affected in the crystal irradiated at 5×10^5 Gy. However for crystals irradiated below 10^5 Gy the mechanism process is not altered, and the decrease in the light output is due to internal transmission loss. It was also observed that the damage for irradiation is not permanent and it obeys a bi-exponential function. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Radiation damage; CsI(Tl) crystals; Scintillation mechanism; Light output

1. Introduction

Although a number of recent studies have produced a great deal of information about radiation damage in many types of scintillator crystals, the complex effects that radiation can cause in most of these materials is not well understood. For example, it is believed that the loss of light output, which characterizes radiation damage in CsI(Tl), is predominantly due to decreased absorption length, not to decreased scintillation light yield [1–3]. However, Woody et al. studied the radiation damage in CsI and CsI(Tl) and reported that the results from the light output and transmission measurements indicate an apparent loss in light output that exceeds what would be expected simply from the loss in internal transmission [4].

Recently, Kubota et al. studied the radiation damage of NaI(Tl) by fast neutron irradiation. They reported that the decrease in the light output from irradiated NaI(Tl) crystals is attributed to the blocking of the energy transfer process of V_k centers in the host lattice to Tl^+ activators by the lattice disorders produced by fast neutrons [5]. The effect of this blocking of the energy transfer processes was clearly observed as a difference in the decay curves for the NaI(Tl) crystals before and after fast neutron irradiation.

We carried out a study of radiation damage in CsI(Tl) crystals to see if the scintillation

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mechanism was affected by the irradiation. CsI(Tl) crystals were irradiated with gamma rays at different doses, ranging from 10^{-1} to 5×10^5 Gy. After the irradiation, the scintillation light output and decay curves were measured. The effect of the scintillation mechanism damage was clearly observed as a difference in the decay curves for the unirradiated crystal and the irradiated crystal for dose of 5×10^5 Gy. As a complementary experiment, emission and transmittance spectra were measured before and after each irradiation. In addition, recovery was also studied as a function of time.

2. Experimental procedure

The CsI(Tl) crystals sized $1 \times 1 \times 1\,cm^3$, with all sides highly polished were obtained from Crismatec. The crystals were irradiated by a ^{60}Co gamma ray source, which is a gamma cell type with activity of 3.15×10^{14} Bq and average dose rate of 5.8 kGy/h (0.58 Mrad/h). The total delivered dose ranged from 10^{-1} to 5×10^5 Gy in air at room temperature.

The light outputs were measured with a quartz window photomultiplier (Hamamatsu Photonics H3378-51). The scintillator surfaces, except the coupling face, were coated with "white" tape or "black" tape. The output signals from the photomultiplier were shaped with an RC network having a shaping time constant of about 5 μs. The gamma rays of 0.662 MeV from a ^{137}Cs source were used to excite the CsI(Tl) crystal.

The emission spectra from the CsI(Tl) crystals were measured with a Shimadzu Bausch and Lomb monochromator. A RaD-Be source was used to excite the crystals. The energy-analyzed photons from the monochromator were detected by a photomultiplier R943-02 (Hamamatsu Photonics). Anode current was measured with an electronic picoammeter (Advantec). The spectra have not been corrected for the spectral response of the detection system.

Transmission was measured with a model V-530 pulsed UV/VIS spectrometer (JAS Co.). The single photon counting method was used to measure the scintillation decay curves of CsI(Tl) crystals for the time range of 300 ns. The experimental method is similar to that reported in Ref. [5]. The decay curves were also measured by a Tektronix 784A oscilloscope for the time range of 5 μs. All the measurements were performed at room temperature.

3. Results and discussion

3.1. Light output

Fig. 1 shows the relative change in light outputs from "white" taped crystals as a function of dose. The measurements were made immediately and 3 days after irradiation. The light output of the sample for a dose of 10^3 Gy could not be measured immediately after irradiation due to the strong, long-lived phosphorescence. This effect is well known and has been reported in Refs. [4,6].

The light outputs from the above crystals coated with "black" tape were measured 3 days after irradiation and they are also shown in Fig. 1. It is observed that the degree of the decrease in the light outputs of the irradiated crystals measured with "black" tape is smaller compared with that measured with "white" taped crystal. By considering the enhanced multiple reflection in "white" coated crystal, it may be concluded that the transmission loss is considered mainly as causing a decrease in light output for "white" taped irradiated crystal.

The previous results from Refs. [2,3,6,7] are also shown for comparison. Our crystals with $1 \times 1 \times 1\,cm^3$ show a noticeable damage for doses higher than 10^2 Gy. A similar result was reported in Ref. [2] for CsI(Tl) crystals with $\varnothing\ 2.54 \times 2.54\,cm^3$. It is to be noted here that in the previous studies the size of CsI(Tl) crystals was $1 \times 1 \times 10\,cm^3$ in Ref. [6] and 37 cm in length in Ref. [3].

3.2. Emission spectra

Emission spectra were measured for the crystals with a total delivered dose of 10^5 and 5×10^5 Gy 66 days after irradiation. Hereafter, the unirradiated crystal and the crystals for the total

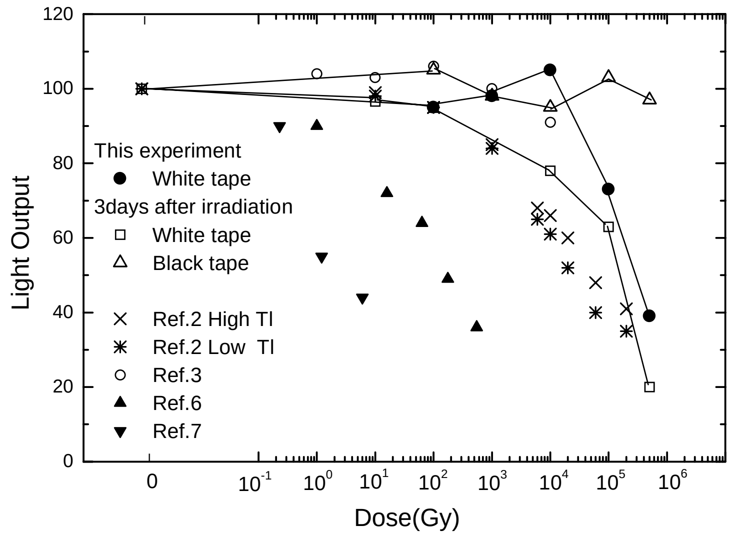

Fig. 1. Relative light output as a function of dose. The line is a guide to the eyes.

delivered dose of 10^5 and 5×10^5 Gy are labeled as U, R and RR, respectively. Fig. 2 shows the emission spectra from three crystals U, R and RR coated either with "white" tape or "black" tape. An appreciable decrease in the emission intensities for the wavelength of less than 600 nm is observed from R and RR crystals coated with "white" tape. However, only a slight decrease is observed from these two crystals when coated with "black" tape. The slight decrease in the emission intensity observed for irradiated crystals coated with the black tape may be attributed to the less reflection occurred, diminishing the degree of the transmission loss.

3.3. Transmission spectra

Fig. 3 shows the transmission spectra for the three crystals U, R and RR 60 days after irradiation. In this figure, the transmittance for 650 nm for the three crystals is assumed to be 100%, because we observed no appreciable difference in the intensities for the wavelength of more than 600 nm. It is seen that the transmission spectrum for the crystal RR has the absorption band around 380, 420–460, 515 and 560 nm. The

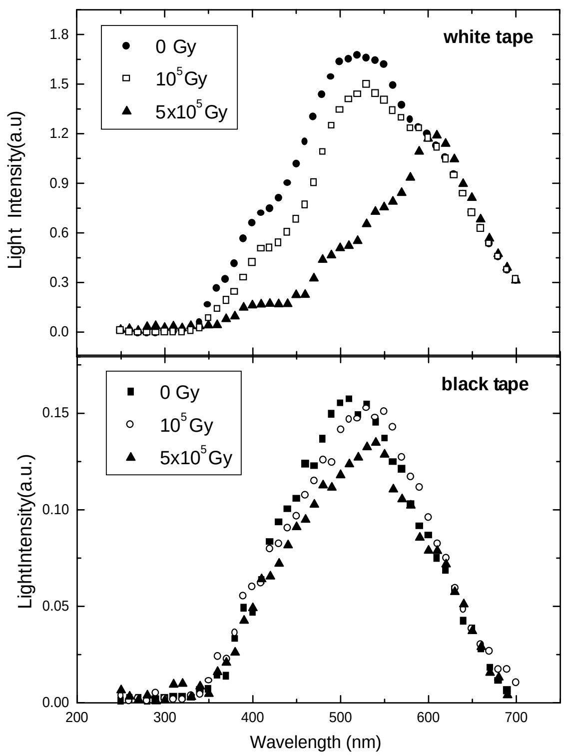

Fig. 2. Emission spectra from the CsI(Tl) crystals for doses at 0, 10^5 and 5×10^5 Gy, coated with "white tape" (upper) and "black tape" (lower).

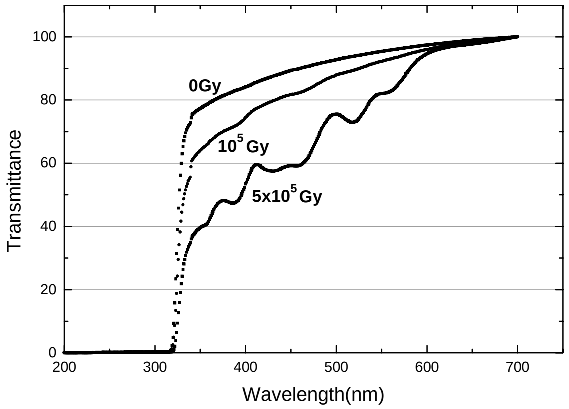

Fig. 3. Transmittance spectra from the CsI(Tl) crystals irradiated at 0, 10^5 and 5×10^5 Gy.

effect of these absorption bands is apparently seen as the dips for the emission spectrum of the crystal RR coated with "white" tape as shown in Fig. 2.

3.4. Decay curves

No difference in their decay curves could be detected from the U and RR crystals coated with "white tape" or "black tape". Fig. 4a shows the decay curves of the time range of 300 ns for U, R and RR measured 3 days after the irradiation. It is seen that the degree of the rising part decreases in the order of R and RR. Fig. 4b and c, shows the decay curves of the time range of 5 μs for U and RR measured after 60 days. An apparent difference is seen in the rising part of the decay curves, i.e. a relatively faster rise is observed in RR compared with that in U. The decay curve for R, which is not shown here, is similar to that for U.

Hamada et al. studied the decay curves for CsI(Tl) with different Tl concentrations under electron, alpha particle and pulsed UV excitations. Their results were well explained in terms of the three energy-transfer processes from the host lattice to Tl^+ ion activators [8]. The three energy-transfer processes that convert Tl^+ ions to the excited state $(Tl^+)^*$ are:

(i) the prompt process of a hole and an electron capture at a Tl^+ site,
(ii) the V_k diffusion process to Tl^0, and
(iii) the electron release process from Tl^0 to Tl^{++}.

Here, Tl^0 and Tl^{++} were produced through $(Tl^+ + \text{electron}) \rightarrow Tl^0$ and $(Tl^+ + \text{hole}$ (or V_k center)) $\rightarrow Tl^{++}$, respectively.

The rising part of the decay curve has been interpreted in terms of emission from the $(Tl^+)^*$ through process (ii) [8]. Therefore, the decrease in the degree of the rising part as shown in Fig. 4 is explained as being due to the less contribution of the emission from the process (ii).

By considering the above three processes, the decay curve is given by

$$I(t) = I_{i}(t) + I_{ii}(t) + I_{iii}(t), \quad (1)$$

where $I_i(t)$, $I_{ii}(t)$ and $I_{iii}(t)$ correspond to the decay curves for three processes and they are

$$I_i(T) \propto ((Tl^+)^*_0/T)\exp(-t/T), \quad (2)$$

$$I_{ii}(t) \propto (Tl^0)_0 \times \left(\exp(-t/T) - \exp(-t/T_v)\right)/(T - T_v), \quad (3)$$

and

$$I_{iii}(t) \propto (Tl^{++})_0 \times \left(\exp(-t/T_r) - \exp(-t/T)\right)/(T_r - T), \quad (4)$$

where, T is the lifetime of $(Tl^+)^*$, T_v is the V_k diffusion time and T_r is the electron release time from Tl^0.

The measured decay curves for U, R and RR were fitted to Eq. (2). In this fitting T is fixed to 585 ns obtained under pulsed UV excitation [8]. Table 1 summarized the fitted T_v and T_r, light output and the light intensity ratio $I_i : I_{ii} : I_{iii}$, where I_i is the light intensity from i-th process.

Here, we consider the effect of the irradiation for the three energy transfer processes. Since the prompt process (i) occurs at a Tl^+ site provided if a hole and an electron are produced within a trapping radial distance of 1.4 nm [9], it is expected that this process is less affected by the crystal defects produced by irradiation. On the other hand, in the processes (ii) and (iii), V_k centers and electrons should diffuse the order of 10 nm in a host lattice as described in Ref. [9]. Lattice disorders may prevent V_k centers from diffusing crystal defects, such as vacancies that may capture electrons. Therefore, it is expected that the energy-transfer processes (ii) and (iii) are affected by

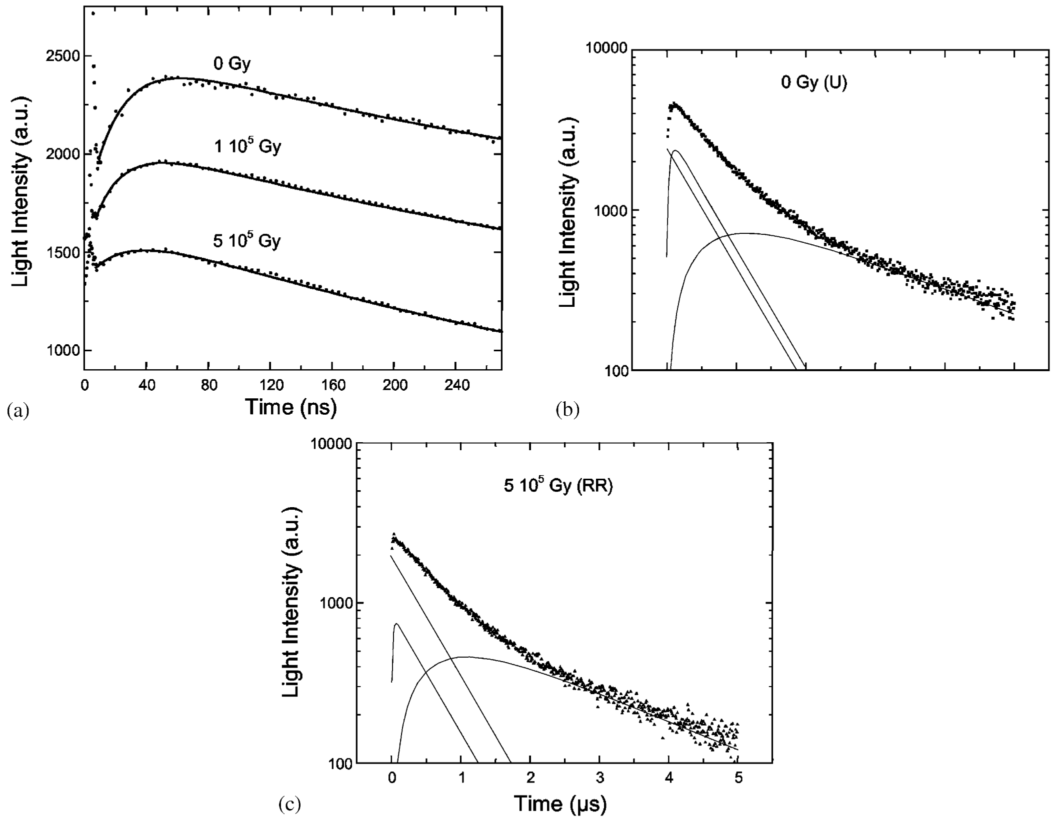

Fig. 4. Decay curves of the time range of 300 ns for CsI(Tl) crystals irradiated at 0, 10^5 and 5×10^5 Gy (a). Decay curves of the time range of 5 μs for crystals irradiated at 0 Gy (b) and 5×10^5 Gy (c).

Table 1
Fitted decay times T_v, T_r, light output and intensity ratios for *i*th component for the CsI(Tl)crystals, for doses of 0 Gy (U), 10^5 Gy (R) and 5×10^5 Gy (RR)

Sample	Fitted decay time (ns)			Light output (%)		Intensity ratio (%)
	T	T_v	T_r	w^a	w^b	$I_i:I_{ii}:I_{iii}$ [c]
U	585	50 ± 5	2700 ± 200	100	100	23:28:49
R	585	40 ± 5	2700 ± 200	63	83	23:29:48
RR	585	18 ± 5	2400 ± 200	20	55	32:15:53

[a] Immediately after irradiation.
[b] 150 days after irradiation.
[c] Errors are about 10%.

lattice disorders, which were produced by gamma-rays.

Less contribution of the emission from the process (ii) as shown in Fig. 4 may occur due to the blocking of the energy transfer of V_k centers. According to this explanation, if any scintillation mechanism is damaged, a relative increase in I_i and a relative decrease in I_{ii} and (or) I_{iii} are expected in irradiated crystals.

If the scintillation mechanism of CsI(Tl) is not affected in irradiated crystals and the loss of the light output is predominantly due to the loss of transmission, the intensity ratio for irradiated crystals should be the same as that for U. It is observed that the intensity ratio for R is nearly the same as that for U within an experimental error. Thus, the decrease in the light output for R should be predominantly explained in terms of the loss in internal transmission.

The intensity ratio for RR is apparently different from that for U: i.e., a relative increase in I_i and a relative decrease in I_{ii} compared with those of U. This result implies that the scintillation mechanism of the V_k diffusion process (ii) is affected in the irradiated crystal RR.

3.5. Recovery

The variation of light outputs was measured during 160 days after the irradiation. In order to evaluate the systematic stability of the system, the light output of unirradiated CsI(Tl) crystal was also measured as the reference. Fig. 5 shows the relative light output as a function of time passed after irradiation for R and RR. The two crystals showed a relatively fast recovery in the light output after irradiation, followed by a gradual recovery. The variation of the light outputs $L(t)$ was fitted to the following equation:

$$L(t) = C_0 + C_1(1 - \exp(-t/T_1)) + C_2(1 - \exp(-t/T_2)), \quad (5)$$

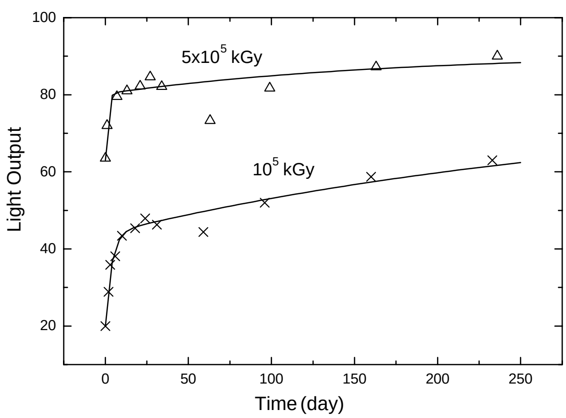

Fig. 5. Recovery in light output of irradiated samples with 10^5 and 5×10^5 Gy. The lines are fitted to Eq. (5), in the text. The light output for the non-irradiated sample is 100.

Table 2
Recovery in light output of CsI(Tl) crystals, for doses of 10^5 Gy (R) and 5×10^5 Gy (RR)

Sample	C_0	C_1	C_2	T_1 (day)	T_2 (day)
R	63	17	10	1.2	321
RR	20	24	34	3.7	168

where C_0 , C_1 and C_2 are constants. The fitted values are summarized in Table 2. Recovery in the light output for 4 days was about 20% of that for U. Permanent losses in the light output, which were given by $100 - [C_0 + C_1 + C_2]$, were expected to be about 10% and 22% of those of U, for the crystals R and RR, respectively.

4. Conclusion

The higher decrease in the light output in the crystal coated with "white tape" is due to the transmission loss of the multiple reflection providing from the white cover. The scintillation mechanism of the V_k diffusion process is affected in the crystal irradiated at 5×10^5 Gy, while for crystal irradiated below 1×10^5 Gy the light output loss is attributed predominantly to internal transmission loss, not to scintillation mechanism.

References

[1] R.Y. Zhu, Nucl. Instr. and Meth. A 413 (1998) 297.
[2] M.A.H. Chowdhury, et al., Nucl. Instr. and Meth. A 432 (1999) 147–156.
[3] D.G. Hitliin, G. Eigen, in: F. de Notaristefani, P. Lecoq, M. Schneegans (Eds.), Heavy Scintillators for Scientific and Industrial Applications, Proceedings of the Crystal 2000 International Workshop, Editions Frontières, Paris, 1993, pp. 467–478.
[4] C.L. Woody, et al., IEEE Trans. Nucl. Sci. NS-39 (1992) 524.
[5] S. Kubota, F. Shiraishi, Y. Takami, J. Phys. Soc. Jpn. 68 (1999) 298.
[6] M. Kobayashi, S. Sakuragi, Nucl. Instr. and Meth. A 254 (1987) 275.
[7] G.J. Bobbink, et al., Nucl. Instr. and Meth. A 227 (1984) 470.
[8] M.M. Hamada, et al., IEEE Trans. Nucl. Sci. NS-48 (2001) 1148.
[9] S. Kubota, F. Shiraishi, Y. Takami, J. Phys. Soc. Jpn. 68 (1999) 291.