Note

Peak 4 Instability in LiF:Mg,Ti (TLD-600/700) : Comparison for γ -Ray and Fe-Ion Irradiation[†]

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Instability of peak 4 in LiF:Mg,Ti (TLD-600/700) was experimentally examined by comparing the glow curves obtained for ^{137}Cs γ -rays and Fe-ion beam. After post-annealing at $100^\circ\text{C} \times 30$ min, thermoluminescence (TL) intensity around peak 4 notably decreased only for γ -ray irradiation. In Fe-ion irradiated samples, post-annealing effects were scarcely observed in the TL intensities in the peak-4 region. These results suggest that the peak-4 luminescent centers are unstable and more vulnerable than other peaks to highly-dense ionization.

Key Words: LiF:Mg,Ti, TLD, glow curve, post annealing, heavy ion,
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1. Introduction

It is well known that TLDs of LiF:Mg,Ti (TLD-100/600/700) exposed to low-energy neutrons or heavy charged particles (HCPs) show considerable differences of their glow-curve shapes. Using this feature, many attempts for discrimination of different-quality radiations have been made in neutron- and HCP-mixed fields^{1)–9)}. In many cases of previous attempts, the ratio of peak 7 (around 250°C) to peak 5 (around 200°C) has been used because of the contrasting characteristics in their LET dependencies and easiness in two-peak separation. However, the TL intensity of peak 7 is too weak to measure γ -rays below ~ 2 mGy¹⁰⁾. Dependence of peak-7 supralinearity

on γ -ray energy⁴⁾ is also disadvantageous. For these reasons, Yossian and Horowitz⁹⁾ proposed utilizing peak 4 (around 170°C) in place of peak 7 for separately evaluating γ -ray dose in neutron-mixed fields. This technique is based on the fact that peak-4 intensity relative to peak 5 in $^6\text{LiF:Mg,Ti}$ (TLD-600) is smaller for neutrons than that for ^{60}Co γ -rays. Such a glow-curve change for different-quality radiations (neutrons and γ -rays) is attributable to differences of secondary charged particles, i.e. Compton electrons induced by γ -rays, and alpha particles ($E=2.05$ MeV) produced by low-energy neutrons through the reaction with $^6\text{Li}^{11)}$. Because the alpha particles have high LET values ($\text{LET}_\infty = 160 - 230 \text{ keV } \mu\text{m}^{-1}$ in water)¹²⁾, the luminescent centers of peak 4 are assumed to be vulnerable to dense ionization induced by HCPs. If this assumption was correct, a reduction in peak 4 intensity should be observed for irradiation by heavy ions with a comparable LET value.

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2. Materials and Methods

The dosimeters employed herein are polycrystalline chips of $^6\text{LiF:Mg,Ti}$ (HARSHAW TLD-600) and $^7\text{LiF:Mg,Ti}$ (HARSHAW TLD-700) with dimensions of $3.2 \times 3.2 \times 0.9 \text{ mm}^3$. Following a previous experiment on gamma-neutron dosimetry⁹⁾, the TLD chips were annealed at 400 °C for one hour and followed by a fast quench in air to room temperature at a cooling rate of about $50^\circ\text{C min}^{-1}$. In general, TL intensity of LiF:Mg,Ti increases with increasing cooling rate¹³⁾.

Two chips of each TLD, four chips in total, were packed in a rectangular case of tissue-equivalent resin (Toughwater phantom, Kyotokagaku) with dimensions of $80 \times 80 \times 6 \text{ mm}^3$ including a 2 mm-thick cover. One dosimeter package was exposed to one of three dose levels : 10, 50, and 100 mGy as water absorbed dose of Fe-ion beam or ^{137}Cs γ -rays in National Institute of Radiological Sciences (NIRS). The Fe-beam irradiation was carried out in the Biology room of Heavy Ion Medical Accelerator in Chiba (NIRS-HIMAC). The absorbed doses were measured with an air-filled ion chamber calibrated to standard radiation sources. The beam uniformity in the exposed area ($100 \times 100 \text{ mm}^2$) was confirmed using plastic nuclear track detectors (CR-39) and a beam-profile monitor installed in the beam line. The energy of Fe ($Z=26$, $A=56$) at the beam exit was 500 MeV per nucleon and the velocity relative to light (β) was 0.75. The unrestricted LET in water ($\text{LET}_\infty \cdot \text{H}_2\text{O}$; abbreviated to LET) at the position of a target was calculated to be 197

$\text{keV } \mu\text{m}^{-1}$. This value is similar to the LET level ($160 - 230 \text{ keV } \mu\text{m}^{-1}$) of alpha particles produced through the reaction of $^6\text{Li}(n, \alpha)^3\text{H}$. Change of LET inside a chip (0.9 mm thick) was estimated to be less than 2 %.

Next day after irradiation, half of the samples were post-annealed at 100 °C for 30 min as shown in Table 1. The irradiated samples are grouped into (1) without post-annealing and (2) with post-annealing. The exposed samples were read two days after irradiation with an automatic TLD reader (HARSHAW 5500) by heating to 360 °C at a heating rate of 10°C s^{-1} . The TL intensity was automatically recorded as a function of temperature on to a personal computer. Since TL-peak positions change by heating rates^{14),15)}, we shifted the reader-indicated temperatures to give the dosimetric main peak (peak 5) at 200 °C for the TLD-600 chips of group-(1) exposed to γ -rays; by means of this adjustment, we can compare the results in the present study with those of previous studies.

3. Results and Discussion

The glow curves of TLD-600 and those of TLD-700 exposed to 50 mGy(H_2O) of γ -rays and Fe-ion beam are shown in Fig. 1 and Fig. 2, respectively; Figs. 1a and 2a are obtained for γ -rays, and Figs. 1b and 2b for Fe-ions. The shapes of dose-normalized glow-curves were almost the same among the different dose levels (10, 50, and 100 mGy) for each group. In both figures, the scales of vertical axis for Fe-ions (right figures) are half of those for γ -rays (left figures); total TL

Table 1 Conditions for annealing and cooling of the dosimeters (TLD-600/700)

Group	Pre-annealing	Post-annealing	Cooling
(1)	400 °C $\times$ 1 h	—	Naturally in air at
(2)	400 °C $\times$ 1 h	100 °C $\times$ 30 min	about $50^\circ\text{C min}^{-1}$

Fig. 1 Glow curves of $^6\text{LiF:Mg,Ti}$ (TLD-600) exposed to 50 mGy(H_2O) of ^{137}Cs γ -rays (a) and Fe-ion beam (b).

The data are grouped into (1) without post annealing and (2) with post annealing at $100^{\circ}\text{C} \times 30$ min.

Note that the scale of vertical-axis in (b) is half of that in (a).

Fig. 2 Glow curves of $^7\text{LiF:Mg,Ti}$ (TLD-700) exposed to 50 mGy(H_2O) of ^{137}Cs γ -rays (a) and Fe-ion beam (b).

The data are grouped into (1) without post annealing and (2) with post annealing at $100^{\circ}\text{C} \times 30$ min.

Note that the scale of vertical-axis in (b) is half of that in (a).

intensities for the Fe-irradiated samples were about 40% of those achieved with the same-dose γ -rays. Such a reduction of TL-efficiency for high-LET particles is probably caused by saturation of trapping centers, as seen for high-dose γ -ray exposure^{(16)–(18)}. An analytic calculation using a track-structure model⁽¹⁹⁾ showed that the energy deposition in nanometer scale adjacent to a path of Fe-ion can be greater than 100 kGy(H_2O). Under a highly-dense ionization, probability of

electron-hole trapping is expected to be reduced to a large extent. These phenomena have been successfully explained *a priori* by microdosimetric approaches based on the target-hit theory^{(20)–(22)}.

After being post-annealed at $100^{\circ}\text{C} \times 30$ min, main-peak (Peak 5) positions shifted to higher temperature for both γ -ray and Fe-ion irradiation; this shifting appeared more clearly in γ -ray irradiated samples than in Fe-ion irradiated ones. Although the glow-curve changes after post-

annealing are attributable to Peak-4 instability, it is not easy to extract only Peak 4 from the overlapped peaks (Peaks 3 – 5). In a previous study using a technique of computerized glow-curve deconvolution (CGCD), several unique peaks were observed for neutron-irradiated TLD-600⁹⁾. Thus, difference of Peak-4 intensity between γ -ray and Fe-ion irradiated samples was discussed using the TL-intensity ratio of the group-(2) (with post-annealing) to the group-(1) (without post-annealing). Plots of the calculated TL ratios as a function of temperature from 100 °C to 200 °C are shown in Fig. 3a for TLD-600 and in Fig. 3b for TLD-700. The data obtained for the same temperature from the samples exposed to different dose levels (10, 50, and 100 mGy) are averaged in these plots. For γ -irradiation, notable TL reductions around Peak 4 were observed for both dose-meters after post-annealing. Whereas, the effects of post-annealing are hardly seen for Fe-ion irradiated samples; the ratios between groups (2) and (1) were almost unity around Peak 4. This fact suggests that the Peak 4 induced by a Fe-ion is much smaller than the γ -ray induced Peak 4; in other words, Peak-4 luminescent centers have already been depleted in Fe-ion irradiation. These findings agree with the results of a previous CGCD analysis⁹⁾, in which the deconvoluted Peak-4 intensity relative to Peak 5 was much smaller for neutrons (i.e. alpha-particles) than that for γ -rays. The flat shapes of the glow curves of Fe-ion irradiated samples (Fig. 3) also support the results of the CGCD analysis⁹⁾, in which neutron (alpha-particle) irradiated samples showed several unique overlapping peaks (3a, 3b, 4a, etc.).

From the results in the present study, the Peak-4 luminescent centers are considered to be unstable and depleted more easily than other peaks by dense energy deposition brought by heating (post-

Fig. 3 The TL-intensity ratio of group-(2) to group-(1) (See Table 1) as a function of temperature for TLD-600 (a) and TLD-700 (b); the data obtained for different dose levels (10, 50, and 100 mGy) are combined. Notable reductions of TL-intensity around Peak 4 are seen only for γ -ray irradiated samples.

annealing) or highly localized ionization. This vulnerability of Peak 4 must be responsible for the observed instabilities of Peak 4 in size, position, and shape during long-term storage^{23),24)}.

The vulnerability of Peak 4 might be explained based on instability of impurity-vacancy ($\text{Mg-Li}_{\text{vac}}$) dipoles, because annealing at 400 °C $\times$ 1 h results in most of Mg being in the form of $\text{Mg-Li}_{\text{vac}}$ dipoles and subsequent annealing establishes a new equilibrium of Mg in the form of trimers²⁵⁾. Thus, Peak-4 luminescent centers are

possibly present with Mg-Li_{vac} dipoles. The contrasting feature of Peak 5 which increased after post-annealing (Figs. 1 and 2) suggests that the Peak-5 luminescent centers are present with Mg trimers. This subject is now examined on a theoretical basis. In conclusion, it is strongly suggested that evaluation of radiation quality using Peak 4 is susceptible to its vulnerability and requires a rigorous handling of the dosimeters.

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