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LET dependence of TSL properties in Ce:LiCaAlF6 crystals

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ABSTRACT

We have investigated thermally-stimulated luminescence (TSL) properties of Ce-doped $LiCaAlF_6$ crystals irradiated with ionizing radiations of different linear energy transfers (LET) – X-rays and He ion beams. It was found that the TSL glow peaks mainly appeared around 120 and 250–400 °C, and the ratio of the former peak intensity to the latter was found to decrease with increasing the LET. Furthermore, together with the TSL properties measured with X-rays, diffusion length of electron-hole pairs was estimated to lie in a range of 30–120 nm depending on the LET and concentration of Ce.

1. Introduction

For the past decades, great developments have been achieved in thermally-stimulated luminescence (TSL) experiments with the growth of the important applications of radiation dosimetry such as personal dose monitoring [1], medical imaging [2] and radiation therapy [3]. Under irradiation, TSL material stores and accumulates the incident excitation energy as trapped electrons and holes and then release the absorbed energy as light by thermal stimulations. The irradiation can be any form as long as it ionizes the host such as UV lights, X-rays, charged particles and neutrons. Other than the application point of view, TSL is a useful tool to characterize material properties such as charge trapping levels, defects and impurities in solids.

Particularly, demands of TSL dosimeters have been rapidly increasing around the field of high-energy particle accelerators in clinical and industrial applications, and we are encouraged to make more efforts on the investigation of TSL behavior for heavy charged particle irradiation. Since heavy charged particles show quite high linear energy transfers (LETs), they generate tracks of local ionization events [4,5]; and LET-dependent response of TSL properties were previously reported [6]. In this point of view, the phenomenon induced by high LET is largely different from the case of X- and γ-ray irradiations which cause electronic excitation at low LET. Such high-LET irradiation involves large probability of excited state interactions; and this phenomenon must be discussed by taking consideration of the ionization density over the three dimensional scale, whereas LET is however a one-dimensional quantity of ionization density. The distribution of ionization density over the vicinity volume of ion track varies with LET since the diffusion length of electron-hole pairs along the direction perpendicular to the ion trajectory depends on the LET.

The aim of this study is to investigate LET dependence of TSL properties and analyze the ionization density in a three dimensional scale and the diffusion lengths of high LET heavy charged particles. TSL glow curves, i.e. the TSL intensity as a function of heating temperature, are generally comprised of multiple glow peaks. In some materials, relative intensities of glow peaks appear to be different with different X-ray exposure doses. Glow peaks at different temperatures are generally representation of electrons (and/or holes) released from trapping centers of different depths or groups, and the total number of charges which can be captured by each group of trapping centers is supposed to vary. Therefore, the appearance of TSL glow peaks with different relative intensities indicates that one or more of trapping centers have been filled up, and the extent of saturation (or relative glow peak intensities) should depend on the density of ionization. In this case, the relative intensities of TSL glow peaks can be used as a measure of ionization density by taking account of the three dimensional distribution surrounding the ion tracks. A similar effect is expected with high LET irradiation. The higher the LET, the more the electrons

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and holes generated, so high LET irradiation is equivalent to large dose irradiation, and the ionization density should be estimated in the same manner. In addition, we can also estimate a diffusion length associated with LET and ionization density in a three dimensional scale.

The material of interest in this study is Ce:LiCaAlF$_6$ crystals. It was first developed for optical lens applications due to the wide band gap energy, and then applied for solid state UV laser when doped with Ce^{3+} [7–9]. In addition to these studies on optical devices, Ce-doped LiCaAlF$_6$ was found to show high TSL properties for the X-ray irradiation [10]. Following these researches, we have developed and investigated ^{6}Li enriched Ce-doped LiCaAlF$_6$ for the neutron scintillation detectors [11–13] since the shortage of ^{3}He gas commonly used for neutron detections became a serious problem worldwide [14,15] and alternative thermal neuron detectors are urgently required. During these research and development, the Ce:LiCaAlF$_6$ scintillator was proven to show discrimination function of neutron and γ-ray induced events by using the difference of the pulse shape depending on LET [16] although conventional scintillator-based neutron detectors were not able to distinguish between the events of neutrons and γ-rays. In ^{6}Li enriched scintillators, ^{6}Li(n,α)^{3}H reaction occurs so the scintillator is excited by charged particles. Therefore, it is very important to understand the physical phenomena involved under high LET conditions. Up to now, scintillation properties particularly luminescence dynamics under high LET have been studied [17] while steady state properties such as TSL have been investigated after X-ray (low LET) irradiation [18]. In order to understand responses of LiCaAlF$_6$ against ionizing radiation, steady state analysis under high LET is required. Further, together with the knowledge from the earlier studies, we attempted to derive the diffusion lengths of the electron-hole pairs by heavy He charged particle beam with different LETs.

2. Experimental

The Ce-doped LiCaAlF$_6$ crystal samples used in this study was grown by the Czochralski method by Tokuyama Corp. The concentration of Ce added was 1 and 4 mol.% of Ca. Fig. 1 shows the samples used in this research. The sample sizes of 1 and 4% Ce-doped crystals were about 10 × 2 × 1 mm^3 and 10 × 10 × 1 mm^3, respectively. Hereafter, the samples are referred by their Ce concentrations.

The samples were irradiated by He ion beams and X-rays. The irradiation of He beam was performed at the Heavy Ion Medical Accelerator in Chiba (HIMAC) synchrotron facility at the National Institute of Radiological Sciences (NIRS), Japan. The radiation doses for all the irradiations were fixed to 10 Gy of water equivalent quantity. The LET was controlled by changing the He energy by varying the thickness of binary filters (BF) and was calculated using the SRIM code [20]. In the calculation, deceleration of particles only in the BFs and the sample was considered and that in the air was neglected. In addition, fragmentation in the BFs is also ignored. The LET values dealt in this paper are averaged values over the entire depth of the samples. For X-ray irradiation, a conventional X-ray tube installed in an X-ray diffractometer (MiniFlex600, Rigaku) was used. The X-ray tube was equipped with a Cu anode target and Be window. The tube voltage and current were set to 40 kV and 1.5 mA, which yield the dose rate of 35 Gy/min in air.

TSL glow curves were measured using our original setup. An irradiated sample was placed on a ceramic heater, and the sample was heated at a rate of 1 °C/s. The temperature of the heater was controlled by a Sakaguchi controller system (SCR-SHQ-A). During the heating, the TSL signal from the sample was collected by an optical fiber and guided to a CCD-based spectrometer (QEPro, Ocean Optics) to measure the spectrum as well as intensity as a function of temperature.

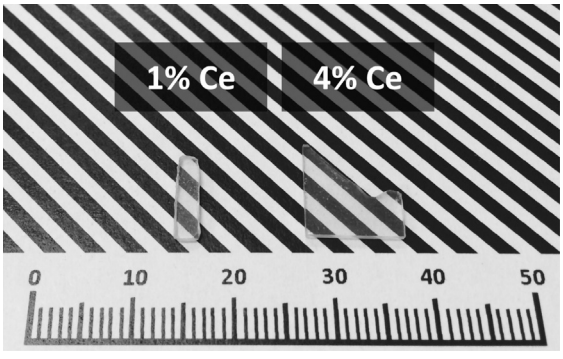

Fig. 1. Samples of 1% and 4% Ce:LiCaAlF$_6$ crystals.

3. Results and discussion

Fig. 2 represents the TSL spectra of 1% and 4% Ce-doped LiCaAlF$_6$ samples measured at 300 °C after irradiations by X-rays and He ion beam. The irradiation dose was fixed to 10 Gy. The emission is mainly due the Ce ion activated as emission center which appears to be a single band emission around 300 nm. The emission spectrum seems to be independent on the radiation types but slightly on the concentration of Ce ion. These emission spectral features are consistent with those of previous report with X-ray irradiations [18].

Figs. 3 and 4 show TSL glow curves of the Ce:LiCaAlF$_6$ samples measured after X-ray irradiation with various doses. In the 1% Ce-doped sample, TSL glow peaks are observed around 120 and 330 °C whereas the 4% Ce-doped sample shows the glow peaks around 120, 270 and 350 °C. These peak positions are consistent with reported values in the past studies [18], and the splitting of peaks around 200–400 °C would be due to the charge imbalance between Ce^{3+} and Ca^{2+}. For both of the samples, the intensity ratio of the high-temperature peak, I_{high}, to that of the low-temperature peak, I_{low}, decreases with increasing the radiation dose. In the evaluations, I_{high} and I_{low} are defined as the peak TSL intensities of the higher and lower peaks, respectively. As demonstrated in the insets of Figs. 3 and 4, the relative intensity I_{high}/I_{low} decreases monotonically with increasing the radiation dose in the semi-logarithmic representations.

TSL glow curves of the 1% and 4% Ce-doped samples measured after irradiation (10 Gy) with He ion beam of various LETs are shown in Figs. 5 and 6, respectively. For both of the samples, the

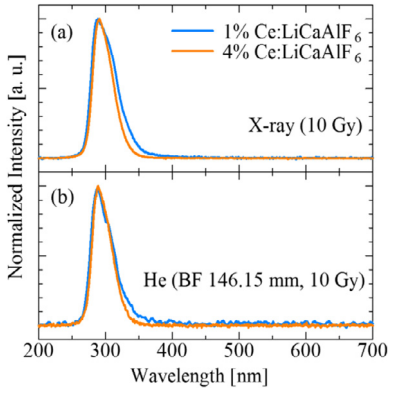

Fig. 2. TSL spectra of the 1% and 4% Ce:LiCaAlF$_6$ crystals (a) after 10 Gy X-ray exposure and (b) after He ions exposure.

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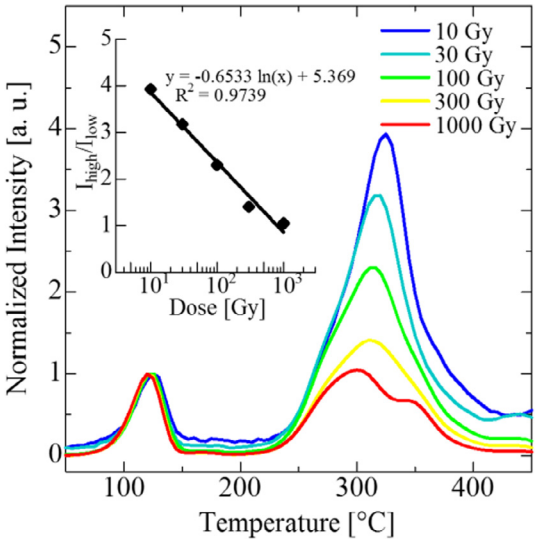

Fig. 3. TSL glow curves of the 1% Ce:LiCaAlF$_6$ crystal measured after X-ray irradiation of various radiation doses. The inset shows the relationship between the relative intensity of glow peaks I_{high}/I_{low} and irradiation dose.

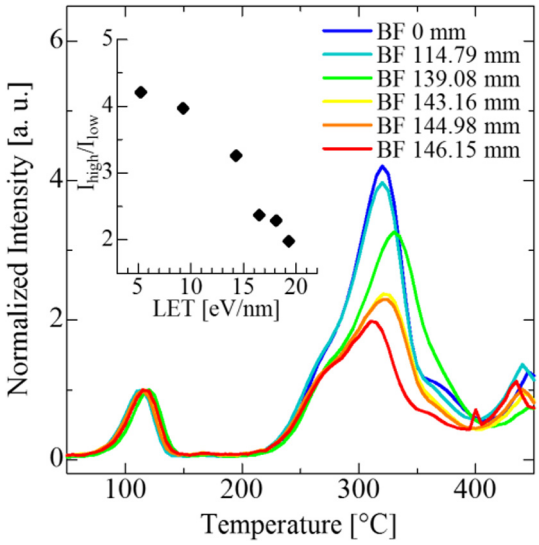

Fig. 5. TSL glow curves of the 1% Ce:LiCaAlF$_6$ crystal measured after irradiation of He ion beam at 10 Gy with varying LETs. The inset shows the relative intensities of glow peaks I_{high}/I_{low} as a function of LET.

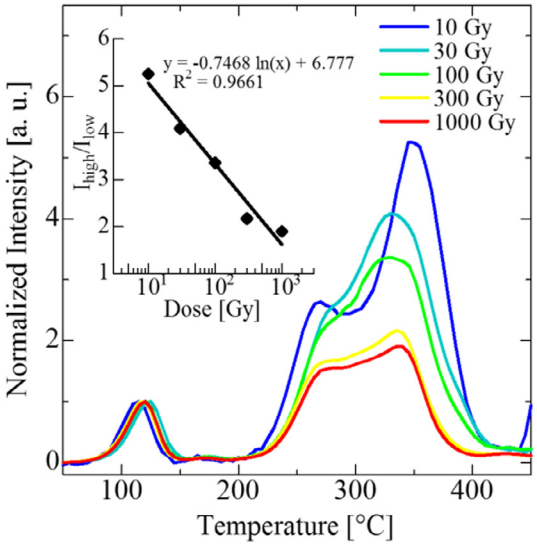

Fig. 4. TSL glow curves of the 4% Ce:LiCaAlF$_6$ crystal measured after X-ray irradiation of various radiation doses. The inset shows the relationship between the relative intensities of glow peaks I_{high}/I_{low} and irradiation dose.

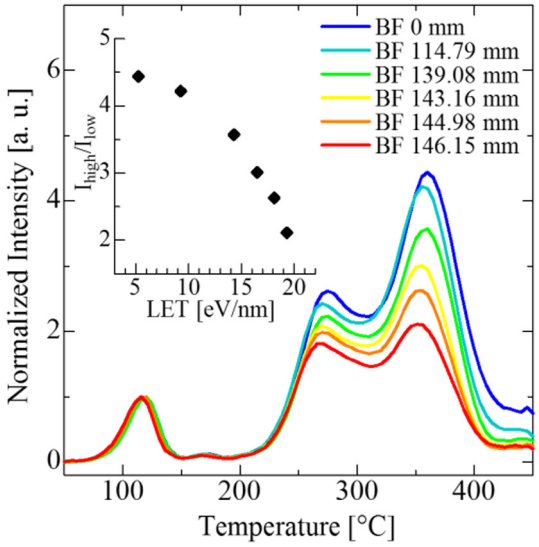

Fig. 6. TSL glow curves of the 4% Ce:LiCaAlF$_6$ crystal measured after irradiation of He ion beam at 10 Gy with varying LETs. The inset shows the relative intensities of glow peaks I_{high}/I_{low} as a function of LET.

relative TSL intensity of around 200–400 °C decreases in association with increasing LET as observed for high dose X-ray irradiation. However, as illustrated in the insets of Figs. 5 and 6, unlike X-ray irradiation, the decrease in I_{high}/I_{low} with increasing LET is not monotonic.

With the TSL data presented above, we have estimated the diffusion lengths of electron-hole pairs as follows. Hereafter, irradiated X-ray dose and LETs are referred as A and B, respectively. Then, they can be expressed as A [Gy] = A [J/kg] = $A \times 2988$ [J/m^3] since the density of LiCaAlF$_6$ is 2.988 g/cm^3. On the other hand, B [eV/nm] can be expressed as $B \times 1.602 \times 10^{-19} \times 10^9$ [J/m]. Assuming that the energy is deposited uniformly in the cylindrical volume of ion track whose diameter corresponds to the diffusion length of electron-hole pairs, the ionization density can be expressed as $B \times 1.602 \times 10^{-10}/\pi r^2$ [J/m^3]. Here, r is the diffusion length of electron-hole pairs. Assuming that the excitation densities are the same for the same value of I_{high}/I_{low}, the calculated diffusion lengths of 1% and 4% Ce-doped LiCaAlF$_6$ crystals are respectively $r_{1\%} \simeq 40$–120 nm and $r_{4\%} \simeq 30$–70 nm as listed in Table 1. These values are significantly smaller than that estimated earlier for GaAs [19]. This difference is reasonable because electron and hole mobilities in LiCaAlF$_6$ are considered to be much lower than those in GaAs. The estimated diffusion lengths are larger at lower LETs. This result may be correlated with the velocity of the He ions: the initial diameter of the energy deposited region is known to be larger at higher velocity of the impinging ions. In addition, suggested by the results, the diffusion lengths are significantly smaller in the sample with higher concentration of Ce. This can be attributed to a difference in concentrations of trapping sites: probability of charge trapping is proportional to concentration of trapping centers. The trap concentration is expected to be higher for higher Ce concentration since a larger number of defects

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Table 1
Diffusion lengths of electron-hole pairs calculated for He ion irradiation with different LETs.

LET [eV/nm]	$r_{1\%}$ [nm]	$r_{4\%}$ [nm]
5.25	120	62
9.26	140	71
14.3	99	58
16.5	53	43
18.1	53	34
19.3	43	25

are to be created due to the charge imbalance between Ce^{3+} and Ca^{2+}. These results show approximately $r_{1\%}/r_{4\%} \simeq \sqrt[3]{(4\%/1\%)}$ and are quite reasonable.

4. Conclusions

We investigated the TSL properties of 1% and 4% Ce-doped $LiCaAlF_6$ crystals measured after irradiations of X-rays of different and large doses and He ion beams of different LETs. In both of the samples, the relative TSL intensity I_{high}/I_{low} decreases with increasing LET of ion beam irradiation. Calculated from the corresponding I_{high}/I_{low} values measured with X-ray irradiations, diffusion lengths of electron-hole pairs were estimated to be about 30–120 nm, which depends on the LET as well as the concentration of Ce.

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